Crystallographic Analysis of Transformation Behavior of Acicular Ferrite from B1-type Compounds in Steels

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Crystallographic analysis was carried out on acicular ferrites (AFs) of steel that nucleated on B1-type compounds embedded in steels during the isothermal transformation from austenite (γ) to ferrite (α), and the orientation relationship between α , γ , and the compounds and the AF formation behavior were correlated by varying the B1-type compound, steel, and transformation temperature. The results demonstrated that all the AFs satisfy the Kurdjumov–Sachs orientation relationship with the prior γ , and in addition, some of the AFs have specific low-index orientation relationships with the B1-type compounds and nucleate earlier than others. Among the specific low-index orientation relationships, furthermore, AFs satisfying the Baker–Nutting orientation relationship, which gives the lowest interfacial energy between α and a compound, nucleate preferentially.

KEY WORDS: acicular ferrite; hetero-nucleation; crystal orientation relationship; transformation.

1. Introduction

Intragranular ferrite (IGF), which nucleates at inclusions dispersed in austenite (γ) grains, has attracted considerable attention as a means of suppressing the degradation of toughness due to grain-coarsening in the heat-affected zone of steel welds. Thus, many studies have been conducted to clarify the formation mechanisms of IGF, and several factors that promote ferrite (α) nucleation have been reported^{1–5)} such as lattice matching between α and inclusions, thermal stress or strain due to the difference in the thermal expansion between γ and inclusions, and changing the chemical composition in the vicinity of the γ /inclusion interface.

Regarding lattice matching, B1-type oxides and nitrides (B1) with the NaCl-type lattice structure have been reported to be effective for IGF nucleation^{6,7)} because they have a crystal orientation relationship with low lattice mismatch with α , called the Baker–Nutting orientation relationship (B-N OR); $\{100\}_{B1} // \{100\}_{\alpha}$, $\langle 100 \rangle_{B1} // \langle 110 \rangle_{\alpha}$. However, to rigorously discuss the effect of the lattice coherency on the nucleation of IGF, the change in the γ -to- α transformation behavior with the transformation temperature should be considered. It is well known that the morphology of IGF is usually divided into two types, polygonal type and acicular type. In the case of polygonal ferrite (PF), ferrites form by a diffusional transformation at relatively high temperatures, and thereby, the B-N OR can be easily satisfied between α

and B1 upon the nucleation of α .^{6,7)} In the case of acicular ferrite (AF), on the other hand, ferrites form by a displacive transformation at relatively low temperatures and it is well known that a specific crystal orientation relationship exists between the product phase α and the mother phase γ , which is called the Kurdjumov–Sachs orientation relationship (K-S OR); $\{111\}_{\gamma} // \{110\}_{\alpha}$, $\langle 110 \rangle_{\gamma} // \langle 111 \rangle_{\alpha}$. Under this constraint in the relationship between α and γ , it is still unclear whether a specific crystal orientation relationship between AF and B1 can exist because of the assumption that B1-type compounds do not have a specific orientation relationship with austenite.⁸⁾ Although several studies have been conducted to consider the crystal orientation relationship between α , γ , and inclusions,^{9–12)} the kinetics of the IGF nucleation at inclusions, as well as the effect of the crystal orientation relationship between them, has not yet been clarified.

In this study, we focused on the formation behavior of IGF at relatively low temperatures to clarify the crystal orientation relationship between α , γ , and B1 and its correlation with the transformation behavior of AF. In particular, the selection of the specific crystal orientation relationship between α and B1 was evaluated.

2. Experimental Procedure

Two types of steels, C–Mn steel and C–Ni steel, were prepared and their chemical compositions are shown in **Table 1**. The bainite transformation start temperature (B_s) and martensite transformation start temperature (M_s) calculated using empirical equations^{13,14)} are also shown in Table 1.

