In Situ EBSD Analysis on the Crystal Orientation Relationship between Ferrite and Austenite during Reverse Transformation of an Fe-Mn-C Alloy

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The crystal orientation of nucleating austenite during reverse phase transformation of a C-Mn steel has been investigated by in situ EBSD technique using a heating stage of FE-SEM at temperatures ranging from RT to 800°C. It has been found that the multiple interfaces between a nucleating austenite grain and the surrounding parent ferrite grains are preferentially selected as the Kurdjumov-Sachs (K-S) relationship. More than 50% of austenite grains are surrounded with two or more parent ferrite grains satisfying the K-S relationship with a deviation within 7°. Based on these findings, a nucleation model at triple junction is proposed, which supposes that the orientation of nucleating austenite is selected to satisfy the K-S relationship or the orientation relationship close to K-S relationship at two of the interfaces to ferrite grains, and at the other interfaces to minimize the misorientation from the K-S relationship. [doi:10.2320/matertrans.MAW201602]

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1. Introduction

A conventional steel transforms several times between the austenite (γ) and the ferrite (α) phases during the steel production process, and the texture control by cooling and heating treatments must be predicted by simulation models. The mechanism of variant selection has been intensively studied on the γ→α phase transformation in the cooling process3–7) because the eventual texture after cooling has a significant influence on the various mechanical properties of the final products. The crystal orientation selection rule during the reverse phase transformation (α→γ) in the heating process is also important for controlling the microstructure in the steel making process. For instance, the variant of transformed γ on heating affects not only the γ texture but also the successive martensite morphology after quenching in recently developed heating and cooling processes, by affecting the mobility of the α/γ interfaces8–13).

The mechanism of variant selection of transforming γ has been discussed in the literature. In studies on the texture memory effect during α→γ→α transformation14–22), it is commonly suggested that the γ texture after the reverse transformation is not randomly selected but is determined under the rule on the variant selection of transforming γ. Various models to explain the variant selection rules of transforming γ during reverse transformation have been proposed.

Tomida et al. proposed the variant selection model for α→γ transformation based on the orientation relationship at multiple interfaces with different α grains showing the texture memory effect. They assumed that the orientation of γ during reverse transformation is selected in order to satisfy the Kurdjumov-Sachs (K-S) relationship23) or near K-S relationship at two or more interfaces with adjacent α grains at the same time6,7). They call this hypothetical relationship the double K-S relationship. Here, the term "double K-S relationship" is used not only to refer to having the K-S or near K-S relationship with two adjacent parent grains but also to refer to having such relationship with more than three parent grains. This model had firstly developed for the forward phase transformation and was later applied to the texture change in the α−γ−α transformation15). Based on the model, they have developed a calculation method to predict the texture change during α→γ→α transformation with about 10° of the tolerance angle for the near K-S relationship and shown that the texture change has been quantitatively predictable.

Concerning the model of the double K-S relationship, Lischewski et al. observed the α→γ phase transformation of a C-Mn steel containing micro-alloying elements by in situ electron back-scattering diffraction (EBSD) measurement, and analyzed the orientation relationship between the parent α and the transformed γ19,20). A considerable number of transformed γ grains satisfy the multiple K-S relationship with adjacent α grains. They considered the reason for the high frequency of the multiple K-S relationship to be preferential nucleation of the K-S variant with lower interface energy.

For the case of γ nucleation at a triple junction or corner of the α matrix, the orientation relationships at three or four α/γ interface must be considered. All α/γ interfaces around the nucleus contribute to reducing the interfacial energy for stable nucleation by satisfying the specific orientation relationship, and some of the interfaces will play a role in the growth of the nucleus to a stable size with large mobility. The present study has focused on the orientation relationship between parent α grains and transformed γ grains to clarify whether the orientation relationship are the dominant factors in the transformation of γ or some deviation from the ideal double K-S relationship is required. The reverse transformation of γ from recrystallized α microstructure is investigated, and based on a crystallographical analysis between parent α grains and transformed γ grains, the mechanism of the variant selection rule is discussed.
2. Experimental Procedure

