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

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Bainite in steel is an industrially useful structure. However, the controlling factor of its transformation start point is not known clearly. In this study, to clarify the effect of carbon content on the bainite transformation start temperature (B_s), we evaluated the dilatation curve and the microstructure in low carbon Fe–9Ni alloys. As a result, B_s decreased with increasing of carbon content. Furthermore, the driving force of partitionless transformation from fcc to bcc at B_s , which was calculated considering nickel segregation, was approximately constant at 400 J/mol in all alloys. This value is consistent with the driving force required for partitionless growth of ferrite, as reported in a previous study. This consistency suggests that B_s depends on the martensitic growth behavior of lath-shaped ferrite, which is determined by the super-cooling starting from the T_0 line.

KEY WORDS: bainite; martensite; phase transformation; driving force.

1. Introduction

Bainite in steel is a very useful structure because it has a relatively high strength along with good formability and toughness. Many types of high strength steels contain bainite; therefore, it is important to accurately know the bainite transformation start temperature (B_s).

Three definitions have been proposed for B_s .^{1–4)} The first one defines B_s as the upper critical temperature for the formation of bainite;^{1–3)} this temperature is called microstructural- B_s .³⁾ Figure 1 shows a schematic for this B_s through a time-temperature-transformation (TTT) diagram and continuous-cooling-transformation (CCT) diagram for a low carbon alloy.^{1,2,4)} As shown in Figs. 1(a) and 1(b), this temperature appears at the terrace of the diagrams for alloys in which the C-curve of diffusion transformation (ferrite and pearlite) is much slower than that of bainite transformation. The second definition is that B_s is the upper limiting temperature for an incomplete bainite transformation in a TTT diagram;⁴⁾ this temperature is called kinetic- B_s . Although kinetic- B_s appears in alloys that contain Mo,⁵⁾ Cr,⁶⁾ Mn,⁷⁾ and Nb,⁸⁾ its appearance in low-alloyed Fe–C alloys,^{2,7–9)} in which the C-curves of bainite and diffusion transformation (ferrite and pearlite) overlap extensively, as shown in Fig. 1(c), is unclear. The third definition is that B_s is the transformation start temperature during a discrete heat treatment.^{10–12)} Although it is valuable for steel making, B_s

depends on many heat treatment conditions that affect the pre-existent microstructure of bainite at the start of its transformation (*e.g.*, cooling rate). In this study, we used the first definition of B_s ; this B_s corresponds to the upper limit of the partitionless growth of ferrite.¹⁾

Several models have been proposed for determining B_s .^{1,2,4)} The most reliable model is Bhadeshia's model.¹⁾ It suggests that B_s is parallel to and below the T_0 temperature, at which fcc and bcc of the same composition have the same free energy. In the model, bainite transformation is displacive transformation and requires the driving force of martensitic transformation, which is larger than that required for the displacive growth of bainitic ferrite. This model can explain some results showing a decrease in B_s with increasing carbon content.^{10,13,14)} However, in some results,^{2,9,15,16)} it shows that B_s does not depend or slightly depends on the carbon content. Tsuzaki *et al.*²⁾ proposed another model for the B_s of low carbon steels. They assumed that a B_s independent from carbon content corresponds to the martensite transformation start temperature (M_s) in ultra low carbon steel and proposed that the nucleation of bainitic ferrite starts in the carbon-free regions of low carbon steel. However, there is no evidence for the existence of carbon-free regions, neither experimentally nor theoretically. Therefore, these models cannot explain the B_s above the T_0 temperature in high carbon steels.^{3,9,17)}

The aim of this study is to clarify the mechanism of bainite transformation start. To this end, we evaluate the effect of carbon content on B_s in Fe–9Ni alloys,^{2,18)} in which nickel does not exhibit a solute drag like effect.⁶⁾ We

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Fig. 1. Transformation start line on the schematic CCT and TTT diagrams for low carbon alloys. (a) CCT diagram wherein the ferrite and bainite regions are well separated.¹⁾ (b) TTT diagram wherein the ferrite and bainite regions are well separated.^{1,2,4)} (c) TTT diagram in which the ferrite and bainite regions extensively overlap.^{2,4)}

focus on low carbon alloys in this paper. We define that bainite, which transforms just below B_s, contains a group of lath-shaped ferrites with/without cementite. This definition

confirms to the upper bainite categorized by Ohmori *et al.*¹⁹⁾

2. Experimental Procedure

Table 1 shows the chemical compositions and T₀ temperatures of the five Fe–9Ni alloys used in this study. To avoid ferrite transformation around B_s, Alloys A, B, and C contained a small amount of boron. Alloy A was an ultra low carbon alloy containing 0.0031 mass%C. Alloys B and D contained 0.05 mass%C, and Alloys C and E contained 0.10 mass%C. Other elements in these alloys were present in a very small amount. The T₀ temperatures shown in Table 1 were calculated via Thermo-Calc using the TCFe8 material database. In the calculation, we considered only Fe, C, and Ni and neglected other elements.

