In-Situ SEM/EBSD Observation of $\alpha/\gamma$ Phase Transformation in Fe-Ni Alloy

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The in-situ SEM/EBSD technique has been applied to observe the dynamic of ferrite($\alpha$) $\rightarrow$ austenite($\gamma$) phase transformation and to examine the orientation relationship between $\alpha$ and $\gamma$ phases in an Fe-9.5 at% Ni alloy. Random grain boundaries and grain-boundary triple junctions were found to act as preferential nucleation sites for $\gamma$ phase. The effectiveness of triple junctions as the $\gamma$ phase nucleation site in phase transformation increased with increasing number of random boundaries intersecting at a triple junction. Approximately 90% of allotriomorphic $\gamma$ phase possessed either a Kurdjumov-Sachs (K-S) or a Nishiyama-Wasserman (N-W) orientation relationship (OR) with the $\alpha$ parent grains. The $\gamma$ allotriomorphs with a special OR with one of the adjoining $\alpha$ parent grains preferentially grew into the $\alpha$ grain having the special OR with them, while the allotriomorphs with special ORs with both of the $\alpha$ grains grew into both adjoining $\alpha$ grains symmetrically.

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1. Introduction

Recent developments of automated scanning electron microscope (SEM)/electron backscattered diffraction (EBSD) systems make it possible to investigate microstructural evolution on the basis of crystallographic data. $^{1,2}$ To date, electromigration of Al, $^3$ microstructural change in Al during tensile testing at high temperatures, $^3$ phase transformation in Ti$^5$ and Co, $^5$ grain growth in Cu thin films, $^5$ phase transformations in Fe,$^7$ grain boundary migration in Al$^9$ and abnormal grain growth in nanocrystalline Ni$^9$ have been studied using the in-situ SEM/EBSD technique. These previous studies demonstrated that the in-situ SEM/EBSD is an advanced technique suitable for the study of microstructural change in microstructure. The fact has motivated us to observe ferrite ($\alpha$)/austenite ($\gamma$) phase transformation in iron using the in-situ SEM/EBSD technique.

Microstructures in iron alloys and steels are often controlled by manipulation of the $\alpha/\gamma$ phase transformation. Thus, it is of great importance to understand the nature of the phase transformation. Many researchers have studied $\alpha/\gamma$ phase transformations and martensitic transformations using various techniques such as X-ray diffraction (XRD), transmission electron microscopy (TEM), SEM/electron channeling pattern (ECP) and SEM/EBSD. It has been reported that the product phases often form with specific orientation relations with the parent phase, such as the Kurdjumov-Sachs (K-S) and Nishiyama-Wasserman (N-W) orientation relationships, to reduce the energy of interphase boundaries. In order to study the $\alpha/\gamma$ phase transformation using conventional techniques, it is necessary to preserve a high-temperature phase such that it still exists at room temperature. Because this is sometimes difficult, in-situ TEM$^{1,11}$ and in-situ optical microscopy (OM)$^{12}$ have been applied to understand the nature of this phase transformation. Onink et al.,$^{10}$ who carried out in-situ TEM observation of $\gamma$ to $\alpha$ phase transformation in Fe-C, found that the $\alpha$ grains grew along the $\gamma/\gamma$ grain boundaries, and the $\alpha/\gamma$ interphase boundaries migrated in the manner that acceleration and deceleration stages were repeated. Watanabe et al.$^{13}$ carried out in-situ observations of $\alpha/\gamma$ phase transformation in an Fe-4.2 at%Cr using optical microscope equipped with a heat stage. They determined grain boundary character by SEM/EBSD technique before the in-situ observations. It was found that the greater the number of random boundaries at triple junction, the greater its effectiveness as a nucleation site for the $\gamma$ phase. Recently, Kirch, et al. reported in-situ SEM/EBSD observations of $\alpha/\gamma$ phase transformation in a low carbon steel up to 1273 K using a laser powered heat stage.$^7$ They found that nucleation sites are predominantly at matrix grain boundaries and triple junctions, and that the temperature window of the $\alpha \rightarrow \gamma$ phase transformation was shifted upwards by approximately 15 K to higher temperatures in comparison to the $\gamma \rightarrow \alpha$ phase transformation.

