Effect of Carbon Content on Bainite Transformation Start Temperature in Middle–High Carbon Fe–9Ni–C Alloys

Hiroyuki KAWATA, Toshiyuki MANABE, Kazuki FUJIWARA and Manabu TAKAHASHI

1) Steel Research Laboratories, Nippon Steel & Sumitomo Metal Corporation, 20-1 Shintomi, Futtsu, Chiba, 293-8511 Japan.

Corresponding author: E-mail: kawata.z84.hiroyuki@jp.nssmc.com
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Bainite in steel is an industrially useful structure. However, the controlling factor of its transformation start point is not clearly known. In this study, we measured the bainite transformation start temperature (Bs) in Fe–9Ni–C alloys containing 0.3–0.9 mass%C via microstructure observation of the specimens held isothermally between 600 K and 798 K. Bs existed between 758 K and 773 K in all alloys used, and was independent of carbon content. Especially, Bs was higher than To, at which fcc and bcc of the same composition have the same free energy, at more than 0.3 mass%C. This result was completely different from that of our previous study on low carbon Fe–9Ni–C alloys, in which Bs decreased with the increase in carbon and kept the certain driving force of partitionless transformation from fcc to bcc. Bs in middle–high carbon alloys corresponded to the temperature of the intersection point between To, at which the driving force is 400 J/mol, and the γ+θ phase boundary. This suggests that the nucleation and growth of bainitic ferrite in austenite containing solute carbon higher than To is promoted by the precipitation of cementite in austenite.

KEY WORDS: bainite; martensite; cementite; phase transformation; To.

1. Introduction

Bainite in steel is a very useful structure and has high strength with good formability and toughness. Many types of high strength steels contain bainite with various carbon contents. For example, the high toughness steels contain bainite with low carbon content. On the contrary, in the low-alloyed TRIP steels, bainite transformed from residual austenite that contains a high carbon content of over 1 mass%. It is important to understand the bainite transformation behavior over a wide carbon range. Especially, it is valuable to know the bainite transformation start temperature (Bs) with various carbon contents for understanding about bainite transformation mechanism.

There are many studies that deal with the behavior of Bs with carbon content. Bhadeshia proposed that Bs is parallel and below To, at which fcc and bcc systems of the same composition have the same free energy. In a previous study, we confirmed this suggestion in Fe–9 mass%Ni alloys with 0.003–0.10 mass% carbon. This phenomenon suggests that Bs depends on the driving force of partitionless transformation from fcc to bcc, which corresponds to the supercooling from To.

However, in middle–high carbon content regions, Bs is independent of To. It was reported that Bs can be over To in high carbon content regions, in Fe–C, Fe–2Mn–C, and Fe–Cr–C alloys; in middle carbon content regions, Tsuzaki et al. showed that Bs does not change with increasing carbon content in Fe–9Ni–C alloys.

This study clarifies the mechanism of bainite transformation. To this end, we evaluate the effect of carbon content on Bs in Fe–9Ni alloys, in which nickel does not exhibit a solute drag like effect. The effect of carbon content in low carbon region has already been reported. In this paper, we focus on middle-high carbon alloys. We defined that bainite, which transforms just below Bs, contains a group of lath-shaped ferrites with/without cementite. This definition confirms to the upper bainite categorized by Ohmori et al.
affects the transformation behavior.\textsuperscript{4} Figure 1 shows the histogram of nickel distribution measured by electron probe microanalysis (EPMA) throughout the thickness of these sheets. Local nickel content in these sheets differs from 7.6 to 10.0 mass\%. Nickel depresses the bainite transformation.\textsuperscript{12} The bainite transformation must start in the local nickel poor region. Therefore, the effective nickel content (\(\text{WNi,E}\)) that dominates the bainite transformation start is the local nickel content in the nickel poor region.

Table 2 shows the \(\text{WNi,E}\) in these alloys sheets, and \(T_0\) temperatures that were calculated via Thermo-Calc using the TCFE8 material database with \(\text{WNi,E}\). We adopted the nickel content at 1\% from the bottom of the distribution for \(\text{WNi,E}\) because the dilatation technique used in this study can detect 1\% progress of the transformation from fcc to bcc empirically.