An alloy with a chemical composition of Fe-1 mass%Mn-0.1 mass%C was melted by vacuum induction furnace. The ingot was hot-rolled to 2 mm in sheet thickness at a final rolling temperature of 840°C, and the rolled sheet was cooled to 700°C by water spray and subsequently cooled in a furnace to room temperature at a cooling rate of 20°C/h. The microstructure obtained after hot-rolling and cooling was measured to consist of 95% ferrite and 5% pearlite in volume fraction by an optical microscope observation. The hot-rolled sheet was then cold-rolled to 1 mm in thickness. A high temperature EBSD measurement was carried out using a field emission SEM (JSM-7800F) equipped with a special heating stage made by TSL Solutions Japan. The sample dimensions for the heating stage were 5×7×0.5 mm. The surface of the specimen was polished mechanically and then chemically with colloidal silica suspension. The observed surface of the sample is three quarters of the original sheet thickness, which is considered to be the representative of the microstructure of the sample prepared.

With the specimen mounted on the heating stage for EBSD measurement, two sets of chromel-alumel thermocouples were fixed to measure the temperature, the first was on the sample surface and the other was on the heating plate. The EBSD measurement area and its sampling step were carefully determined at each temperature in order to avoid artifacts such as sample drift and vaporization of surface elements in a vacuum.

3. Results

The change in microstructure and crystal orientation during α−γ transformation with heating was investigated using high temperature EBSD measurement at a temperature above 700°C. The temperature region of the reverse transformation is between 700°C and 850°C in the present sample. Figures 1(a), (b) and (c) show image quality mappings of the EBSD band patterns obtained by the EBSD measurement, where the α matrix is indicated by gray-scale level and the γ grains confirmed by the crystal orientation information are superimposed by red color images. Figure 1(d) indicates a graph of the sample temperature change on heating during the in situ EBSD measurement. Since it is taken about 10 minutes for an EBSD measurement, the sample was kept for 10 minutes at each temperature. As seen in Fig. 1 (b), reverse transformed γ grains clearly appeared at 800°C and most of them are observed at the triple junction of the α grains. When the temperature was increased to 825°C, the γ grains grew into the neighboring α grains and some γ grains newly transformed at triple junctions. Typical changes in size of growing γ grains are indicated by arrows in Fig. 1 (b) and (c).

In order to check the surface effect on the reverse transformation, the texture of the γ grains was analyzed using the discrete binning method24) of the EBSD analysis software (OIM analysis), where each orientation of measured points is distributed into discrete Euler space to obtain the orientation distribution function (ODF). The results are represented in Fig. 2 (a) and (b), which are the ODF’s of the 0° and 45° section in φ2, respectively. The transformed γ grains are strongly

Fig. 1 Image quality mappings of microstructure observed at (a) 700°C, (b) 800°C, (c) 825°C (γ grains are indicated by red color) and (d) a graph showing the sample temperature change during in situ SEM-EBSD measurements. Arrows in Fig. 1 (d) indicate measured temperatures at 800°C and 825°C.

Fig. 2 ODF’s of the transformed γ measured by in situ EBSD analysis, where (a) 0° section and (b) 45° section in φ2 are indicated (the contour lines correspond to the level intensity by 1 time to random texture).

Fig. 3 Image quality mapping of the microstructure measured at 730°C, which is used for the evaluation shown in Table 1 (The corresponding austenite grains are labeled as A to K).
oriented to (011)[211] as shown in 0° section and (110)[112] and (112)[132] position as shown in 45° section. These distribution profiles correspond to the typical major orientations of the γ texture that appears after α-γ reverse transformation in steels[14,15]. Thus, the observed orientations of the transformed γ grains in the present experiment are confirmed to be selected in the same mechanism as that in the transformation in the bulk of the specimens.

By performing similar high temperature in situ EBSD measurements up to 800°C several times, orientation mapping data including parent α grains and 154 transformed γ grains were investigated. The heating conditions in the present experiments are almost similar to that indicated in Fig. 1(d).

