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In situ determination of misorientation angle of grain boundary by field ion microscopy analysis

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ABSTRACT

We proposed an advanced analysis technique for characterizing a grain boundary using field ion microscopy (FIM) for atom probe analysis. The technique enables quick and precise estimation of the misorientation angle of the grain boundary by matching the calculated crystallographic pole positions with the actual FIM image including the grain boundary. We investigated the accuracy in estimation of the misorientation angle using target grain boundaries which had been analyzed by electron backscatter diffraction pattern (EBSD) analysis. From the comparison between EBSD and FIM analyses, we found that the technique enables the determination of the misorientation angle with a high accuracy of $\pm 0.4^{\circ}$, which is comparable with that achieved by EBSD.

1. Introduction

Atom probe tomography (APT) is widely used for the quantitative observation of elements segregating at grain boundaries in materials, because atom probe can detect all elements from light to heavy with very high spatial resolution of atomic lattice size and sufficiently low detection limit of 10 at ppm level [1,2]. In particular, the application of steel materials requires to quantitatively observe light elements, such as hydrogen, boron, carbon, and nitrogen, that strongly influence the microstructure formation and mechanical property of steels. For example, boron and carbon segregation at the grain boundary decreases the ductility-embrittlement transition temperature (DBTT), while sulfur and phosphor increase it [3,4]. It should be noted that the amounts segregating at the grain boundary strongly depend on the character of the grain boundary in addition to the thermal history. The grain boundary character is described by five macroscopic degrees of freedom; a unit rotation vector (rotation direction), the rotation angle (misorientation angle) and the unit normal to the plane [5]. The misorientation angle of the grain boundary is the most important parameter to discuss the segregation and precipitation at the grain boundary. Therefore, the grain boundary in the needle tip must be characterized before atom probe measurements.

Recently, analysis of 3D reconstruction data with the grain boundary using the five macroscopic degrees of freedom was attempted by electron backscatter diffraction (EBSD) analysis [6]. However, in most studies using atom probe, characterization of the observed grain boundaries has not been sufficiently performed because it needed much efforts [7]. In a few studies, it is reported that the grain boundary in the needle tip is characterized using Kikuchi pattern analysis with transmission electron microscope (TEM) before the atom probe measurement [8,9]. It is an appropriate method for grain boundary characterization, but requires much time and a high technique to estimate the misorientation angle. It is also reported as the method for characterizing the grain boundary from a field ion microscopic (FIM) image [10–13]; however, it cannot be applied easily and further cannot obtain the result with sufficient accuracy of less than $\pm 1^{\circ}$. Recently, characterizations of grain boundary orientation using special distribution maps (SDMs) and 3D Hough transformation have been proposed in applications using local electrode atom probe (LEAP) [14,15]. However, the precision of these methods is limited primarily by the accuracy of APT reconstruction, which deteriorates by the uncertainty of the reconstruction parameters (detection efficiency, compression factor and geometric factor) based on a reconstruction protocol [1,16]. Further, these methods cannot be applied before atom probe measurement. In the grain boundary analysis, a quicker (in situ), easier, and more accurate technique before the atom probe measurement is required for long time.

We propose an advanced technique for characterizing the grain boundary just before the atom probe measurement by using FIM. We have already applied this technique to the atom probe analysis of grain boundaries in ferritic steels [17]. However, the accuracy has not been quantitatively investigated. In this paper, the quantitative accuracy in the determination of the misorientation angle of the grain boundary is investigated using target grain boundaries,

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which had been characterized by electron backscatter diffraction pattern (EBSD) analysis.

2. FIM analysis technique for grain boundary characterization

2.1. FIM projections

Newman et al. reported that linear projection is a better approximate than geometric, stereographic, and orthographic projections to explain the positions of crystallographic poles in FIM images [18–21]. Fig. 1(a) shows the diagram of the FIM projections [19]. In the study, it is assumed that the center of the trajectory of ions **O′** is fixed at one point on the straight line that connects the center of FIM screen **O″** and that of the hemispherical cap of the needle tip **O**, where the distance between the hemispherical cap center **O** and trajectory center **O′** is x and the radius of curvature of the hemisphere cap is r. The geometric and stereographic projections correspond to the cases of $x=0$ and $x=r$, respectively [18–20]. In this figure, the point **P** on the tip surface is projected to the point **Q** on the screen. Linear projection is described by the formula $L=k\theta$, where k is constant and L is the distance between the screen center **O″** and projected point **Q** on the FIM screen. It should be noted that the trajectory center is not exactly fixed at one point for linear projection. Fig. 1(b) shows the relation between the distance L on the screen and the central angle θ under various positions of the trajectory center, where the projection factor is defined as x/r. To compare these projections in terms of linearity, the distance of the vertical axis is normalized by the slope at $\theta=0$, namely, $\partial L/\partial\theta|_{\theta=0}$ in figure. In case the factor x/r is about 1.8, the relation is mostly approximate to the linear projection since the relation is given as a straight line in the angle range ($<70°$). Calculated lines are deviated upward from the linear relation for projection factors smaller than 1.8, and downward for larger projection factors. Pole positions on the FIM image can be adjusted by the scale factor as well as the projection factor. The scale factor controls the magnification of the FIM image, which is related to the distance between the screen and needle tip.