These sheets were cut to small specimens with dimensions of 10 mm \(\times\) 80 mm. These specimens were heated via electrical heating and cooled by \(\text{N}_2\) gas. The transformation behavior during the heat treatment was evaluated based on a dilatation curve obtained using a laser displacement meter. The specimens were austenitized at 1473 K for 60 s. After austenitization, the specimens were cooled to several temperatures at 30 K/s and were held isothermally. The holding time was changed according to the transformation behavior. After holding, the specimens were cooled at 30 K/s.

The microstructures etched with nital were observed using an optical microscope (OM). In order to avoid the surface effect, the observation field was limited to the inside of the steel plate, and observation was not carried out in the range of 0.13 mm depth from the surface. For all specimens, we observed over 20 prior austenite grains in the observation field. Especially, in the specimens held isothermally around Bs for a short time, which contained a small amount of transformed structure, we observed throughout the observation field.

3. Results

3.1. Time–temperature–transformation (TTT) Diagrams

Figure 2 shows the time–temperature–transformation (TTT) diagrams of alloy F, G, H, and I. These diagrams show the transformation start time measured by the dilatation curve at each temperature for each alloy. In alloys F and G, the transformation start time makes C–curves between 600 K and 773 K, and the transformation start over 773 K becomes faster with the increase in temperature. Therefore, in these alloys, there is a clear bay around 773 K. Also, in alloys H and I, the transformation start time makes C–curve between 600 K and 773 K. However, in these alloys, the dilatation curves during isothermal holding for 10 000 s at more than 773 K did not show the expansion which correspond to the transformation start. In them, the transformation start time showed terraces around 773 K and not bays.

Figure 3 shows the schematic TTT diagram which consists of two C-curves corresponding to the diffusion transformation (ferrite and pearlite) and the bainite transformation, respectively.\textsuperscript{15} In generally, the terraces in Fig. 2 correspond to Bs.\textsuperscript{3,4,6,13} These TTT diagrams indicate that Bs in low carbon alloys which

\begin{table}[h]
\centering
\caption{Chemical compositions and \(T_0\) temperatures of the alloys used in the current and previous\textsuperscript{4} studies.}
\begin{tabular}{lcccccccc}
\hline
Alloy & Chemical compositions/mass\% & \hline
 & C & Si & Mn & P & S & Ni & B & N \\
A & 0.0031 & <0.01 & 0.02 & 0.0005 & 0.001 & 9.06 & 0.0023 & 0.0013 \\
B & boron added & 0.052 & <0.01 & 0.02 & 0.0005 & 0.001 & 9.06 & 0.0024 & 0.0014 \\
C & & 0.094 & <0.01 & 0.02 & 0.0005 & 0.001 & 9.09 & 0.0024 & 0.0017 \\
D & & 0.050 & <0.01 & 0.01 & 0.0005 & 0.001 & 9.03 & <0.0003 & 0.0017 \\
E & & 0.099 & <0.01 & 0.01 & 0.0005 & 0.001 & 9.06 & <0.0003 & 0.0017 \\
F & boron free & 0.30 & <0.01 & 0.01 & 0.0005 & 0.001 & 9.03 & <0.0003 & 0.0020 \\
G & & 0.50 & <0.01 & 0.01 & 0.0005 & 0.001 & 9.01 & <0.0003 & 0.0017 \\
H & & 0.69 & <0.01 & 0.01 & 0.0006 & 0.001 & 8.95 & <0.0003 & 0.0016 \\
I & & 0.89 & <0.01 & 0.01 & 0.0006 & 0.001 & 9.00 & <0.0003 & 0.0015 \\
\hline
\end{tabular}
\end{table}

\begin{table}[h]
\centering
\caption{Effective nickel content and \(T_0\) temperature for the bainite and martensite transformation start.}
\begin{tabular}{lccccc}
\hline
Alloy & F & G & H & I \\
\hline
WN_{\text{E}}/\text{mass}\% & 7.67 & 7.60 & 7.75 & 7.59 \\
\(T_0/\text{K}\) & 799 & 729 & 660 & 593 \\
\hline
\end{tabular}
\end{table}
decrease with increasing carbon content. 4)

Figure 4 shows optical micrographs of alloy G and I specimens held at 723 K for the time to finish the transformation. 723 K is the peak of C-curves in all TTT diagrams evaluated (Fig. 2). In alloy G (Fig. 4(a)), there are pearlite (P), degenerated pearlite (DP), and lath-shaped structures. The lath-shaped structure contains cementite, which looks elongated along with the lath boundary. We concluded that this structure is upper bainite (UB), categorized to B-II11) from its morphological features. In alloy I (Fig. 4(b)), although the transformation temperature, 723 K, is much higher than \( T_0 \), 593 K, the microstructure contains P and UB (B-II), similarly.