The orientation relationship between the transformed γ grains and parent α grains was analyzed based on the K-S relationship, which is generally recognized as the orientation relationship between α and γ in iron. One of the microstructures evaluated at 730°C is shown in Fig. 3, where the γ grains are labeled A to K. Most of the γ grains have three or four interfaces to the neighboring α grains. The misorientation from the K-S relationship is calculated between each γ grain and the neighboring α grains. For example, taking γ grain D in Fig. 3, 24 α variants of the K-S relationship were calculated from the orientation of γ. Then, the rotation angle between those variants and the neighboring α grain were calculated by using the following equations[25],

\[
M = g'^T(V_i R_i g)^T \quad (1)
\]

\[
\Delta \theta = \arccos((M_{11} + M_{22} + M_{33} - 1)/2) \quad (2)
\]

Here, \(R_i\) represents the rotation matrix for the cubic symmetry that is the combination of rotations of 90° about the basal crystal axes. \(V_i\) is the rotation matrix to change the crystal coordinate from fcc to bcc according to the K-S relationship, which can be expressed as a rotation of 90° about the (112) axis of γ crystal[4,15]. \(g'\) and \(g\) are the orientation matrices of the γ and α respectively.

The misorientation \(\Delta \theta\) for γ grains A to K toward their neighboring α grains are listed in Table 1. In this table, \(\Delta \theta\) for each γ grain is shown in the columns named Interfaces 1 to 4 in ascending order. The following two things are indicated:

i) At Interface 1, the orientation relationships quite close to the K-S relationship with \(\Delta \theta\) of 3° or less is satisfied for seven γ grains.

ii) When the orientation relationships within 10° in \(\Delta \theta\) are taken into account[30], six γ grains have such orientation relationships at Interface 2 and Interface 3 in addition to the K-S relationship at Interface 1.

By analyzing all 154 γ grains measured in the present study, the frequency of γ grains satisfying Conditions i) and ii), that is the double K-S relationship with the tolerance angle of 3° and 10°, was 53%.

In order to analyze the deviations from the K-S relationship at the α/γ interfaces, two kinds of mutually perpendicular misorientations are measured. One is the misorientation of the close-packed-planes \(\Delta \theta_{cpp}\) and the other is that of the close-packed-directions \(\Delta \theta_{cpd}\). These misorientations are calculated using the following equations:

\[
\Delta \theta_{cpp} = \arccos((g'^T P_{cpp}) \cdot ((g)^T P_{cpp}^T)) \quad (3)
\]

\[
\Delta \theta_{cpd} = \arccos((g'^T P_{cpd}) \cdot ((g)^T P_{cpd}^T)) \quad (4)
\]

Here, \(P_{cpp}\) and \(P_{cpd}\) are the vectors of the normal direction of the close-packed-plane and the close-packed-direction respectively.

Figure 4 shows the relationship between the \(\Delta \theta_{cpp}\) and \(\Delta \theta_{cpd}\) of all α/γ interfaces of the 154 γ grains measured in the present study. In this figure, the red points indicate the interface with the smallest \(\Delta \theta\) of each γ grain (Interface 1) and the blue points are with the second smallest \(\Delta \theta\) (Interface 2), and the white points are with the third or fourth smallest \(\Delta \theta\) (grouped as Interface 3). The K-S relationship and the Nishiyama relationship[26] are also shown in the same notation.

Many of the α/γ interfaces have an orientation relationship quite close to the K-S relationship, that is, the orientation relationship with a misorientation of less than 2° in \(\Delta \theta_{cpp}\) and less than 2° in \(\Delta \theta_{cpd}\) is held at 16% of all the interfaces[19,27]. These orientation relationships are satisfied at Interface 1. Also, within a misorientation of 2° in \(\Delta \theta_{cpp}\) and 6° in \(\Delta \theta_{cpd}\), there are 28% of all the interfaces; they are of Interface 1 and Interface 2. Orientation relationships with larger misorientations in \(\Delta \theta_{cpp}\) and \(\Delta \theta_{cpd}\) are satisfied mainly at Interfaces 2 and 3 but less at Interface 1.

The frequencies of α/γ interfaces are re-plotted against the misorientation from the K-S relationship. The result is shown in Fig. 5. The number of orientation relationships is the highest at the K-S relationship (\(\Delta \theta = 0°\)) and the number decreases rapidly until \(\Delta \theta \sim 7°\). 40% of all interfaces satisfy the orientation relationships within 7° in \(\Delta \theta\). 65% of the interfaces satisfy the orientation relationship within 15° in \(\Delta \theta\) and 80% are within 21°. Almost all the orientation relationships are within 31° in \(\Delta \theta\), although \(\Delta \theta\) is crystallographically possible to be up to 43°. The significance of the distribution of misorientation must be considered along with the statistics without particular orientation relationship, which is discussed in the next section.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>The misorientation from the K-S at each austenite grain in Fig. 3.</th>
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<tbody>
<tr>
<td>γ grain</td>
<td>The misorientation from the K-S relationship Δθ</td>
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<tr>
<td></td>
<td>Interface 1</td>
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<tr>
<td>A</td>
<td>0°</td>
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<tr>
<td>B</td>
<td>1°</td>
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<tr>
<td>C</td>
<td>11°</td>
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<td>K</td>
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4. Discussion