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Three different single-crystalline B1-type compounds, TiO, TiN, and MgO, having similar lattice parameters were also prepared. The steels were cut to rod specimens with 6 mm height and 8 mm diameter. The B1-type compound particles with diameters from about 3 to 30 μm were sandwiched by the two rod specimens, which were then hot-pressed in the height direction to about 50% reduction at 1100°C by a thermomechanical simulator (Thermecmaster-Z, Fuji Electronic Industrial). The specimens were cooled then reheated to 1150°C and held for 2400 s to embed the B1-type compound particles inside the austenite grains as a result of their grain growth. Subsequently, the specimens were cooled rapidly at 50°C s⁻¹ by He gas and held isothermally at 400, 500, or 600°C to induce the ferrite transformation. Finally, these specimens were gas-quenched to room temperature. A schematic of this thermomechanical cycle is shown in Fig. 1. Note that none of the IGFs nucleated when the specimens were cooled to room temperature at the same cooling rate without isothermal holding.

The heat-treated specimens were cut normal to the interface between the two rod specimens. The observation area was etched by 2% nital after mechanical polishing and the microstructure was observed by optical microscopy. Additionally, the same area was observed by field-emission scanning electron microscopy (FE-SEM; JSM7001, JEOL) and crystallographic analysis was conducted by electron backscattering diffraction (EBSD; EDAX/TSL) with OIM software (OIM DC and OIM analysis 7). In the crystallographic analysis, not only the B-N OR but also other low-

index ORs such as $\{100\}_{\text{B1}}//\{110\}_{\alpha}$, $\langle 110 \rangle_{\text{B1}}//\langle 110 \rangle_{\alpha}$ and $\{110\}_{\text{B1}}//\{112\}_{\alpha}$, $\langle 100 \rangle_{\text{B1}}//\langle 110 \rangle_{\alpha}$ (hereafter, called OR2 and OR3, respectively) were also considered because these low-index ORs were also found in our previous studies.^{7,11)}

The procedure of evaluating the crystal orientation relationship between α , γ , and B1 is as follows. The $\langle 100 \rangle$ directions of the B1-type compound of interest are set to parallel to RD, TD, and ND in the pole figure. Then specific low-index ORs, the B-N OR, OR2, and OR3, are plotted according to the orientation of the B1-type compound as shown in Fig. 2. In this study, a misorientation within 15° from the exact orientation relationship is allowed for each specific low-index OR according to our previous study^{7,11)} in which it was found that the misorientation allowance depends on the B1-type compound and the maximum allowance is about 15° even in the case of a diffusional PF transformation. In this case, the B-N OR and OR3 partially overlap in the pole figure, and the ferrites in the overlapping area, if they exist, are considered to satisfy the B-N OR. Twenty-four variants of the K-S OR in one austenite grain are also plotted in the pole figure using the martensite phase surrounding the B1-type compounds, because the martensite is formed from the remaining austenite by quenching after isothermal holding for a certain period during AF formation. By comparing the experimental results with the above analytical results, the crystal orientation relationship between α , γ , and B1 is evaluated.

3. Results and Discussion

The effect of the transformation temperature on the morphology and the orientation relationship of IGF was investigated using the C-Mn steel. The specimens were austenitized at 1150°C and then cooled to and isothermally held at 400, 500, or 600°C for 10–20 s as shown in Fig. 1. In the case of the isothermal transformation at 600°C, only polygonal ferrites (PFs) were observed around the B1-type compounds (TiO) as shown in Fig. 3(a). Both PFs and acicular ferrites (AFs) were observed at 500°C (Fig. 3(b)), and only AFs were observed at 400°C (Fig. 3(c)). By decreasing the transformation temperature, it was confirmed that the morphology of IGF changed from a polygonal to acicular shape.

The orientation relationship between α , γ , and B1 was also investigated and the result corresponding to Fig. 3(b) is shown in Fig. 4. The crystal orientation of the TiO is rotated and its $\langle 100 \rangle$ directions are set to parallel to RD,

Table 1. Chemical compositions (in mass%) of the steels used in this study and their transformation temperatures.

	C	Si	Mn	Ni	Fe	Bs/°C	Ms/°C
C-Mn steel	0.35	0.20	1.76	—	Bal.	524	337
C-Ni steel	0.32	0.20	0.20	10.3	Bal.	475	215

Fig. 1. Thermomechanical cycle used in this study.