On the contrary, the microstructures that were generated around the terrace are different from Fig. 4. Figure 5 shows optical microscope images of the specimens held at 773 K for 10,000 s. (a) Alloy F and (b) Alloy G. α: allotriomorphic ferrite, P: Pearlite, DP: Degenerated Pearlite.
shows the microstructures in alloys F and G held at 773 K. This temperature corresponds to the terrace in Fig. 2. These microstructures contain allotriomorphic ferrite and DP, in which the cementite particles make a wavy dot line. However, in these microstructures, there is no UB that contains lath-shaped ferrite with elongated cementite. It suggests that the temperature of terrace is just above or as much as Bs, at which the velocity of bainite transformation is much smaller than that of ferrite and pearlite transformation.

3.2. Microstructure during Early Stage of Transformation

To clarify the morphological features of microstructures explicitly, we observed the specimens, which were held at several temperatures and quenched after the transformation start time. The fraction of transformed volume estimated by

Fig. 6. Optical microscope images of Alloy G samples held isothermally (a) at 798 K for 10 000 s, (b) at 773 K for 2 000 s, (c) at 748 K for 1 000 s, (d) at 733 K for 1 000 s, and (e) at 648 K for 1 200 s. α: allotriomorphic ferrite, P: Pearlite, DP: Degenerated Pearlite, UB: Upper Bainite, α′: lath martensite that transformed during quench after isothermal holding, γ: residual (untransformed) austenite, prior γ g. b.: prior austenite grain boundary.
dilatation curves is less than 5% in them. The transformation mainly started around prior austenite grain boundaries (γ g. b.) in these specimens.

**Figure 6** shows the microstructure during the early stages of transformation in alloy G, which contains 0.5 mass% carbon. In the specimens held above the bay, as shown in Figs. 6(a) and 6(b), the microstructures contain a small amount of allotriomorphic ferrite grains around γ g. b. These ferrite grains are surrounded by lath martensite (α′), which transformed during quenching after isothermal holding, and residual austenite (γ). However, we could not observe UB lath in these specimens.

On the contrary, Figs. 6(c), 6(d), and 6(e) show the microstructures in the early stages of transformation in alloy G held below the terrace in the TTT diagram (Fig. 2). At 748 K (Fig. 6(c)), a lath-shaped ferrite structure exists around γ g. b. with elongated cementite particles. This structure corresponds to B-II type UB. The UB layer can distinguish from (α′ + γ) layer by the etched color, which depends on the solute carbon content. Therefore, we could observe some P and/or DP layers, which consists of allotriomorphic ferrite and cementite lamellar structures. The specimen held at 733 K shows similar microstructure in Fig. 6(d), which contains UB (B-II) and P (P and/or DP) around γ g. b.

Figure 6(e) shows the microstructure in alloy G held at 643 K. In this specimen, we could observe the lath-shaped black layer around γ g. b. with OM, and could not detect B-II type UB. This black layer consists of lath-shaped ferrite and many small cementite particles. This morphological feature completely corresponds to B-III type UB. Figure 7 shows the microstructures during the early stages of transformation in alloy I, which contains 0.9 mass% carbon. In the specimen held above the terrace, as shown in Fig. 7(a), the microstructure contains DP and pro-eutectoid cementite plate (θ). There is no UB layer.

![Fig. 7. Optical microscope images of Alloy I samples held isothermally (a) at 773 K for 10 000 s, (b) at 758 K for 10 000 s, (c) at 743 K for 3 000 s, (d) at 733 K for 2 000 s, and (e) at 723 K for 1 200 s. θ: pro-eutectoid cementite plate and other captions are similar with those in Fig. 5.](image-url)
With the temperature decreasing, UB appeared in alloy I, similarly with alloy G. At 758 K, Fig. 7(b), the microstructure during the early stage of transformation contains B-II type UB with P and/or DP, and θ. At 743 K and 733 K, as shown in Figs. 7(c) and 7(d), respectively, the microstructure contains UB (B-II) and P, and θ disappeared. At 723 K, as shown in Fig. 7(e), UB changed from B-II type to B-III type, similarly with alloy G.