One main motivation of the present study is to reveal the influence of grain boundary character and triple junction character on $\alpha/\gamma$ phase transformation in iron, particularly on the orientation relation between the nucleated $\gamma$ phase and parent $\alpha$ phase. In addition, it is of interest to observe how the nucleated $\gamma$ phase grows into the $\alpha$ phase, and how the interphase boundaries make an interaction with the grain boundaries in the $\alpha$ parent phase during the phase transformation.

2. Experimental Procedures

We used an Fe-9.5 at%Ni alloy, in which the $\alpha$ to $\gamma$ phase transformation starts at a lower temperature than that in pure iron, for in-situ SEM/EBSD observations of the $\alpha/\gamma$ transformation, because of the limitation in maximum temperature (approximately 1000 K) of the heating stage used.

Samples for EBSD observations were prepared according to the following procedures. Electrolytic Fe (99.99%) and Ni (99.99%) were arc-melted into a button-shaped sample in an
Ar atmosphere. The sample was annealed at 1473 K for 24 h to achieve a homogeneous composition. Thereafter, it was rolled to a plate of 0.3 mm in thickness (reduction rate: 65%) at room temperature, and annealed at 873 K for 3.6 ks and at 1013 K for 600 s, and then slowly cooled in the furnace. Specimens for in-situ SEM/EBSD observation, of dimensions 8 mm x 5 mm, were cut from the plate using a spark cutter and mechanically polished using waterproof SiC papers, and finally buff-finished to a mirror surface using colloidal silica.

The in-situ SEM/EBSD observations were conducted with a Hitachi cold FEG-SEM (S4200) equipped with an OIM system from TSL. A heat stage for a Hitachi cold FEG-SEM (S4200) equipped with an OIM (max. 40 V, 45 W) with a size of 1000 K. Specimens were heated using a ceramic plate heater (max. 40 V, 45 W) with a size of 10 x 10 x 0.175 mm³. A tungsten sheet was put between the ceramic heater and the specimen in order to prevent diffusional bonding and to achieve uniform temperature. A CA-thermocouple was used for the temperature measurements. The temperature was precisely controlled by PID within an error of ±0.1 K.

The SEM was operated at 30 kV accelerating voltage, 10 μA emission current and 15 mm working distance. The electron beam was scanned with a step size between 0.1 and 0.2 μm. The analysis time per one step was 0.015 s using the two phase indexing procedure, and the time per one flame was ~3600 s. The α to γ phase transformation behavior was observed at temperatures between 774 and 1013 K. The specimen was heated at a heating rate of 30 K/min, and held at 774 or 783 K for EBSD observation. Then, the temperature was ramped up again to the next observation temperature. Such a procedure was repeated up to the maximum temperature of 1013 K.

3. Results

3.1 Initial microstructure

Figure 1 presents an inverse pole figure (IPF) map of the as-prepared specimen just before α/γ phase transformation. The microstructure consisted of a mixture of larger grains and smaller grains with the average grain sizes of 20 and 4.7 μm, respectively. The grains contained some residual strain due to the γ → α phase transformation occurring after annealing.

Fig. 1 An inverse pole figure micrograph of an Fe-9.5 at% Ni alloy before in-situ observations.

