Development of In-situ Orientation Mapping and Microstructure Observation System for Ferrite/Austenite and Martensitic Transformations in Steel

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The unique in-situ SEM/EBSD observation system, which is available to carry out orientation mapping and microstructure observation at high temperature, was developed to observe ferrite/austenite and martensitic transformation of carbon steel directly. The system has the heating capacity over 1 000°C and rapid cooling ability of the cooling rate faster than 100°C/s. The SIM imaging shows very good performance to observe quick change in crystalline grain’s shape, and microstructural evolution of secondary recrystallization grains was successfully observed. Furthermore, the in-situ observation system was applied to observe martensitic transformation of plain carbon steel and nickel contained carbon steel. Crystalline grain shape and its orientation before and after martensitic transformation of the steel are observed at the same sample position. And direct comparison of the microstructures between austenite and martensite is successfully realized.

KEY WORDS: in-situ observation; SEM; SIM; EBSD; phase transformation; martensite.

1. Introduction

In the development of steel products, metallographic structure characterization provides very important information of material structures such as crystalline grain shape, texture, crystal orientation, and grain boundary character. These allow understanding mechanical and chemical properties of the products. Since steel products show phase transformation between austenite which has face centered cubic (fcc) and ferrite which has body centered cubic (bcc) at around 800°C to 900°C, it is important to understand an effect of the transformation on microstructure evolution during heat treatment in manufacturer process. It is well known that microstructure of the prior austenite before phase transformation greatly influences microstructure evolution of crystalline phases such as ferrite, bainite, and martensite in the steel products.

A transmission electron microscope (TEM) and a scanning electron microscope (SEM) are powerful tools to obtain micro- and nano-scale information of internal structure in the steel products, and have been widely used until now. “In-situ observation” with a TEM1–5) and a SEM6–10) under heat treatment has been developed to observe microstructural evolution. Specially, combination between SEM observation and electron back scatter electron diffraction (EBSD) is available to provide microstructure information such as crystalline grain’s phase and its orientation,11) therefore, in-situ observation based on SEM/EBSD measurement during heat treatment recently has been known as very popular technique12–23).

Since bainitic and martensitic transformation are occurred by rapid cooling of austenite such as oil quench and water quench, a rapid cooling system is required to directly observe the above transformation in an in-situ SEM/EBSD observation system. However, it is difficult to cool down a sample quickly because the sample is placed in a vacuum chamber of the system.

A unique in-situ SEM/EBSD observation system has been developed in this study. The system can be applied to orientation mapping and microstructure observation at higher temperature than that of ferrite/austenite transformation of steel materials (~900°C). The system equips a SEM, a focused ion beam (FIB) device, and an EBSD detector to analyze orientation of crystalline grains. Furthermore, the system equips a special laser heating and quick cooling system to observe phase transformation and change in grain morphology during heating and cooling. In this paper, basic configurations of the in-situ SEM/EBSD observation system are presented. And then, in-situ observation of martensitic transformation of carbon steel, an application of the system, is presented.

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2. The In-situ SEM/EBSD Observation System under High Temperature

A commercially available SEM/FIB instrument was used as a base machine for developing the in-situ SEM/EBSD observation system. Figure 1 shows a schematic illustration of the system. The system consists of a field-emission electron-beam gun, a gallium ion-beam gun, an EBSD detector, and the special heating stage. Primary specifications are summarized in Table 1. The electron-beam is used for obtaining SEM images and EBSD maps. The ion-beam is mainly used for scanning ion microscope (SIM) images. The SIM images taken by channeling contrast imaging technique provide the shape of crystalline grains. The ion-beam gun is installed with tilting angle of 70° from the electron-beam gun. The heating stage is directly mounted on the standard sample stage of the SEM/FIB instrument. On the heating stage, a sample is also pre-tilted by 70° from the horizon in order to acquire EBSD maps without tilting the standard sample stage. Therefore, EBSD maps and SIM images can be acquired alternately at the same position without moving and tilting the sample. A sample is set on a special sample holder, and the sample holder is mounted on the heating stage. The sample is irradiated from backside of the sample holder by high power laser beam which can heat it quickly. The available heating temperature range is from room temperature up to 1000°C and the heating speed faster than 100°C/s is also available. An acceptable size of the sample is up to 10 mm × 10 mm and an appropriate thickness is thinner than 1 mm. A thermo-couple and a pyrometer are used to measure and to control temperature of the sample. The pyrometer, which can quickly respond to variation of temperature, is mainly used to control the temperature. When the temperature is lower than 325°C, the thermo-couple is used to control instead of the pyrometer because the pyrometer does not work at the temperature. In order to cool down the sample as quickly as possible, this heating stage has the special cooling system in which a metal block contacts on the sample holder from back side face and remove heat of the sample. The cooling speed faster than 100°C/s is available.