Figure 8 shows the change of constituent in the microstructures, which transformed in early stages, with temperature and carbon content. The open dots correspond to the microstructures containing B-II type UB, and the solid dots correspond to the microstructures containing B-III type UB. Other dots show the microstructures containing no UB region. Between 0.3 and 0.9 mass%C, UB appeared below 758 K. Above 773 K, we could not observe UB in all alloys used in this study. And, the morphological features of UB changed from B-II type to B-III type, similarly with alloy G.

These results (Figs. 5, 6, 7, and 8) indicate that Bₜ exists between 758 K and 773 K, regardless of the carbon content. The broken line in Fig. 8 shows T₀ calculated for Fe–7.6 mass%Ni–C ternary system. 7.6 mass%Ni corresponds to W₅₆ in alloys used (Table 2). In alloy F containing 0.3 mass%C, Bₜ is as much as T₀. Therefore, in other alloys containing 0.5 mass% carbon or more, the Bₜ temperatures are both higher than T₀.

4. Discussion

Figure 9 shows the Bₜ measured in the current and previous studies along with the carbon content. The broken line in Fig. 9 shows T₀ calculated for Fe–7.6 mass%Ni–C ternary system. 7.6 mass%Ni corresponds to W₅₆ in alloys used (Table 2). In alloy F containing 0.3 mass%C, Bₜ is as much as T₀. Therefore, in other alloys containing 0.5 mass% carbon or more, the Bₜ temperatures are both higher than T₀.

These results match well with those reported by Aaronson regarding Bₜ in Fe–C alloys. Ohmori tried to express the behavior of Bₜ in high carbon alloys with the model proposed by Tsuzaki. This model assumed that the nucleation of bainitic ferrite occurs in the carbon-poor regions. In this model, the highest temperature, at which the nucleation is allowed to occur, corresponds to Mₘ in the ultra low carbon alloy containing the same amount of substitution atoms. In low carbon alloys, since Mₘ in the ultra low carbon alloy is lower than T₀', the nuclei can grow displacively. Therefore, Bₜ in low carbon alloys corresponds to Mₘ in the ultra low carbon alloy. According to Tsuzaki’s original model, Bₜ in high-carbon alloys corresponds to T₀' because the nuclei cannot grow above T₀'. Ohmori proposed the expansion of this model in regard with the precipitation of cementite in austenite. Since the precipitation of cementite decreases the carbon content in austenite around itself from over T₀'.

Completely corresponded to the Bₜ model proposed by Bhadeshia.

However, in this study, Bₜ is independent of carbon content between 0.3 and 0.9 mass%C. Alloys F, G, H, and I almost have the same Bₜ. Over 0.3 mass%C, Bₜ is higher than T₀ (bold black line in Fig. 9). It seems that the bainite transformation does not need the driving force of partitionless transformation. The Bₜ in Fe–9Ni–C alloys plotted versus carbon content in Fig. 9 seems to have the inflection point at around 0.2 mass%C.
to below $T'_0$, the nuclei of the bainitic ferrite can grow in the carbon poor region introduced by the precipitation of cementite. Therefore, above $T'_0$, bainite transformation can proceed with cementite precipitation.

In this study, $B_s$ between 0.3 and 0.9 mass%C corresponds to $M_s$ in alloy A containing 0.003 mass%C. It seems that Ohmori’s suggestion is correct. However, as $M_s$ can increase in alloy A by further carbon reduction, there is no necessity that $B_s$ in alloys F, G, H, and I coincide with $M_s$ in alloy A. It is difficult to explain the behavior of $M_s$ in middle–high carbon alloys by Ohmori’s suggestion based on Tsuzaki’s model.

We perceived in Ohmori’s idea that the precipitation of cementite induced bainite transformation in high carbon alloy is useful. In this study, the microstructure during the early stage of transformation just below $B_s$ always contains P and/or DP around UB. Tsuzaki et al. also demonstrated the microstructure containing DP around UB below $B_s$ in Fe–9 mass%Ni-0.28 mass%C alloy. Figure 10 shows $B_s$, $M_s$, $T_0$, $T'_0$, and the $\gamma$($\gamma + \theta$) phase boundary in Fe–9 mass%Ni-C alloys. White and black dots correspond to $B_s$ and $M_s$ measured in the current and previous studies. $T_0$, $T'_0$, and the $\gamma$($\gamma + \theta$) phase boundary are calculated via Thermo-Calc. In the calculation, we used representative $W_{Ni,E}$, 7.6 mass%, for Ni content to regard the effect of Ni segregation. The $\gamma$($\gamma + \theta$) phase boundary was calculated in the ortho equilibrium condition. In this figure, $B_s$ in low carbon alloys exist along with $T'_0$ and that in middle–high carbon alloys correspond to the temperature of the intersection point between $T'_0$ and the $\gamma$($\gamma + \theta$) phase boundary. This intersection point, arrowed in Fig. 9, located on 0.205 mass%C, 755 K. Similarly, $B_s$ above $T'_0$ in Fe–C and Fe–Mn–C alloys are close to the temperatures of their calculated intersection points.