Ingots of the alloys melted in vacuum were heated at 1 523 K and hot-rolled over 1 173 K to make plates with a 3 mm thickness. These plates were grinded to remove the decarburized layer around both surfaces and cold-rolled to sheets with a 1 mm thickness. These sheets were cut to small specimens with dimensions of 10 mm × 80 mm.

These specimens were heated via electrical heating and cooled by N₂ or He gases. The transformation behavior during the heat treatment was evaluated based on a dilatation curve obtained using a laser displacement meter.

We performed two tests. The first test was a continuous cooling test. The specimens were austenitized at 1 473 K or 1 223 K for 60 s. To save time, the cooling rate was changed twice for each specimen. After austenitization, the specimens were cooled to just above A_{e3}, which was calculated, at a relatively high cooling rate, *i.e.*, 20 K/s. Below A_{e3}, the specimens were cooled at several cooling rates between 0.01 and 300 K/s until the transformation was complete. Then, they were cooled at a rate of 30 K/s.

The second test was an isothermal holding test. The specimens were austenitized at 1 473 K or 1 223 K for 60 s and then cooled to several temperatures at 30 K/s. 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 influence of the surface, 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 B_s for short time, which containing small amount of ferrite and/or bainite, we observed the whole observation field.

Table 1. Chemical compositions and T₀ temperatures of the alloys used in this study.

Alloy	Chemical compositions/mass%								T ₀ /K (calculated)
	C	Si	Mn	P	S	Ni	B	N	
A	0.0031	<0.01	0.02	0.005	0.001	9.06	0.0023	0.0013	884
B	boron added	0.052	<0.01	0.02	0.005	9.08	0.0024	0.0014	863
C		0.094	<0.01	0.02	0.005	9.09	0.0024	0.0017	846
D	boron free	0.050	<0.01	0.01	0.005	9.03	<0.0003	0.0017	865
E		0.099	<0.01	0.01	0.005	9.06	<0.0003	0.0017	845

3. Results

3.1. Ultra Low Carbon Fe–9Ni–B Alloy

Figure 2 shows the dilatation curves during continuous cooling with various cooling rate after austenitization at 1 473 K for Alloy A. Under rapid cooling, *i.e.*, faster than 10 K/s, the volume expansion corresponding to transformation started at ~765 K and finished at ~700 K. On the other hand, at a cooling rate of 10 K/s or less, the expansion started at ~833 K. Alloy A showed two distinct transformation temperatures, 765 K and 833 K corresponding to grey lines in Fig. 2.

Figure 3(a) shows an OM image of the Alloy A specimen cooled at 200 K/s, in which the transformation occurred between 765 K and 708 K. The prior austenite grain in the center of Fig. 3(a) consists of some packets containing many parallel laths, as illustrated in Fig. 3(b). This morphological feature is similar to that of a typical lath martensite structure²⁰⁾ and upper bainite structure.²¹⁾

Figure 3(c) shows an OM image of the Alloy A specimen cooled at 1 K/s. In this specimen, phase transformation started at 834 K and finished at 753 K. Most of this specimen transformed above 765 K, which was the transformation start temperature during the rapid cooling of Alloy A. However, in this image, the microstructure is similar to that shown in Figs. 3(a) and 3(b). The prior austenite grain illustrated in Fig. 3(d) consists of some packets, similarly.

In other words, microstructures having the same morphological characteristics were generated at two different transformation start temperatures. The difference between these temperatures was ~70 K. These two temperatures likely correspond to M_s and B_s .

To clarify the transformation behavior around 833 K, which is a high transformation temperature, we performed an isothermal holding test. **Figure 4** shows the dilatation curves during isothermal holding at several temperatures. The vertical axis shows normalized dilatation, which is expressed by the following equation:

$$f = \frac{D - D_\gamma}{D_\alpha - D_\gamma},$$

where f represents normalized dilatation, D represents dila-

Fig. 2. Dilatation curves during continuous cooling in the Alloy A specimen.