3. Experimental Procedure

A pure iron (99.9%) plate was used to demonstrate heating performance of the special heating stage. Two kinds of carbon steel samples of plain carbon steel, hereafter referred to as “Fe–C steel”, and nickel contained carbon steel, hereafter referred to as “Fe–C–Ni steel” were used to observe martensitic transformation. The detailed chemical compositions of the carbon steel samples are shown in Table 2. The sample plates were cut to be 3 mm × 3 mm × 0.5 mm, and were mounted on the sample holder with a silver paste. SIM images were taken under ion beam current of 50 pA. EBSD maps were taken under electron beam current of 100 nA. Observed area is 280 μm × 500 μm. In case of coarse crystalline grains such as austenite grains, an EBSD map was taken by scanning step size of 1.5 μm or 3.0 μm within 3 minutes. In case of fine crystalline grains such as martensite grains, an EBSD map was taken by scanning step size of 0.5 μm.

Obtained data were processed by software “OIM Analysis” produced by EDAX Inc. to obtain inverse pole figure (IPF) maps and pole figures. Numerical calculation software developed by one of the authors24) was applied to reconstruct orientation of prior austenite by analyzing the relationship among variants of the martensite corresponding to the prior austenite. In the software, orientations of prior austenite are

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**Table 1.** Primary specification of the in-situ observation system.

<table>
<thead>
<tr>
<th>SEM</th>
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<tbody>
<tr>
<td>High tension range</td>
<td>0.1 – 30 kV</td>
<td></td>
</tr>
<tr>
<td>Beam current range</td>
<td>4 pA – 100 nA</td>
<td></td>
</tr>
<tr>
<td>Spatial resolution</td>
<td>2.5nm@1kV, 1.2nm@15kV</td>
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<table>
<thead>
<tr>
<th>SIM</th>
<th></th>
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<tbody>
<tr>
<td>High tension range</td>
<td>1 – 30 kV</td>
<td></td>
</tr>
<tr>
<td>Beam current range</td>
<td>1 pA – 50 nA</td>
<td></td>
</tr>
<tr>
<td>Spatial resolution</td>
<td>5nm@30kV, 1 pA</td>
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</table>

<table>
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<tr>
<th>EBSD</th>
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<tbody>
<tr>
<td>CCD</td>
<td>640 × 480 pixels</td>
<td></td>
</tr>
<tr>
<td>Solid angle</td>
<td>&gt;70°</td>
<td></td>
</tr>
<tr>
<td>Acquiring speed</td>
<td>500 points/s</td>
<td></td>
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</table>

<table>
<thead>
<tr>
<th>Heating Stage</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>UVH laser</td>
<td>938 nm, 100 W</td>
<td></td>
</tr>
<tr>
<td>Sample size</td>
<td>10 mm × 10 mm × 1 mm (max.)</td>
<td></td>
</tr>
<tr>
<td>Temperature (Temp. control)</td>
<td>25°C – 100°C (Pyrometer)</td>
<td></td>
</tr>
<tr>
<td>Heating speed</td>
<td>–100°C/s</td>
<td></td>
</tr>
<tr>
<td>Cooling speed</td>
<td>–100°C/s</td>
<td></td>
</tr>
</tbody>
</table>