We can explain this agreement between $B_s$ and the intersection point with Bhadeshia’s model and Ohmori’s idea. Figures 11 and 12 show the illustrations that describe the bainite transformation behavior proposed in this study. Figure 11 shows the construction process of UB, and Fig. 12 describes the change of solute carbon content which controls the driving force of bainite transformation. In fully austenitic high carbon alloy held below the $\gamma$($\gamma + \theta$) phase boundary (Figs. 11(0) and 12(A)), the cementite particles precipitate (Fig. 11(1)) and decrease the solute carbon content in austenite around themselves, $X_1$ (Fig. 12(B)). If the holding temperature, $T_t$ is lower than the intersection point, $X$ becomes lower than the carbon content of $T'_0$ as in Fig. 12(C), and the carbon poor region around the cementite gets the driving force of the partition-
less transformation from fcc to bcc, which is enough for the displacive growth of bainitic ferrite. After the nucleation and/or growth of bainitic ferrite (Fig. 11(2)), X increases with the carbon partitioning from bainitic ferrite to austenite26 (Figs. 11(3) and 12(D)). After that, since the increase in carbon content promotes the precipitation of cementite, the cementite grows and/or precipitates around the bainitic ferrite (Fig. 11(4)), and X decrease toward X1 again (Fig. 12(C)). By repeating this process, the bainitic ferrite and the cementite nucleate and grow continuously, and they make B-II type UB, as in Fig. 11(6).

In Figs. 11 and 12, we simply described the proposed relation between the cementite and the bainitic ferrite. The trigger of the bainite transformation is not only the simple cementite particle but also P and DP if they reduce the solute carbon content in the austenite around them. At low temperatures, because of the increasing driving force of the precipitation of cementite in bainitic ferrite, the cementite would nucleate into the bainitic ferrite before carbon partitioning from bainitic ferrite to austenite, and the microstructure would become B-III type UB.

In this model, the bainite transformation rate is limited by the cementite precipitation. This process looks like the so-called “inverse bainite”3,19. And, this process is similar to the transformation behavior of cementite and pearlite from hyper-eutectoid alloy.

We proposed this model on the assumption that the kinetics of cementite nucleation and growth is fast enough. In the current and previous studies, the alloys used do not contain the elements that delay cementite formation. If the cementite precipitation is depressed, the proposed process cannot proceed smoothly, and B3 in high carbon alloys would become lower than the temperature of the intersection point between T0′ and the γ′(γ + θ) phase boundary; particularly, if the cementite precipitation cannot occur, we assume that B3 should correspond to T0′ in all carbon content.

5. Summary
To clarify the effect of carbon content on the B3 of Fe–9Ni–C alloys containing 0.3–0.9 mass% C, we observed the microstructure during the early stage of transformation during isothermal holding. We compared the behavior of B3 with that in the low carbon Fe–9Ni–C alloys measured in our previous study. In this study, bainite refers to a lath-shaped ferrite group with cementite that generates over Ms.

(1) In TTT diagrams, an increase in the carbon content delayed the transformation start. However, regardless of carbon content, the peaks and upper terraces of C–curves were 723 K and 773 K, respectively.

(2) With a decrease in the transformation temperature from 798 K to 600 K, the microstructure during the early stage of transformation changed from simple or complex structures containing allotriomorphic ferrite, pearlite, DP, and/or pro-eutectoid cementite plate to UB. Similarly, the morphological feature of UB changed from B-II type to B-III type with a decrease in temperature.

(3) B3 existed between 758 K and 773 K in all alloys used. It was independent of carbon content, which was different from the case of low carbon alloys.

(4) B3 measured in this study corresponded to the temperature of the intersection point between T0′ and the γ′(γ + θ) phase boundary.

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