Using synchrotron radiation XRD technique to investigate active slip systems during tensile deformation in pure magnesium

Masahiro Hirata, Daisuke Okai, Hiroki Adachi

University of Hyogo, Himeji, Hyogo, Japan

em20j003@steng.u-hyogo.ac.jp

Abstract. Active slip systems in pure magnesium specimens with different grain sizes were investigated by the in-situ X-ray diffraction (XRD) technique during tensile testing. From the XRD measurement results, the inhomogeneous strain anisotropy formed by dislocations was obtained. Inhomogeneous strain anisotropy differs in each slip system because hexagonal close-packed metals such as magnesium have anisotropic elasticity. In this study, the dislocation amounts belonging to each active slip system were evaluated utilizing this difference. Coarse-grained specimens had isotropic inhomogeneous strain corresponding to basal slip. Fine-grained specimens showed high elongation and had anisotropic inhomogeneous strain. It is considered that the improvement in ductility occurred by non-basal slip activation near grain boundaries, which increased with grain refinement. Similarly, the ratio of non-basal slip evaluated from the crystal rotation axis increased in the fine-grained specimens.

1. Introduction

Magnesium and magnesium alloys are some of the lightest metal materials in practical use and are used in a variety of applications. However, they have low ductility caused by their low plastic deformability, which is a disadvantage in practical use. In von Mises' criterion [1], five independent slip systems are required to freely deform each grain in polycrystalline materials. Magnesium has a hexagonal close-packed (HCP) structure and basal slip system as the main slip system, which is less than the required five independent slip systems and leads to the observed low ductility. Therefore, since the activation of non-basal slip systems such as prismatic slip and pyramidal slip can increase the ductility, it is important to evaluate the non-basal slips. The critical resolved shear stress (CRSS) of the basal slip in pure magnesium single crystals at room temperature has been reported to be approximately 0.5 MPa [2]. On the other hand, the CRSS of non-basal slips is far larger than that of the basal slip, and the activation of the non-basal slips rarely occurs. However, the activation of the non-basal slips has been reported under several conditions, such as in samples with grain refinement [3, 4] and those undergoing deformation at higher temperature [5]. The identification of active slip systems has been mainly conducted by transmission electron microscopy (TEM). Although the Burgers vector can be identified relatively easily by this method, it is difficult to evaluate active slip systems from the results of many grains because the identification of the slip plane requires time and effort.

Due to its HCP structure, magnesium has different values of inhomogeneous strain anisotropy for each slip system [6]. It was speculated that qualitative evaluation of the dislocation amount belonging to each active slip system is possible by comparing the calculated and measured values of inhomogeneous strain anisotropy using these differences. Here, "inhomogeneous strain anisotropy"
means a change in the size of the inhomogeneous strain field introduced by dislocations existing in the crystal depending on the crystal orientation. Therefore, in this study, we attempted to qualitatively evaluate the dislocation amount belonging to each active slip system by obtaining the inhomogeneous strain anisotropy, using the results of in-situ X-ray diffraction (XRD) measurements during tensile deformation of pure magnesium with different grain sizes and comparing the results with the calculated values.

2. Experimental procedures

Samples with a diameter of 30 mm and a thickness of 1 mm were prepared from a 99.9% pure magnesium ingot for high pressure torsion (HPT) processing [7, 8]. The rotation anvil was turned 7 times at a speed of 0.2 rpm under a compressive stress of 2 GPa. The HPTed samples were annealed at 473 K for 7.2, 18 or 36 ks in order to obtain samples with several grain sizes, and then tensile specimens having a parallel length of 10 mm, a width of 3 mm and a thickness of 0.5 mm were prepared by a wire-electrical discharge machine. The average grain sizes of the HPTed sample and annealed samples were measured by electron backscatter diffraction (EBSD) measurements at an acceleration voltage of 15 kV using an orientation imaging microscopy (OIM) system (TSL Solutions) with a scanning electron microscope (JSM-6500F, JEOL). The average grain size, d, was defined as the interval between large-angle grain boundaries of a misorientation angle >15°. In the HPTed sample, d=2.1 µm, and in the samples annealed at 473 K for 7.2, 18 or 36 ks, d=4.8, 6.4, and 8.3 µm, respectively. In this paper, these samples are called the 2.1, 4.8, 6.4, and 8.3 µm samples, respectively.