**Table 2.** Chemical composition of carbon steel plates (mass%).

<table>
<thead>
<tr>
<th>Sample</th>
<th>C</th>
<th>Si</th>
<th>Al</th>
<th>Mn</th>
<th>Ni</th>
<th>N</th>
<th>Ceq</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe–C steel</td>
<td>0.12</td>
<td>0.005</td>
<td>0.024</td>
<td>–</td>
<td>–</td>
<td>0.0008</td>
<td>0.12</td>
</tr>
<tr>
<td>Fe–C–Ni steel</td>
<td>0.12</td>
<td>0.005</td>
<td>0.023</td>
<td>–</td>
<td>1.99</td>
<td>0.0007</td>
<td>0.17</td>
</tr>
</tbody>
</table>

Ceq=+%C++%Si/24+%Ni/40
calculated by the specific orientation relationship such as the Kurdjumow-Sachs (K-S) relationship between austenite and martensite. The method of Humbert et al. is expanded and modified to increase accuracy of judgments whether the martensite variants belong to the common austenite.

4. Results and Discussion

4.1. Demonstration of the In-situ SEM/EBSD Observation

As a demonstration of performance of the in-situ SEM/EBSD observation system, in-situ observation of the phase transformation from ferrite phase to austenite phase in pure iron was carried out. Figure 2 shows IPF maps showing microstructure evolution of the pure iron plate during heating from room temperature to 900°C. The pure iron plate had primary recrystallized grains of around 50 μm in diameter at room temperature. With the sample temperature increasing, no remarkable change was observed until 696°C. At 800°C, large ferrite grains were observed instead of the primary recrystallized grains. This means secondary recrystallization occurred between 700°C and 800°C. The secondary recrystallization proceeded too fast to dynamically observe the growth of the secondary recrystallized grain by EBSD. SIM imaging was applied to observe growing behavior of the secondary recrystallized grains because SIM images are recorded as a movie at high frame rate. Figure 3 shows SIM images captured from a movie taken at the temperature between 700°C and 800°C. The growth of the secondary recrystallized grain was successfully observed as changes in contrast of the grains. The grain boundary between the primary recrystallized grains and the secondary recrystallized grain does not show simple round shape. The growth behavior of the secondary recrystallized grain suggests that there is preferred orientation relationship between the primary recrystallized grains and the secondary recrystallized grain so as to enhance grain growth.

Figure 4 shows IPF maps during heating from 900°C to 950°C. A phase transformation from ferrite to austenite started at 912°C. The austenite grains were growing with the sample temperature increasing, and the transformation completely finished at 922°C. During the transformation, the secondary recrystallized grains of ferrite were still growing while the austenite grains were growing. The growing points are indicated by the black arrows in Fig. 4(b). The austenite grains mainly appeared on the grain boundary which has misorientation angle larger than 10°.

Thus, the developed system is very powerful to directly observe the growth of crystalline grains during secondary recrystallization and phase transformation at high temperature, and it presents important information to understand their growth behavior.

4.2. Observation of Martensitic Transformation of Carbon Steel

The Fe–C sample and the Fe–C–Ni sample are heated at the temperature higher than their austenite transformation temperature, and then they were rapidly cooled down to room temperature. Figure 5 shows variation of sample temperature for each sample. Both the samples were cooled to 300°C within 5 s after the cooling started, and the cooling rate was faster than 100°C/s.

Fig. 3. Time resolved microstructure evolution of secondary recrystallized grain (S.R.G) during between 700°C and 800°C taken by SIM.