It has been proofed that the in situ EBSD measurement at high temperature has been a great experimental technique to observe the reverse phase transformation from bcc to fcc in steel. Although the sample drift and other contaminations at high temperature must be taken care for the EBSD measurement in SEM, the short time measurement less than 10 minutes at each temperature is sufficient to obtain a good quality mapping of orientational information.

In the present study, the orientation relationship between reverse transformed γ grains and parent α grains has been discussed on the basis of misorientation from the K-S relationship that is a representative relationship between bcc and fcc transformation. It has been clarified that the K-S relationship or the orientation relationship with a misorientation up to 7° from the K-S relationship is often satisfied at two or more α/γ interfaces. The latter orientation relationships are mostly those having the close-packed planes of the two phases parallel to each other, which lie between the K-S relationship and N relationship. The frequency of the double K-S relationship observed in the present study is 53%, which is consistent with that reported by Tomida et al. and by Lischewski et al.

Lischewski et al. proposed in their study on the reverse transformation of steel that the γ nucleus with multiple interfaces of the K-S relationship or approximate K-S relationship are favorable to nucleate since such a nucleus is expected to have low interfacial energy. In their model of γ nucleation, it is supposed to occur at α grain boundary near a triple junction with satisfying an approximate K-S relationship at either two or three of the interfaces to α grains. This condition on nucleation accounts for the variant selection of transformed γ by the double K-S relationship and thus the texture change in α−γ−α phase transformation.

To thoroughly understand the precise condition of γ nucleation, the contribution of all interfaces including the third interface must be considered. Supposing that nucleation prefers to take place at the corner or triple junction of grain boundaries in the parent phase since the shape of the nucleus at those sites is thermodynamically stable, the nucleus is likely to face three or four different parent grains during the nucleation process. For such a nucleation condition, the orientation relationships at all interfaces may be effective.

In the following, to clarify the condition of γ nucleation at the triple junction, the influences of the orientation relationships at all of the α/γ interfaces are taken into account. The frequency of the α/γ interfaces to be misoriented from the K-S relationship is evaluated against Δθ for α/γ Interfaces 1, 2 and 3. In addition, Δθ distribution without orientation relationship has been calculated. In the calculation, a hundred thousand pairs of α and γ orientations are generated by computer, and Δθ for each pair of orientations is estimated in the same way explained above. The results are shown in Figs. 6 (a) to (c).

At Interface 1 (Fig. 6 (a)), 67% of the orientation relationships satisfy the K-S relationship quite accurately with Δθ of 3° or less. The K-S relationship is preferentially selected as the orientation relationships for the reverse transformation from α to γ.

At Interface 2 (Fig. 6 (b)), 50% of the orientation relationships are within Δθ up to 7°, and most of which are with Δθ from 3° to 7°. The interfaces certainly have orientation relationships near the K-S relationship, however, the distribution becomes broader than that of Interface 1 and mostly the interfaces are with Δθ larger than 3°. The misorientation from the K-S relationship would be due to the restriction on selectable γ orientations by holding the K-S relationship at Interface 1.

Actually, there is only a small possibility of γ orientation satisfying the K-S relationships at two interfaces with Δθ less...
At Interface 3 (Fig. 6 (c)), the misorientation from the K-S relationship becomes larger, whereas the frequency is still larger than that without any orientation relationship, for instance at $\Delta \theta = 9^\circ$ and $13^\circ$. The reason for the large deviation at Interface 3 is that there is no selectable orientation relationship close to the K-S relationship whereas the tendency to reduce the misorientation $\Delta \theta$ at Interface 3 is still effective.