Fig. 2. (001) α pole figures for (a) B-N OR, (b) OR2, and (c) OR3.

Fig. 3. Optical micrographs of IGFs nucleated on TiO (a) after 20 s at 600°C, (b) after 10 s at 500°C, and (c) after 10 s at 400°C.

Fig. 4. Crystal orientation analysis for PFs and AF nucleated on TiO after 10 s at 500°C (corresponding to Fig. 3(b)). (a) Image quality map with highlighted TiO, PFs, and AF. (001) α pole figures for (b) PFs with a circle having 15° misorientation from the B-N OR and (c) AF with a circle having 15° misorientation from OR2.

Fig. 5. Fractions of IGFs categorized on the basis of the crystal orientation relationship between α , γ and B1 generated at 400, 500, and 600°C.

TD, and ND as shown in Fig. 4(a). The crystal orientations of the PFs specified in Fig. 4(a) are plotted in the circle which is centered at the B-N OR and has a 15° radius, but away from the K-S OR (Fig. 4(b)). The crystal orientation of the AF specified in Fig. 4(a) is plotted in the circle which is centered at OR2 and has a 15° radius, and also placed in the K-S OR (Fig. 4(c)). The result demonstrates that the PFs satisfy the B-N OR with TiO but not with the prior γ and the AF satisfies OR2 with TiO and the K-S OR with prior γ simultaneously in the case of Fig. 4. The orientation relationship between α , γ and B1 was evaluated for about 50 IGF grains nucleated at TiO at each isothermal transformation temperature, and the fractions of the orientation relationships are shown in Fig. 5. In Fig. 5(a), the orientation relationships are categorized into four types, where “SORs” means that the IGF satisfies one of the specific low-index ORs with the compound (B-N OR, OR2, and OR3) but not with the prior γ , “KS” means that the IGF satisfies the K-S OR with the prior γ but not with the compound, “SORs&KS” means that the IGF satisfies both the K-S OR with the prior γ and one of the specific low-index ORs with the compound, and “other” means that no specific low-index

ORs were found between α , γ and B1. From the results for 500 and 600°C, most of the PFs, delineated by the dashed line in the figure, have specific low-index ORs with the compounds. The results for 400 and 500°C show that every AF, delineated by the chain line in the figure, satisfies the K-S OR with the prior γ and many of the AFs also satisfy specific low-index ORs with the compounds. Details of the SORs between α and B1 are summarized in Fig. 5(b). In the case of PF, the fraction of IGFs satisfying the B-N OR with the compound is more than 80%. This result clearly demonstrates that the B-N OR is preferentially selected during the diffusional transformation from γ to PF. On the other hand, in the case of AF, the fraction of BN is smaller but the fraction of other low-index ORs, OR2 and OR3, is larger. Thus, a clear difference between PF and AF in terms of the crystal orientation relationship with the compound was found; in the case of PF, the B-N OR dominates, while in the case of AF, not only the B-N OR but also other low-index ORs, OR2 and OR3, are selected.

The change in the selection of the orientation relationships between α and B1 during the isothermal transformation of AF at 400°C with time was evaluated using combinations

Fig. 6. Change in the fraction of IGFs satisfying a specific orientation relationship with the B1-type compounds, TiO, TiN, and MgO with the holding time at 400°C.

of the C–Mn steel and different B1-type compounds, and the results are shown in **Fig. 6**. For every condition, about 50–70 IGF grains were analyzed. From the result using TiO, in the early stage of the transformation such as after 10 s of isothermal holding, more than 80% of IGFs satisfy the low-index ORs with the compound as well as the K-S OR with the prior γ , while the fraction of IGFs that do not satisfy any low-index ORs with the compound becomes large with increasing holding time up to 30 s. In the cases of TiN and MgO, the fraction of IGFs satisfying the low-index ORs with the compound is relatively large in the early stage of the transformation and becomes small with time. These results suggest that IGFs having specific crystal orientation relationships with both the prior γ and the compound form first, and then IGFs only satisfying the K-S OR with the prior γ start to form after a relatively long incubation time.