Fig. 2. Microstructure evolutions during heating, (a) at room temperature, (b) at 696°C, (c) at 800°C and (d) at 902°C. Secondary recrystallization of ferrite crystallite grains occurred between 700°C and 800°C. (Online version in color.)
Figures 6(a) and 6(b) show IPF maps of the Fe–C steel at 890°C and after the rapid cooling to room temperature, respectively. Figures 7(a) and 7(b) show IPF maps of the Fe–C–Ni steel at 878°C and after the rapid cooling to room temperature, respectively. The IPF maps were obtained from the same position of the sample before and after the rapid cooling. All of the crystalline grains shown in Figs. 6(a) and 7(a) are austenite phase having fcc structure. The microstructures in Figs. 6(b) and 7(b) have very fine needle-like shape, and suggest that martensitic transformation.
occurred during the rapid cooling. In order to confirm if the fine microstructure consists of martensite variants, \{110\} pole figures of the selected areas indicated by “A” and “B” in Fig. 6(b) and by “C” and “D” in Fig. 7(b) are shown by green dots in Figs. 6(d) and 7(d), respectively. The selected areas correspond to the prior austenite grains which had existed before the rapid cooling. Grain boundaries of the prior austenite grains are drawn by black lines in Figs. 6(b) and 7(b). The black dots in the figures indicate orientations of the martensite variants estimated with orientation of the prior austenite grain on the basis of the K-S relationship. The red dots in the figure are \{111\} pole figures of the prior austenite. The green dots gather around the black dots, and show characteristic patterns similar to the black dots. These results conclude that the microstructures in Figs. 6(b) and 7(b) have the near K-S relationship with the corresponding prior austenite grains, namely, martensitic transformation successfully occurred in the Fe–C steel and the Fe–C–Ni steel by the rapid cooling of 100°C/s in the in-situ SEM/EBSD observation system.

Since partitioning of the austenite grain into several parts in the initial stage of martensitic transformation\(^2\) brings slight and random inclination of the remaining austenite occurring during the martensitic transformation, the partitioning and the inclination of austenite affect fluctuation in orientation of the martensite variants. The fluctuation is observed as dispersion of the green dots around the black dots in the pole figures. Although the austenite grains inside a sample are surrounded by other crystalline grains and deformation is restricted during martensitic transformation, the austenite grains in a surface have free surfaces, therefore, fluctuation in orientation of the martensite variants seems to be observed more remarkably in the surface than at the inside of the sample. To confirm the surface effect on the fluctuation, EBSD data were obtained from inside position of the sample. Figure 8(a) shows an IPF map obtained from a cross-section of the Fe–C–Ni sample after the rapid cooling. Figure 8(c) shows \{110\} pole figures of the selected areas indicated by “E” and “F” in Fig. 8(a). These observed areas are located 100 \(\mu\)m away from surface of the sample. No remarkable difference is found in the dispersion of the pole figures between the surface and the inside position of the Fe–C–Ni sample. This qualitatively indicates that there is a very small effect of surface on the fluctuation in orientation of the martensite variants. The results in Figs. 8(c) also approximately matched with the black dots which indicate orientation of the variants of martensite estimated with orientation of the prior austenite grain on the basis of the K-S relationship as well as those in Figs. 6(d) and
Here, the orientation of the prior austenite grain was determined under the presumption that \{11\overline{1}\} of the prior austenite grain lie on the red arrowed positions in Fig. 8(c).

Small difference in position between the green dots and the black dots in the pole figures, which is observed in Figs. 6(d) and 7(d), is also observed in Fig. 8(c). Figure 9 shows enlarged views of a part of the pole figures surrounded by rectangular frames in Figs. 6(d), 7(d), and 8(c). The difference in position between the pole figure and the black dots is found in Figs. 9(a), 9(c), and 9(e). The difference in position suggests that the orientation relationship between the martensite variants and the prior austenite deviates from the K-S relationship by a few degrees. Therefore, the orientation of martensite variants should be estimated on the basis of the modified K-S relationship taking the deviations from the K-S relationship at the martensite/austenite interfaces, that is, the misorientation of the close-packed-direction and of the close-packed-plane into account. With the misorientation of the close-packed-direction of 2.0° and that of the close-packed-plane of 1.2°, the black dots match well with the pole figure of the martensite variants in the Fe–C–Ni sample as shown in Figs. 9(d) and 9(f). Slight difference in position between the pole figure of the martensite variants and the black dots still remain in the Fe–C sample as shown in Fig. 9(b).