3.2 In-situ EBSD observation

Snapshots of the transient microstructure during α to γ phase transformation at different temperatures are shown in Fig. 2. In these micrographs, the α phase and the γ phase are displayed in light gray and dark gray colors, respectively, and the character of grain boundaries are shown by differently colored lines; low-angle, Σ3, CSL boundaries (Σ ≤ 29) other than Σ3 and random boundaries are shown by thin black, bold white, thin white and bold black lines, respectively. The γ phase is found to nucleate preferentially at α/α grain boundaries and grain boundary triple junctions, compared with the grain interior of the α phase, as was already suggested many years ago by J. W. Cahn. However, not all of grain boundaries and triple junctions behave equally as nucleation sites for the γ phase. Similar observations have been reported on the phase transformation of an Fe-4.18 at%Cr alloy, Ti,4) Co5) and low-carbon steel.7) Although the character or misorientation of grain boundaries could be related to the tendency for formation of γ phase, little reliable experimental evidence is available. The allotriomorphic γ phase, which designates the γ phase nucleated at α/α grain boundaries, tends to grow along the α/α grain boundaries to either a corner of curved grain boundary or a triple junction, and then grows towards the α grain interior away from the grain boundary. It is interesting to see that some γ allotriomorphs (γ3, γ4 in Fig. 2(b)) grew preferentially into one of the adjoining α grains, while the allotriomorph γ1, for example, grew into both adjoining α grains symmetrically. The γ phase that nucleated in the grain interior, e.g. γ2 and γ7, was found to grow in an anisotropic manner. From these observations, it is considered that the γ phase grains are likely to grow in such a manner that the resultant α/γ interphase boundaries are energetically favorable.

In addition, we found that grain growth of γ phase is sometimes accompanied by the formation of Σ3 grain boundaries (see Fig. 2). Such Σ3 boundaries can be classified into two types: one is a boundary with a low deviation angle (~1°) from the exact Σ3 CSL relation and the other is the boundary with a relatively high deviation angle. It is considered that the Σ3 boundaries with a low deviation angle would be formed according to the mechanism proposed by Gleiter,14) that is, by a two dimensional nucleation process on the [111] planes of the growing grain. On the other hand, the Σ3 boundaries with a higher deviation angle could be formed as a result of a contact between two different variants of γ grains which are nucleated in the same α grain with a nearly K-S OR.

4. Discussion

4.1 Preferential nucleation site

Considering the nucleation site for the γ phase, we found that the γ phase nucleated much more frequently at grain boundaries and grain boundary triple junctions than in the grain interior. The numbers of γ phase grains nucleated was 75 at the grain boundaries and 31 at the triple junctions, whereas it was only 3 in the grain interior. Table 1 shows the number and the number fraction of γ phase grains nucleated at low-angle boundaries, CSL boundaries (3 ≤ Σ ≤ 29),
random boundaries and the α grain interior. For comparison, the grain boundary character distribution, defined by the fraction of grain boundary length, of the sample observed is also shown in the Table 1. It is evident from Table 1 that the number fraction of γ allotriomorphs found at random boundaries (60%) is considerably higher than the length fraction of random boundaries (41%). From the results, we can say that the random boundaries act as preferential sites for γ phase nucleation on α to γ phase transformation, in agreement with the earlier results of Aaron and Aaronson.\(^{15}\)

The correlation between the character of a triple junction and its effectiveness as nucleation site for γ phase at triple junctions was considered. As suggested by Fortier et al.,\(^{16}\) the triple junctions are classified into four types in view of the

Table 1 The number of γ phase grains nucleated at intragrain, grain boundaries and triple junctions. The number fraction of γ allotriomorphs at low-angle boundaries, CSL boundaries (Σ < 31) and random boundaries, and the grain boundary character distribution determined by length fraction of grain boundaries are also shown in the table.