Fig. 3. Microstructure of the Alloy A specimens cooled continuously at (a) (b) 200 K/s, and (c) (d) 1 K/s. (a) (c) OM image. (b) (d) Schematic image of the prior austenite grain in OM image.

Fig. 4. Dilatation curves during isothermal holding in the Alloy A specimen.

tation, and D_α and D_γ represent the presumed dilatations for a full ferrite structure and an austenite structure at the holding temperature, respectively.

Below 833 K, the volume expansion corresponding to transformation started within 10 s at the latest and stopped at around 100 s. At 808 K and 823 K, an incomplete transformation occurred clearly. On the other hand, the transformation just above 833 K progressed very slowly. Moreover, the incubation period at 848 K was longer than that at 873 K. This means that there is a bay around 833 K in the TTT diagram.

Figure 5 shows an OM image captured in the early stage of transformation during isothermal holding. In the specimen held at 848 K for 10 000 s (Fig. 5(a)), there are some allotriomorphic ferrite grains arrowed in an image over prior austenite grain boundaries. On the other hand, in the specimens held below 833 K (Figs. 5(b) and 5(c)), the microstructures consist of laths and there are no ferrite grains. This indicates that the shape of ferrite changed from

Fig. 5. OM images of the Alloy A specimen held isothermally (a) at 848 K for 10 000 s, (b) at 823 K for 600 s, and (c) at 808 K for 500 s.

an allotriomorph to a lath with decreasing transformation temperature from 833 K, which is a high transformation start temperature.

3.2. Low Carbon Fe-9Ni-B Alloys

Figure 6 shows the CCT diagrams for Alloys B and C, which contain a small amount of carbon with boron. The transformation start points during continuous cooling after austenitization at 1 473 K (○) clearly show the M_s of these alloys. However, B_s is not clear from these points because the velocities of bainite transformation in them were smaller than that in Alloy A. To accelerate the transformation, we decreased the austenitization temperature to decrease the size of prior austenite grains. The transformation start points of the specimens austenitized at 1 223 K (▲) show clear terraces between M_s and T_0 (Fig. 6).

Figure 7 shows OM images of the specimens of Alloys B and C held isothermally just below the terrace for a short time. The volume fractions that were transformed in these specimens were assumed to be less than 5% based on the dilatation curves. In the Alloy B specimen held at 798 K for 1 000 s (Fig. 7(a)), a group of lath-shaped ferrites on a prior austenite grain boundary was observed. This ferrite group was surrounded by lath martensite layer, which transformed during quenching after isothermal holding. Moreover, no cementite was observed in or around this ferrite group. This ferrite group is called B-I type upper bainite.¹⁹⁾

The same microstructure appears in the Alloy C specimen held at 783 K for 10 000 s (Fig. 7(b)). The images in this figure indicate that the terraces in the CCT diagrams are

Fig. 6. CCT diagrams of boron-added low carbon alloys. The dots correspond to the transformation start point during continuous cooling after austenitization at 1 473 K (○) and 1 223 K (▲).

Fig. 7. OM images of boron-added low carbon alloys held isothermally: (a) Alloy B at 798 K for 1 000 s, and (b) Alloy C at 783 K for 10 000 s.

Fig. 8. CCT diagrams of Alloys D and E.

the B_s of Alloys B and C. Moreover, in these specimens, there is no allotriomorphic ferrite, film-like ferrite, and Widmanstätten ferrite.

3.3. Low Carbon Fe–9Ni Alloys

Figure 8 shows the CCT diagrams of Alloys D and E. In the CCT diagram, although M_s is clear, it is difficult to determine the B_s of alloy D. Because the velocity of ferrite transformation in this alloy, which contains no boron, is faster than that in Alloy B containing boron, the terrace would be covered by the ferrite transformation start line in the CCT diagram. In the CCT diagram of Alloy E, although the terrace seems to be around 800 K, it is not clear.

To clarify the B_s , we observed the microstructure in the early stage of transformation during isothermal holding. Figure 9 shows the OM images of Alloy D. In the Alloy D specimen held at 818 K for 3 000 s (Fig. 9(a)), allotriomorphic ferrite was observed over a prior austenite grain boundary. On the other hand, in Fig. 9(b), which shows the microstructure at 808 K, some upper bainite was observed around prior austenite grain boundaries along with allotriomorphic ferrite grains. This result proves that the B_s of Alloy D is between 818 K and 808 K.