In-situ XRD measurement during tensile deformation was conducted using synchrotron radiation (SPring-8) [9]. The energy of the incident X-ray was 30 keV, and the beam size was 0.5 mm long and 0.2 mm wide. A 6-unit MYTHEN detector was positioned 22° above the transmission direction and the camera length was 730.2 mm in order to measure diffraction peaks over a large range at the same time. The tensile test was conducted with an initial strain rate of 3.3×10⁻⁴ s⁻¹ while oscillating the tensile tester in the Z direction at a speed of 1 mm/s and a width of 2 mm, because it is necessary to increase the number of grains in the X-ray irradiation volume. The measurement was performed with a time resolution of 3 s. The diffraction peak angle and full width at half maximum were calculated from the observed diffraction peak profiles from the (100), (002), (101), (102), (110), (103), (200), (112), (201), (004), (202), (104), (203), (211), (114), (212), (105), (204), (213), and (302) diffractions.

Dragomir et al. calculated the inhomogeneous strain anisotropy introduced by the dislocations belonging to each slip system under the assumption that it is a continuous elastic body and only a basal, prismatic or pyramidal slip system exists, respectively [6, 10]. Figure 1 shows the calculated inhomogeneous strain anisotropy of the basal <a> slip: {0001}{2110}, the prismatic <a> slip: {1010}{2110} and the pyramidal <c+a> slip: {2112}{2113} when a density of dislocations belonging to each slip system is 10¹⁴ m⁻². This figure is represented in polar coordinates, the argument is the angle between the basal plane and the direction of inhomogeneous strain, the radius of the curve is the amount of inhomogeneous strain, the vertical axis shows the amount of inhomogeneous strain along the c-axis direction and the horizontal axis shows the amount of inhomogeneous strain along the in-plane direction of the basal plane. The dislocations belonging to different slip systems have different values of inhomogeneous strain anisotropy. For example, the dislocations of the basal <a> slip have a relatively isotropic inhomogeneous strain, the dislocations of the prismatic <a> slip have a 1.2-fold larger inhomogeneous strain anisotropy toward the in-plane direction than that of the basal <a> slip, and the dislocations of the pyramidal <c+a> slip have a 3-fold larger inhomogeneous strain anisotropy toward the c-axis direction than that of the basal <a> slip. Qualitative evaluation of the dislocation amount belonging to the active slip systems was performed by comparing the inhomogeneous strain anisotropy with the measured values with Figure 1.

The crystallite size and inhomogeneous strain are the two main sources contributing to XRD peak broadening. For HCP metals with elastic anisotropy, separation by the Williamson-Hall method [11], which is valid for the analysis of isotropic materials, is not appropriate. Therefore, the modified
Williamson-Hall method [12] was used to separate the two effects [13-15]. The modified Williamson-Hall equation is the following,

$$\Delta K_{hkl} = \frac{0.9}{D} + \left( \frac{\pi M^2 b^2}{2} \right)^{\frac{1}{2}} \frac{1}{\rho^2} \bar{C}_{hkl} \frac{1}{2} K_{hkl} + O\left( K_{hkl}^2 \bar{C}_{hkl} \right)$$

where $K_{hkl} = \frac{2 \sin \theta}{\lambda}$, $\Delta K_{hkl} = \Delta 2\theta \cos \theta / \lambda$, the subscript $hkl$ indicates data for the diffraction peak obtained from the $hkl$ diffraction, $\theta$ is Bragg diffraction angle, $\Delta 2\theta$ is full width at half maximum (FWHM), $\lambda$ is the wavelength of incident X-rays, $D$ is the crystallite size, $b$ is the size of the Burgers vector, $\rho$ is the dislocation density, $M$ is a constant representing the degree of dislocation arrangement and $\bar{C}_{hkl}$ is the average dislocation contrast factor for the $hkl$ diffraction. $O(K_{hkl}^2 \bar{C}_{hkl})$ represents higher-order terms in $KC$, which usually are small compared to the first two terms. The coefficient part of $K_{hkl}$ is the part corresponding to the inhomogeneous strain ($e_{hkl}$) of the conventional Williamson-Hall equation, and can be expressed as the following equation from (1),

$$e_{hkl} = \sqrt{\left( \frac{\Delta K_{hkl}}{K_{hkl}} - \alpha \right)^2}$$

where $\alpha = 0.9/D$. By calculating the amount of inhomogeneous strain in the $hkl$ diffraction plane normal direction from $hkl$ diffraction peak, the inhomogeneous strain anisotropy was obtained.

EBSD measurement has been used as a method for evaluating active slip systems [16, 17]. Kashihara et al. [16] evaluated an active slip system by determining the axis of crystal rotation in deformation along the [100] axis of an aluminum bicrystal by EBSD measurement and comparing it with the calculated rotation axis. In this study, EBSD measurement was performed at the same field of view for the 2.1 and 6.4 µm samples before and after tensile deformation, and the crystal rotation axes of 20 crystal grains were determined. The angle formed by the measured crystal rotation axis and the crystal rotation axis of the basal slip was investigated, and the ratio of the basal slip and non-basal slip was qualitatively evaluated. The results were compared with the qualitative evaluation results of the active slip systems obtained from the inhomogeneous strain anisotropy.