Based on the above results and discussions, reconstructed austenite grains were estimated with orientation of the corresponding martensitic grains on the basis of the modified K-S relationship. Figures 6(c), 7(c), and 8(b) show IPF maps of reconstructed austenite grains estimated with orientation of the corresponding martensitic grains. Grain boundaries of the prior austenite grains are indicated by black lines in the figures. Shapes and orientations of the reconstructed austenite grains approximately match with those of the prior austenite grains for large grains even though small differences are found in the detail of their grain boundaries. It is, however, difficult to reproduce small grains and twin boundary. Furthermore, fine structures corresponding to shape of the martensite variants are observed in the reconstructed austenite grain, and fluctuation of orientation in the reconstructed austenite grain is suggested nevertheless the prior austenite grain has uniform orientation. To evaluate the

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**Fig. 9.** Enlarged views of \{110\} the pole figure of (a) and (b) the Fe–C steel, (c) and (d) the Fe–C–Ni steel, (e) and (f) cross-section of the Fe–C–Ni. (a), (c), and (e) are estimated on the basis of the exact K-S relationship. (b), (d), and (f) are estimated on the basis of the modified K-S relationship. (Online version in color.)

**Fig. 10.** Comparison of angle distribution of \{111\} of the reconstructed austenite (a) between the Fe–C sample and the Fe–C–Ni sample, (b) between the surface and the inside position of the Fe–C–Ni sample. (Online version in color.)
fluctuation quantitatively. Distribution of orientation of the reconstructed austenite crystalline plane was measured with a pole figure of the reconstructed austenite. Figures 6(e), 7(e), and 8(d) show [111] pole figures of the reconstructed austenite in the selected areas indicated by “A” and “B” in Fig. 6(c), by “C” and “D” in Fig. 7(c), and by “E” and “F” in Fig. 8(b), respectively. Orientation of the prior austenite grains corresponding to the selected area is indicated on the pole figure by red dots in Figs. 6(e) and 7(e). The red dots overlap with the maximum of the angle distribution of the reconstructed austenite planes. This means that averaged orientation of the reconstructed austenite exactly agrees with that of the prior austenite. Figure 10(a) shows comparison of angle distribution of {111} of the reconstructed austenite between the Fe–C sample and the Fe–C–Ni sample. The solid lines are the fitting curve by Gaussian function. The full width at half maximum (FWHM) of the distribution function obtained from the Fe–C–Ni sample is 4°. The angle distribution of the Fe–C sample is 1.5 times wider than that of the Fe–C–Ni. Figure 10(b) shows comparison of angle distribution of {111} of the reconstructed austenite between the surface and the inside position of the Fe–C–Ni sample. The FWHM of the distribution function obtained from the surface is almost the same as that from the inside position, and the surface effect is negligible. These results indicate that the partitioning of the austenite grain and the inclination of the remaining austenite mainly affect orientation of the reconstruction of austenite grain. Therefore, the software which takes the effect of the partitioning and the inclination into account is demanded to perfectly reconstruct prior austenite from martensite in addition to the deviations from the K-S relationship at the martensite/austenite interfaces.

5. Conclusions

The in-situ SEM/EBSD observation system developed in this study shows the excellent performance, heating capacity over 900°C and rapid cooling ability of the cooling rate faster than 100°C/s, to observe microstructural evolution during recrystallization, phase transformation between ferrite and austenite, and martensitic transformation. The system has ability to directly observe ferrite to austenite transformation of pure iron and to obtain crystal orientation change during the transformation. The SIM imaging shows very good performance to observe quick change in crystalline grain’s shape, and microstructural evolution of secondary recrystallization grains was successfully observed. Furthermore, the in-situ observation system was applied to observe martensitic transformation of plain carbon steel and nickel contained carbon steel, and then the following results were successfully obtained.

(1) Crystalline grain shape and its orientation before and after martensitic transformation are observed at the same sample position, and direct comparison of the microstructures between austenite and martensite is realized by the developed system.

(2) The orientation relationship between the martensite variants and the prior austenite deviate from the K-S relationship by a few degrees. In the reconstruction of austenite from the corresponding martensite variants, the deviations from the K-S relationship at the martensite/austenite interfaces should be taken into account, for example, the mis-orientation of the close-packed-direction of 2.0° and of the close-packed-plane of 1.2° for the Fe–C–Ni sample.

(3) Not only the deviations from the K-S relationship but the partitioning of the austenite grain and slight and random inclination of the remaining austenite during the martensite should be taken into account in order to perfectly reconstruct prior austenite from martensite.

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