Similarly, the OM observation results for Alloy E are shown in Fig. 10. In the specimen held at 808 K, many allotriomorphic ferrite grains and no bainitic ferrite lath were observed. At 793 K, the reconstructive ferrite was the main product in the early stage of transformation. However, there was a small amount of bainitic ferrite, which was generated at 793 K, as shown in Fig. 10(b). The B_s of Alloy E was between 808 K and 793 K, probably close to 793 K.

Figure 11 shows the TTT diagrams of Alloy D and E. The points in these diagrams express the transformation start

Fig. 9. OM images of the Alloy D specimen held isothermally (a) at 818 K for 3 000 s, and (b) at 808 K for 1 000 s.

times determined by dilatation curves. And their shapes correspond to the microstructure in the early stage of isothermal transformation. These TTT diagrams are similar to the

Fig. 10. OM images of the Alloy E samples held isothermally (a) at 808 K for 10 000 s, and (b) at 793 K for 1 200 s.

Fig. 11. TTT diagrams of Alloys D and E. The dots correspond to the transformation start point and the production of the transformation in the early stage. ▲: ferrite, △: ferrite and bainite, ○: bainite.

schematic TTT diagram shown in Fig. 1(c). In the microstructures just below B_s , bainitic ferrite is a minor structure when compared with the reconstructive ferrite structure, *i.e.*, the structures of allotriomorphic ferrite, film-like ferrite, and Widmanstätten ferrite. This TTT diagram does not show a clear terrace and bay around B_s .

3.4. Effect of Carbon Content on B_s

Table 2 shows the B_s of the alloys used herein. In the boron-added alloys, *i.e.* A, B and C, B_s decreased with increasing carbon content from 0.003 mass% to 0.10 mass%. In the boron-free alloys, the change of B_s with the carbon content was not clear because the initial products just below B_s were very complex, as shown in Figs. 9(b) and 10(b). However, it seems that B_s decreased with increasing carbon content in the boron-free alloys, similarly.

As shown in Table 2, M_s increased with decreasing carbon content, and the boron content had little or no effect on M_s . Alloy A, which contained 31 mass-ppm carbon showed the highest M_s , *i.e.*, 765 K; however, this value was lower than the B_s of all the alloys used herein.

4. Discussion

We considered the mechanism of the relationship between B_s and carbon content in terms of the driving force. Before the consideration, it was important to check the segregation of nickel in the specimens. The specimens were heated at 1 473 K after cold rolling, and were quenched. Figure 12 shows the histograms of the nickel content measured via an electron probe microanalysis (EPMA). The histograms in 5 alloys resemble each other closely with respect to the positions and shapes. The nickel contents were distributed from 7 mass% to 10 mass% in all the alloys used herein.

Bainite and martensite transformations should start in nickel-poor regions. The detection accuracy of the transformation from fcc to bcc using the dilatation technique is ~1% transformation rate, empirically. Therefore, we guessed that the effective nickel content for B_s was the nickel content at 1% from the bottom of the distribution. Table 3 shows the effective nickel content $W_{Ni,E}$, which is the nickel content in the nickel-poor region of each alloy. The temperatures $T_{0,E}$ in Table 3 were calculated as T_0 temperatures using the nickel content at the nickel-poor regions ($W_{Ni,E}$).

Figure 13 shows the driving force of the partitionless transformation from fcc to bcc (ΔG) at B_s and M_s in Table 2. Open and solid points in Fig. 13 correspond to ΔG at B_s and M_s , respectively. Figure 13(a) shows ΔG along with the chemical compositions of the alloys in Table 1; ΔG

Table 2. B_s and M_s of the alloys used in this study.

Alloy	A	B	C	D	E
Carbon/mass%	0.0031	0.052	0.094	0.050	0.099
Boron/mass%	0.0023	0.0024	0.0024	<0.0003	<0.0003
T_0 /K	884	863	846	865	845
B_s /K	833	810	793	808–818	793–808
M_s /K	765	679	653	683	643

corresponds to the supercooling from T_0 in Table 1. In this study, the ΔG at B_s did not depend on the carbon and boron content. All open points in Fig. 13(a) are along the constant energy line (dashed line). On the other hand, the ΔG at M_s increased with the carbon content and was not affected by the boron content.

Although Fig. 13(a) supposed homogenous alloys, all the alloys contain the Ni segregation shown in Fig. 12. ΔG at B_s should be calculated with $T_{0,E}$ and $W_{Ni,E}$ in Table 3. Figure 13(b) shows revised ΔG at B_s and M_s for all the alloys. The values of ΔG at B_s were similar, *i.e.*, 400 J/mol represented by the dashed line in Fig. 13(b), regardless of the carbon and boron contents. This value is consistent with the driving force required for partitionless growth of ferrite, as proposed by Bhadeshia.¹⁾ This consistency suggests that the detection of bainite transformation start requires the bainitic ferrite to grow rather than only nucleating.