**Figure 1** The inhomogeneous strain anisotropy of each slip system created from calculated values. This figure is expressed in polar coordinates in arbitrary units (a.u.); the argument is the angle between the basal plane and the direction of inhomogeneous strain, the radius of the curve is the amount of inhomogeneous strain, the vertical axis shows the amount of inhomogeneous strain in the c-axis direction and the horizontal axis shows the amount of inhomogeneous strain in the in-plane direction.

### 3. Results & Discussion

#### 3.1. In-situ XRD measurement

The nominal stress–nominal strain curves of each sample are shown in Figure 2. The total elongation of the 2.1, 4.8, 6.4 and 8.3 µm samples was 34.5, 10.6, 5.1 and 2.9%, respectively, and the uniform elongation values were 10.1, 9.7, 4.1 and 2.4%, respectively. Similar to previous reports on magnesium alloys, the ductility increased for samples with fine crystal grains [3, 4, 18, 19].
Figure 3(a) shows typical in-situ XRD profiles for the 2.1 µm sample before tensile deformation, during elastic deformation and during plastic deformation, with nominal strain of 0, 0.18 and 2.0%, respectively. In order to clearly observe the change in the shape of the diffraction peak, enlarged views of the (110) diffraction peaks are shown in Figure 3(b). Compared with the diffraction peak before the tensile deformation, the diffraction peaks during the deformation were shifted to the low angle side because the interplanar spacing increased. In addition, the FWHM increased during plastic deformation due to the introduction of inhomogeneous strain.

3.2. Qualitative evaluation of the dislocation amount belonging to each active slip system

Figure 4 shows the changes in the inhomogeneous strain anisotropy in the 2.1 (a), 4.8 (b), 6.4 (c) and 8.3 µm (d) samples during tensile deformation. In each sample, the amount of inhomogeneous strain increased with the progress of tensile deformation. This is due to the increase in the dislocation density. The samples showing high ductility with small grain size and uniform elongation of about 10% (Figure 4(a, b)) and the samples with a lower ductility (Figure 4(c, d)) had different shapes for the inhomogeneous strain anisotropy. At 10.1% in Figure 4 (a) and 9.7% in (b), the amount of inhomogeneous strain in the c-axis direction and the in-plane direction of basal plane was 1.5 and 1.7 times larger than the amount of inhomogeneous strain in the 45° direction, respectively. The anisotropy of the inhomogeneous strain was large. On the other hand, at 4.1% in Figure 4 (c) and 2.4% in (d), the difference in the amount of inhomogeneous strain depending on the direction was 1.1 and 1.2 times, respectively, and the inhomogeneous strain was almost isotropic.

The dislocation amount belonging to each active slip system was qualitatively evaluated by comparison with Figure 1. In (a) and (b) of Figure 4, the 2.1 and 4.8 µm samples have a large amount of inhomogeneous strain along the c-axis and in-plane direction of the basal plane. Therefore, it is considered that the prismatic <a> slip and pyramidal <c+a> slip, which cause large inhomogeneous strain along the c-axis and in-plane direction of basal plane, respectively, were activated. Panels (c)
and (d) of Figure 4 show the isotropic inhomogeneous strain of the 6.4 and 8.3 µm samples; this suggests that there is a lot of activity in the basal $<a>$ slip.

Focusing on the change in inhomogeneous strain anisotropy during tensile deformation, the difference in the direction-dependent amount of inhomogeneous strain at nominal strain of 0.25% in Figure 4(a, b) is 1.3 times at the maximum, which is smaller than the value at nominal strain of 10.1% in (a) and 9.7% in (b). This suggests that the basal $<a>$ slip is active in the early stage of plastic deformation, and the ratios of active prismatic $<a>$ slip and pyramidal $<c+a>$ slip increase as the deformation progresses. On the other hand, in Figure 4(c, d), the difference in the amount of direction-dependent inhomogeneous strain was about 1.1 to 1.2 times. Compared with Figure 4(a, b), there was no significant change in the shapes of the inhomogeneous strain anisotropy curves, and it is considered that a lot of basal slip was activated during tensile deformation.

The non-basal slips are considered to be activated by the additional stress generated by their compatibility in the region near the grain boundary, and fine grains occupy a larger proportion of the region near the grain boundary than coarse grains [4, 20]. Therefore, it is considered that the non-basal slips were more active in samples with fine grains. The reason why high ductility was exhibited in the 2.1 and 4.8 µm samples was that the plastic deformability of magnesium was improved by the activation of the non-basal slips.