Such interfaces with a large misorientation from the K-S relationship are expected to migrate dominantly during the subsequent $\gamma$ growth$^{31,32}$. Based on the tendency of the orientation relationships at each interface, the nucleation of $\gamma$ at the triple junctions is supposed to occur as schematically depicted in Fig. 7. The $\gamma$ nucleus is with the K-S relationship and the orientation relationship near the K-S relationship at the two $\alpha/\gamma$ interfaces of nucleating $\gamma$. At the other interface, although the orientation relationships may also tend to be close to the K-S relationship in order to minimize the misorientation, $\alpha/\gamma$ interface with a large $\Delta \theta$ always occurs, which may contribute to the fast growth of $\gamma$ nucleus since the mobility of the interface for diffusive phase transformation is considered to be large for the interface with large $\Delta \theta$.

To further understand how the orientation of the transformed $\gamma$ at the triple junction is selected to minimize the misorientation from the K-S relationship, the misorientation from the K-S relationship at all the $\alpha/\gamma$ interfaces of each $\gamma$ grain is evaluated by a summation of the misorientation angles $\Delta \theta$ from the K-S relationship at each $\alpha/\gamma$ interface using the following evaluation.

$$ D = \Delta \theta_1 + \Delta \theta_2 + \Delta \theta_3 + \cdots + \Delta \theta_N \quad (5) $$

Here, N represents the number of $\alpha/\gamma$ interfaces, which is at most 4 in the present study.

The ideal minimum value of $D$, $D_{\text{min}}$ for the observed triple junction is estimated by calculating all the D values for the possible orientations of nucleating $\gamma$ in the Euler space, that is $\phi_1:0^\circ - 90^\circ$, $\Phi: 0^\circ - 90^\circ$, $\phi_2: 0^\circ - 90^\circ$. In this calculation, the experimentally measured orientation of $\gamma$ is substituted by all the orientations in the Euler space, whereas the $\alpha$ orientations are the same as those experimentally observed adjoining the $\gamma$ nucleation site. The difference between the D and $D_{\text{min}}$ is expressed as $\Delta D$, which is considered as an indicator of simultaneous minimization of $\Delta \theta$. The evaluation of $\Delta D$ is carried out for all the $\gamma$ grains observed at the triple junction. The distribution of $\Delta D$ is shown in Fig. 8. The number of $\gamma$ grains is highest at $\Delta D = 0^\circ$, and the frequency decreases with increasing $\Delta D$. 59% of observed $\gamma$ grains were within $20^\circ$ in $\Delta D$, and even within $10^\circ$ of $\Delta D$ there are 32% $\gamma$ grains. If the orientation relationship at the third interface was selected randomly, such distribution could not be realized, rather the maximum in the distribution would be found at a finite $\Delta D$. The orientation of nucleating $\gamma$ has a strong inclination to minimize the misorientation from the K-S relationship for all the $\alpha/\gamma$ interfaces. This indicates that
the nucleation process is crystallographically related to not only one or two adjacent parent $\alpha$ grains but all the adjacent $\alpha$ grains at the triple junction or corner of the matrix, which supports the nucleation model proposed in the present study.

5. Conclusions

The orientation relationship between nucleating $\gamma$ and neighboring $\alpha$ grains was studied based on the in situ EBSD measurement at high temperature. As a result of the analysis of the misorientation from the K-S relationship at each $\alpha/\gamma$ interface, the following conclusions were obtained.

(1) The K-S relationship is satisfied at one of the $\alpha/\gamma$ interfaces of transformed $\gamma$ with a deviation of 3° or less.

(2) In addition to the K-S relationship at one of the $\alpha/\gamma$ interfaces, more than 50% of nucleated $\gamma$ grains have another interface with $\alpha$ that holds the orientation relationship close to the K-S relationship with a deviation of 7° or less, which means the double K-S relationship is satisfied. The deviation from the exact K-S relationship is mostly the one with the misorientation of the close-packed-direction up to 6° and the misorientation of the close-packed-plane up to 2°.

(3) At the third and fourth interfaces, the orientation relationship tend to be close to the K-S relationship. The interfaces with large misorientation from the K-S relationship always occur when the $\gamma$ grains nucleate at corners or triple junctions of grain boundaries of $\alpha$.

(4) Based on the present results, a $\gamma$ nucleation model at a triple junction has been proposed, where the $\gamma$ nucleation is accompanied with two parent $\alpha$ grains satisfying the K-S relationship and the orientation relationship close to the K-S relationship. The presence of other interfaces with large deviation from the K-S relationship is required the rapid migration to promote the $\gamma$ growth.

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