To further investigate the selection mechanism of the specific low-index ORs (B-N OR, OR2, and OR3) between AF and B1, the C–Ni steel was subjected to the same experiments as the C–Mn steel, because the driving force of the γ -to- α transformation in C–Ni steel at a given temperature is always smaller than that in C–Mn steel at the same temperature and the selection of the coherent interface between α and B1 is affected by the driving force. It is assumed that a coherent interface with smaller interfacial energy is more likely selected when the driving force is small. **Figure 7** shows the change in the fractions of the orientation relationships for AFs that nucleate at TiN during isothermal holding at 400°C. At the beginning of AF formation, at 7 200 s in this case, more than 80% of AFs satisfy the specific low-index ORs, and the fraction satisfying the B-N OR is the largest among the low-index ORs. Subsequently, the fractions satisfying OR2 and OR3 increase with time, and finally the fraction satisfying other relationships becomes the largest. This result also suggests that the B-N OR was selected preferentially at the beginning of the transformation, then other low-index ORs were also selected with increasing time, and finally more random ORs became dominant rather than low-index ORs such as the B-N OR, OR2, and OR3. Even in the formation of AFs satisfying the K-S OR with the prior γ , it appears that AFs having lower interfacial energy between α and B1 can nucleate with a shorter incubation time on the B1-type compounds.

When AFs nucleate on B1-type compounds with specific

Fig. 7. Change in the fraction of IGFs satisfying the specific orientation relationship with TiN during isothermal holding at 400°C.

ORs, they must satisfy the K-S OR with the prior γ simultaneously. The probability that AF simultaneously satisfies the specific ORs with the compounds and the K-S OR with the prior γ was calculated using the Euler space assuming that B1 and γ are randomly oriented. The matrix representing the rotation given by the Euler angle (Bunge notation), $g=(\phi_1, \phi, \phi_2)$, can be given as

$$R(g) = \begin{pmatrix} \cos\phi_1\cos\phi_2 - \sin\phi_1\sin\phi_2\cos\phi & -\cos\phi_1\sin\phi_2 - \sin\phi_1\cos\phi_2\cos\phi & \sin\phi_1\sin\phi \\ \sin\phi_1\cos\phi_2 + \cos\phi_1\sin\phi_2\cos\phi & -\sin\phi_1\sin\phi_2 + \cos\phi_1\cos\phi_2\cos\phi & -\cos\phi_1\sin\phi \\ \sin\phi_2\sin\phi & \cos\phi_2\sin\phi & \cos\phi \end{pmatrix} \quad (1)$$

Firstly, the $\langle 001 \rangle$ direction of the B1-type compound is fixed at a Euler angle $g_0=(0, 0, 0)$ as a reference. The rotation of α satisfying the specific ORs, the B-N OR, OR2, and OR3, with this B1-type compound can be represented by the following equation:

$$\Delta g_{\alpha}^{OR} \equiv (\phi_1, \phi, \phi_2) = \begin{cases} (45, 0, 0) & : \text{B-N} \\ (45, 90, 45) & : \text{OR2.} \\ (170, 90, 135) & : \text{OR3} \end{cases} \quad (2)$$

In addition, the symmetry due to the cubic structure should be considered. In the case of a cubic structure, there are 24 combinations of planes and directions as shown in Ref. 15). The Euler angle of α satisfying specific ORs with a compound including the symmetry can be given by

$$R(g_{\alpha}) = S_i \cdot R(\Delta g_{\alpha}^{OR}) \cdot S_j \cdot R(g_0) = S_i \cdot R(\Delta g_{\alpha}^{OR}) \cdot S_j \cdot E, \quad (3)$$

where S_i is the rotation of symmetry and E is the identity matrix. The Euler angle for the rotation of the K-S OR is given by¹⁶⁾

$$\Delta g_{KS} \equiv (\Delta\phi_1, \Delta\phi, \Delta\phi_2) = (5.77, 48.19, 5.77). \quad (4)$$