<table>
<thead>
<tr>
<th>Nucleation site</th>
<th>Intragrain</th>
<th>Grain boundary</th>
<th>Triple junction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of γ grains (Fraction)</td>
<td>3</td>
<td>LAB 18(0.24)</td>
<td>CSL 12(0.16)</td>
</tr>
<tr>
<td>GBCD</td>
<td>—</td>
<td>0.45</td>
<td>0.14</td>
</tr>
</tbody>
</table>
connectivity of different types of grain boundaries at triple junctions: R-R-R (3R-TJ), R-R-\Sigma (2R-TJ), R-\Sigma-R (1R-TJ) and \Sigma-\Sigma-R (0R-TJ), where R and \Sigma stand for random boundary and CSL boundary, respectively. The percentages of the triple junctions where 4 phase was precipitated are 50% at 3R-TJs, 38% at 2R-TJs, 26% at 1R-TJs and 0% at 0R-TJs. The percentage was calculated such that the number of 4 phase at different types of triple junctions was normalized by the total number of each type of triple junctions where 4 phase was precipitated. This is consistent with the result reported by Watanabe et al. for 4/\gamma phase transformation in Fe-Cr alloy.\textsuperscript{12}

4.2 Orientation relationship between product 4 phase and parent 4 phase

Figure 3(a) presents a snapshot of the transient microstructure at 883 K, showing the allotriomorph 4\gamma formed at the \alpha_1/\alpha_2 grain boundary in the Fe-9.5 at% Ni alloy. Stereographic projections showing (b) K-S orientation relation between \alpha_2 and 4\gamma, and (c) N-W orientation relation between \alpha_1 and 4\gamma.

Fig. 3  (a) A snapshot of the transient microstructure at 883 K, showing the allotriomorph 4\gamma formed at the \alpha_1/\alpha_2 grain boundary in the Fe-9.5 at% Ni alloy. Stereographic projections showing (b) K-S orientation relation between \alpha_2 and 4\gamma, and (c) N-W orientation relation between \alpha_1 and 4\gamma.

had such a special crystallographic OR with only one of the adjacent \alpha parent grains, the K-S OR predominated compared with the N-W OR. On the other hand, when the allotriomorphs possessed crystallographic ORs with both of adjacent \alpha grains, the K-S OR and N-W OR were observed to an equal extent, (K-S/K-S: 2, K-S/N-W: 3, N-W/N-W: 1).

As described above, we found that random boundaries act as effective nucleation sites for the 4 phase during phase transformation. In addition, the 4 allotriomorphs nucleated at random boundaries frequently have either K-S or N-W ORs. This is probably because the total interface energy decreases as a high-energy random boundary is replaced by a low-energy interphase boundary with a crystallographic orientation relation.

In the case of nucleation at triple junctions, the numbers of 4 phase grains with one or two interphase boundaries which have special ORs (K-S or N-W ORs) with the \alpha parent grains were 3 at 2R-TJs, and 2 at 0R-TJ (3L-TJ) and 1 at 1R-TJ, respectively. Moreover, the number of the 4 phase having the ORs with all sides of \alpha grains was 2 at 0R-TJ (3L-TJ) or 1R-TJ.

4.3 Grain growth of 4 phase

It has been reported that the allotriomorphs which have a crystallographic OR with only one of the adjoining parent grains tend to predominantly grow into the grain with which they do not have a specific OR, because of the lower mobility of interphase boundary with the OR than for the boundary without OR.\textsuperscript{17} However, as seen in Fig. 2, two \gamma allotriomorphs (4\gamma_1, 4\gamma_2), which have a crystallographic OR with one of the adjoining \alpha grains, predominantly grew into the \alpha grain with a specific OR. The area of interphase boundary increases when a product phase grows, and as a result, the total energy of the boundary increases. However, if there is a specific OR
between the parent phase and product phases, the increase in the boundary energy may be lower than if no specific OR exists. Because the phase transformation observed was in the two phase regime, the driving force for α-γ phase transformation due to change in the volume free energy would be insignificant. Therefore, it is likely that interphase boundary motion is governed by interphase boundary energy rather than mobility. This is consistent with the fact that the allotriomorph γ1, which grew into both sides of α grains homogeneously, possessed specific ORs with both sides of adjoining α grains.