If B_s depends on the supercooling from T_0 , the bainite transformation progress in the specimen will vary owing to the nickel segregation. At the temperature just below B_s , the driving force in the nickel-rich region is not sufficient for

the bainitic ferrite to grow without partitioning. Therefore, bainite transformation will progress partially in the specimen. In Alloy A, the addition of 1 mass% nickel reduces the driving force to 100 J/mol at B_s . In order to obtain the driving force of 100 J/mol extra, the supercooling degree of 19 K is further needed from 833 K. It means that the local B_s in the nickel-rich region is lower than the B_s in the nickel-poor region.

Figure 14 shows the distribution of the local B_s in Alloy A calculated with the consideration of nickel segregation shown in Fig. 12(a). In Fig. 14, at 823 K, 10% austenite can

Fig. 12. Histograms of the local nickel content evaluated via EPMA for (a) the boron-added alloys and (b) the boron-free alloys.

Table 3. Effective nickel content and T_0 temperature for the bainite and martensite transformation start.

Alloy	A	B	C	D	E
$W_{Ni,E}/\text{mass}\%$	7.40	7.49	7.62	7.50	7.42
$T_{0,E}/\text{K}$	923	900	879	900	882

Fig. 13. Driving force for the partitionless transformation from fcc to bcc at B_s and M_s . (a) ΔG calculated using the bulk nickel content and (b) ΔG calculated with the effective nickel content ($W_{Ni,E}$).

Fig. 14. Histogram of the local B_s in Alloy A estimated considering nickel segregation.

transform to bainite because its local B_s is higher than 823 K; 50% of austenite has a local B_s temperature lower than 808 K. Therefore, it is assumed that the half of the austenite will remain after the bainite transformation stops at 808 K. This tendency corresponds to the incomplete transformation behavior in Alloy A shown in Fig. 4.

The incomplete transformation of bainite is generally explained by disappearance of the driving force occurring with the progress of bainite transformation.⁸⁾ However, the carbon content of Alloy A is very small, and the driving force hardly changes even when considering the increase of carbon. In this study, the reason for the incomplete transformation in the ultra low carbon alloy would be the effect of nickel segregation in the specimen.

Figure 15 shows the B_s and M_s evaluated in this study with the A_3 , T_0 and T_0' lines for Fe–7.6 mass%Ni–C ternary system. 7.6 mass%Ni corresponds roughly to $W_{Ni,E}$ of the alloys used herein (Table 3). T_0' is the temperature at which the driving force of the partitionless transformation from fcc to bcc accounts for specific value.^{1,8)} The B_s expressed by open dots in Fig. 15 decreases with carbon increasing along with T_0' at which the supercooling from T_0 is 400 J/mol. It indicates that the B_s depends on the possibility of the partitionless growth of ferrite.¹⁾ However, the nucleation behavior of bainite transformation is not clarified by the evaluation of B_s in this study.

Tsuzaki *et al.*²⁾ proposed that the nucleation of bainitic ferrite occurs without partitioning in the local carbon-poor region and that the B_s of the low carbon alloy matches the M_s of the ultra low carbon alloy. In Alloy A, which contains 31 mass-ppm carbon, B_s is 60 K higher than M_s . According to previous studies,^{22–24)} this carbon content can decrease

the M_s by about 60 K in ultra low carbon Fe–C alloys. The B_s of carbon-free alloys may coincide with their M_s , which agrees with Tsuzaki's proposal about the nucleation of bainitic ferrite.

5. Summary

To clarify the effect of carbon content on the B_s of low carbon Fe–9 mass%Ni alloys, we prepared CCT and TTT diagrams and observed the microstructure in the early stage of transformation during isothermal holding. In this study, bainite refers to a lath-shaped ferrite group that generates over M_s .

(1) B_s decreased with increasing carbon content from 0.003 to 0.10 mass%.

(2) The B_s values measured in this study were higher than M_s in Fe–9 mass%Ni–0.002 mass%B–0.003 mass%C alloy.

(3) Supercooling from the T_0 line at B_s was constant in all specimens regardless of the amount of carbon and boron. The driving force of the partitionless transformation, calculated after considering nickel segregation, was approximately 400 J/mol.

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Fig. 15. B_s and M_s plotted on paraequilibrium A_3 , T_0 and T_0' lines of the Fe–7.6 mass%Ni–C ternary system.