Regarding this rotation, the symmetry of the cubic structure also has to be taken into account. Therefore, the Euler angle of γ satisfying the K-S OR with α is represented by

$$R(g_{\gamma}) = S_i \cdot R(\Delta g_{KS}) \cdot S_j \cdot R(g_{\alpha}). \quad (5)$$

From Eqs. (3) and (5), the Euler angle of γ satisfying the K-S OR with α that can satisfy specific orientation relationships is obtained as

Fig. 8. Plane map of Euler space obtained by the calculation in the case of the B-N OR.

$$R(g_{\gamma/\alpha/B1}) = S_i \cdot R(\Delta g_{KS}) \cdot S_j \cdot R(\Delta g_{\alpha}^{OR}) \cdot S_k \cdot E. \dots (6)$$

Since a misorientation within 15° from the exact orientation relationship is allowed when the crystal orientation relationship between α and B1 is evaluated, the fraction of the sphere that is centered at the Euler angle obtained by Eq. (6) and has a 15° radius in the Euler space was calculated to evaluate the probability. In this calculation, the overlapping space between the B-N OR and OR3 is considered to be the B-N OR as well as the experimental analysis. **Figure 8** shows the calculation result of Euler space with the spheres obtained when the ferrite simultaneously satisfies the K-S OR with the prior γ and the B-N OR with the B1-type compound. In this figure, there are some overlapping spaces, but they are not counted more than once in the calculation. Based on the above calculation, the probability that the ferrite simultaneously satisfies both the K-S OR and the B-N OR is about 18.4% when a B1-type compound exists in an austenite grain with a random orientation. The probabilities for the cases of OR2 and OR3 are 19.9% and 9.5%, respectively. Each probability is shown in **Fig. 9**. The ferrites, which are categorized as others with the fraction of 52.2%, satisfy the K-S OR with the prior γ while they nucleate without any specific ORs with B1-type compound. On the other hand, the fractions of ferrites that satisfy the specific ORs with both γ and the B1-type compound in the

Fig. 9. Probabilities that specific orientation relationships are satisfied between α , γ and B1.

early stage of the transformation, obtained by the experiment and shown in Figs. 6 and 7, are larger than the probabilities obtained by the calculation and shown in Fig. 9, and therefore the fractions obtained by experiments do not result from stochastic nucleation events but from nucleation with low-index planes having low interfacial energies, even in the case of a displacive AF transformation.

From the results, it was found that AFs, which nucleate at a relatively low temperature and have to satisfy the K-S

OR with the prior γ , can nucleate without any specific ORs with B1-type compound probably due to the large driving force of transformation. However, we assume that ferrites satisfying the K-S OR with the prior γ and the B-N OR with the compound nucleate preferentially because they have the lowest interfacial energy between α and γ and between α and B1; such nucleation sites where the specific relationships between α , γ and B1 are satisfied are selected first. Subsequently, those satisfying the K-S OR and other low-index orientation relationships, OR2 and OR3, nucleate, and as time proceeds, those satisfying the K-S OR with the prior γ but no specific orientation relationships with the compound start to nucleate at the remaining sites of the compound.

4. Conclusions

(1) It was confirmed that the Baker–Nutting orientation relationship between ferrites and B1-type compounds is preferentially selected in the case of polygonal ferrites, which nucleate at a relatively high temperature.

(2) In the case of acicular ferrites, which nucleate at a relatively low temperature, ferrites satisfying the Baker–Nutting orientation relationship between α and B1 nucleate preferentially in spite of the constraint imposed by the Kurdjumov–Sachs orientation relationship between α and γ in the early stage of the transformation.

(3) Acicular ferrites having other orientation relationships also nucleate as the transformation proceeds.

(4) In the early stage of the transformation, the fraction of acicular ferrites satisfying both the Kurdjumov–Sachs orientation relationship with the prior austenite and specific

crystal orientation relationships with the compound obtained by experiments is higher than that calculated from the Euler space assuming stochastic nucleation events. Nucleation with low-index planes having low interfacial energies takes place intensively in the early stage of the transformation even in the displacive acicular ferrite transformation.

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