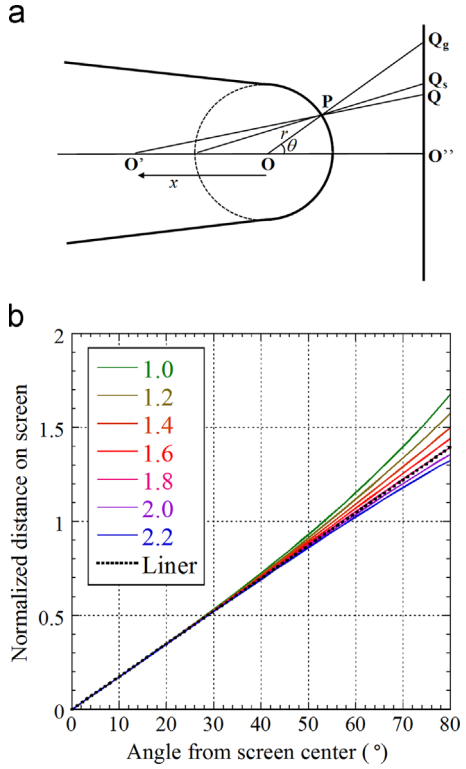

Fig. 1. (a) Schematic of FIM projections. **P** on the tip surface is projected at **Q** on the screen. (b) Relationship between central angle θ and normalized distance from the screen center L, calculated using various projection factors of x/r.

2.2. Pole-fitting method

Fig. 2 shows the diagram of the coordinate system applied in this study. X, Y, and Z are the axes of the coordinate system fixed on the needle tip (the origin **O**). X', Y', and Z' are the axes of the crystalline coordinate system (the origin **O**), which correspond to the [100], [010], and [001] axes in the bcc crystal. The Z-axis of the fixed axes is parallel to the direction of the needle specimen, and the X- and Y-axes are parallel to the directions of X''- and Y''-axes on the screen (the origin **O** ″), respectively. The misorientation angle between the two grains is estimated using the FIM image by the following three steps.

The first step is to determine the direction matrix **A** of the first grain, which corresponds to the rotation from the fixed axes (X,Y,Z) on the needle tip to the crystal axes (X',Y',Z') of the first grain. The picture of the actually observed FIM image and the positions of crystallographic poles calculated by the geometrical configuration shown in Fig. 1 are simultaneously displayed on a PC monitor. The pole positions on the monitor are determined by five parameters, namely three Euler angles (α,β,γ), the ion trajectory center **O′**, and the screen center **O″**. The screen center **O′** is related to the scale factor, while the position of **O″** is related to the projection factor. These parameters can be estimated by matching the calculated positions of plural crystallographic poles with the actual FIM image. The parameters should be determined using at least three poles because a pole position has two information contents.

The second step is to determine the direction matrix **B** about the other grain constructing the grain boundary in the same way as the first one.

The third step is to estimate the misorientation angle of the grain boundary [6]. The direction matrices of the two grains are superimposed through rotation. The rotation matrix is given by

$$\mathbf{M}=\mathbf{B}\mathbf{A}^{-1}, \tag{1}$$

Considering that a cubic crystal has three equivalent axes, the rotation matrix between two grains has 24 combinations. Transformation matrix of the coordinate axes is $\mathbf{R}_j$. The 24 way rotation matrix $\mathbf{M}_j$ is obtained by

$$\mathbf{M}_j=\mathbf{R}_j\mathbf{B}\mathbf{A}^{-1}, \tag{2}$$

where j is the integer number from 1 to 24. The matrix elements of the rotation matrix are assumed as

$$\mathbf{M}_j=\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix}. \tag{3}$$

The rotation angle and rotation axis are obtained by

$$\varphi_j=\cos^{-1}\left(\frac{a_{11}+a_{22}+a_{33}-1}{2}\right) \tag{4}$$

and

$$\mathbf{l}_j=[l_{j\,1},l_{j\,2},l_{j\,3}]=\left[\frac{a_{23}-a_{12}}{2\sin\varphi_j},\frac{a_{31}-a_{13}}{2\sin\varphi_j},\frac{a_{12}-a_{21}}{2\sin\varphi_j}\right], \tag{5}$$

respectively. The misorientation angle of the grain boundary is defined as their minimum angle.

Fig. 3 shows the result of fitting the calculated pole figure to the actual FIM image (8 kV, 70 K, Ne) in ferritic steel. The FIM image shows definite crystallographic poles of the bcc crystal and has the wide view angle corresponding to the full angle of about 120°. The red square marks in the figure represent the 002 pole family, the yellow circle marks represent the 011 family, the green diamond