3.3. Evaluation by crystal rotation

Figure 5 shows the angles formed by the crystal rotation axis obtained by EBSD measurement and the crystal rotation axis of the basal slip in the 2.1 and 6.4 µm samples. The "0.5%" and "1.0%" in Figure 5 are the angles formed by the crystal rotation axis between the condition before tensile deformation and at nominal strains of 0.5% and 1.0%, respectively, and the rotation axis of the basal slip. When the angle is close to 0°, this indicates that the measured rotation axis is close to the rotation axis of the basal slip, which means that the crystal rotation is caused only by the basal slip. The average angle was 52.5° and 55.4° for the 2.1 µm sample and 20.4° and 21.2° for the 6.4 µm sample, respectively, at nominal strains of 0.5% and 1.0%, respectively. Compared with the 6.4 µm sample, the crystal

**Figure 4** The change of inhomogeneous strain anisotropy during tensile deformation in (a) 2.1, (b) 4.8, (c) 6.4, and (d) 8.3 µm samples, shown as % nominal strain.

**Figure 5** The angles formed by the crystal rotation axis obtained by EBSD measurement and the crystal rotation axis of the basal slip in the 2.1 and 6.4 µm samples. The dark gray shows the results of the rotation axis between the condition before tensile deformation and at nominal strain of 0.5%. The light gray shows the corresponding rotation axis at nominal strain of 1.0%.
measurements of inhomogeneous strain anisotropy. Since these results are in agreement with the results obtained by in-situ XRD measurement, it is considered that the evaluation of active slip systems using inhomogeneous strain anisotropy in this study is valid.

4. Conclusions

Active slip systems were qualitatively evaluated from the inhomogeneous strain anisotropy derived from the results of in-situ XRD measurements during tensile deformation of pure magnesium samples with different grain sizes.

(1) Different shapes of inhomogeneous strain anisotropy plots were obtained from samples with different grain sizes.

(2) The anisotropy of the inhomogeneous strain was large in the 2.1 and 4.8 μm samples, suggesting that the prismatic <a> slip and the pyramidal ⟨c+a⟩ slip were active.

(3) The anisotropy of the inhomogeneous strain was small in the 6.4 and 8.3 μm samples, suggesting that the basal <a> slip was active.

(4) As a result of evaluation of the active slip systems based on the crystal rotation, the activation ratio of non-basal slips was higher in the 2.1 μm sample than in the 6.4 μm sample, showing the same tendency as the result obtained from measurements of inhomogeneous strain anisotropy.

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References

[1] Von Mises R and Agnew Z 1928 Math. Mech. 8 161
[2] Numakura H 1998 Materia Jpn. 37 117-124
[3] Kobayashi T, Koike J, Yoshida Y, Kamada S, Suzuki M, Maruyama K and Kojima Y 2003 J. Jpn. Inst. Met. 4 149-152
[4] Zheng R, Bhattacharjee T, Gao S, Gong W, Shibata A, Sassaki T, Hono K and Tsuji N 2019 Sci. Rep. 9 11702
[5] Wu G, Nodooshan H R J, Zeng X, Liu W, Li D and Ding W 2018 metals 8 980
[6] Dragomir I C and Ungár T 2002 J. Appl. Cryst. 35 556-565
[7] Bridgman P W, 1935, Phys. Rev. 48 825
[8] Zhilyaev A P, Langdon T G, 2008 Prog. Mater. Sci. 53 893-979
[9] Adachi H, Mizowaki H, Hirata M, Okai D and Nakanishi H 2020 J. Jpn. Inst. Light Met. 70 274-280
[10] Borbély A, Dragomir J C, Ribárík G and Ungár T 2003 J. Appl. Cryst. 36 160-162
[11] Williamson G K and Hall W H 1953 Acta Metall 1 22-31
[12] Ungár T and Borbély A 1996 Appl. Phys. Lett. 69 3173
[13] Ungár T, Révész Á and Borbély A. 1998 J. Appl. Cryst. 31 554-558
[14] Ungár T, Dragomir I C, Révész Á and Borbély A 1999 J. Appl. Cryst. 32 992-1002
[15] Ungár T, Gubicza J, Ribárík G and Borbély A 2001 J. Appl. Cryst. 34 298-310
[16] Kashihara K and Wert J A, 2006 Mater. Trans. 47 233-238
[17] Kashihara K, 2008 Mater. Trans. 46 419-423
[18] Sakaoka Y, Kuramoto S, Kawabata H and Kurumada A 2019 J. Jpn. Inst. Light Met. 69 332-338
[19] Miura H, Yang X and Sakai T 2008 Mater. Trans. 49 1015-1020
[20] Miyanno H, Takemoto K, Tsushima M, Kitahara H and Ando S 2020 J. Jpn. Inst. Light Met. 70 1117-121