As seen in the left-hand side in Fig. 2, intragranular γ phase grew in an anisotropic manner towards the upper-left direction of the micrograph. Recently, Watanabe et al.12) have discussed the mechanism of anisotropic migration of α/γ interphase boundaries on the basis of the plane matching boundary model.18) One of their authors (T.W.) previously reported twin formation during the α → γ phase transformation in Fe-9.5 at% Ni alloy, and the chief results obtained are as follows. The in-situ SEM/EBSD technique was applied to observe the α → γ phase transformation in Fe-9.5 at% Ni alloy, and the nature of the transformation was examined on the basis of crystallographic data obtained by EBSD, particularly concentrating to the influence of grain boundaries and triple junctions on the α → γ phase transformation. The chief results obtained are as follows.

1. In-situ SEM/EBSD was shown to be a useful technique for observations of the dynamics of microstructural evolution, such as phase transformation in materials.

**4.4 Interaction of growing γ grain with α/α grain boundary**

The allotriomorph γ5 in Fig. 2(c) grew until it met the low-angle boundary GB (α4/α5), and the GB (α4/α5) retarded further growth of the γ5 grain significantly (Fig. 2(d), (e)). Conversely, the γ5 grain appeared to force the GB (α4/α5) to migrate (Fig. 2(f)). The interaction between migrating boundaries can be explained as follows. The allotriomorph γ5 had a near the K-S OR with both α4 and α5 grains. Because the α5 grain does not adjoin to the growing γ5 grain, there is no special OR between the α5 and γ5 grains, and then the interphase boundary formed when they meet each other would not be a low energy boundary. Migration of such an incoherent interphase boundary into α5 grain would be energetically unfavorable, so that the GB (α4/α5) may have migrated to shrink the α5 grain as accompanied by the γ5 grain growth.

**4.5 Twin formations during α/γ phase transformation**

As mentioned in section 3-2, we found that Σ3 boundaries were sometimes formed at the front of growing γ grains during α to γ phase transformation. Kirch, et al. have also reported twin formation during the α to γ phase transformation in low carbon steel.7) Figure 5 presents (111) pole figure showing the trace of the Σ3 boundary (γ1/γ6 boundary shown in Fig. 2(g)) on the surface and the great circle including possible directions of the plane normal to the Σ3 boundary. It is found that the coincident (111) poles for two adjoining grains across the Σ3 boundary appears on the great circle with an experimental error. This finding indicates that the grain boundary plane of the Σ3 boundary is parallel to the [111] orientation, that is, the Σ3 boundary is the coherent twin boundary. We further recognized that Σ3 boundaries formed during α/γ interphase boundary migration were also twin boundaries. From these results, therefore, such twin boundaries are likely to be formed by a two dimensional nucleation process on the [111] plane of the growing γ grain, as Gleiter proposed.14)

**5. Conclusions**

The in-situ SEM/EBSD technique was applied to observe the α → γ phase transformation in Fe-9.5 at% Ni alloy, and the nature of the transformation was examined on the basis of crystallographic data obtained by EBSD, particularly concentrating to the influence of grain boundaries and triple junctions on the α → γ phase transformation. The chief results obtained are as follows.
Grain boundaries and grain boundary triple junctions acted as preferential sites for γ phase nucleation.

Random grain boundaries were the most preferred nucleation sites.

The effectiveness of triple junctions as the γ phase nucleation site in the phase transformation varied depending on the triple junction character: the effectiveness increased with increasing the number of random boundaries interconnected at a triple junction.

The γ allotriomorphs often had a K-S or N-W OR with the α parent grains.

The intragrain γ phase having K-S OR with the α parent phase grew in an anisotropic manner. The segment of the interphase boundary with a tilt part of the \( f_{110}^g/C_{111}^g = f_{111}^g/C_{13}^g \) plane matching interphase boundary could migrate preferentially, while the interphase boundary with a twist part of the \( f_{110}^g/C_{111}^g = f_{111}^g/C_{13}^g \) interphase boundary migrated with more difficulty.

Twin boundaries were often formed in the growing γ grains during phase transformation, probably by a two dimensional nucleation process on the \{111\} plane of the growing γ grain.

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