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Application of Inverse Pole Figure to Rietveld Refinement: II. Rietveld Refinement of Tungsten Liner using Neutron Diffraction Data

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The three-dimensional orientation distribution function of a conical shaped tungsten liner prepared by the thermo-mechanical forming process was analyzed by 1.525 Å neutrons to carry out the Rietveld refinement. The pole figure data of three reflections, (110) (200) and (211) were measured. The orientation distribution functions for the normal and radial directions were calculated by the WIMV method. The inverse pole figures of the normal and radial directions were obtained from their orientation distribution functions. The Rietveld refinement was performed with the RIETAN program that was slightly modified for the description of preferred orientation effect. We could successfully do the Rietveld refinement of the strongly textured tungsten liner by applying the pole density of each reflection obtained from the inverse pole figure to the calculated diffraction pattern. The correction method of preferred orientation effect based on the inverse pole figures showed a good improvement over the semi-empirical texture correction based on the direct usage of simple empirical functions.

Key words: Rietveld refinement, Orientation distribution function, Texture, W-liner, Inverse pole figure

I. Introduction

Since the Rietveld method[1] was used for the profile refinement of diffraction data, it has been widely used for crystal structure refinement of polycrystalline samples. In the Rietveld method, the profile and structural parameters are refined with a given structural model. The greatest advantage of this method is obviously related to its high capability of solving problems involving overlapped diffraction lines. The Rietveld method is to refine the crystal structure of polycrystalline samples under the assumption that crystallites have a random orientation distribution.

It, however, often faced with a problem due to the preferred orientation or texture effect of samples, which results from the processes such as compaction, settling or deformation acting on non-isotropic crystallite shapes. If a sample shows texture effect, the peak intensity is not only a function of crystal structure and instrument but also a function of texture, and there is a great deviation between the theoretical and observed intensities.

In order to correct the texture effect of a sample, most of Rietveld programs are incorporated by the semi-empirical texture correction method based on the concept of rigid platelets or needle floating in a viscous medium.[2] Also many researches are adopted in making powder, rotating sample and special sample preparation techniques to reduce the preferred orientation effect. These techniques are useful for correcting the preferred orientation effect for a weakly textured sample, but there is a weak point for the highly textured sample that greatly changes the intensity ratio of reflections in the whole diffraction pattern owing to the strong preferred orientation effect.[3,4]

The texture effect on the diffraction pattern of a unique sample direction is directly proportional to the pole density of the corresponding inverse pole figure that can be obtained from the three-dimensional orientation distribution function (ODF) of the sample. It can be obtained from its pole figure, which is a stereographic projection showing the distribution of poles for a specific crystalline plane using specimen axes as a reference axis. Pole figure data can be obtained with X-ray or neutron diffraction by setting a detector on the center of a diffraction peak for each *hkl* plane, and the sample is tilted and rotated. The observed intensity is proportional to the number of lattice planes in that orientation. To calculate the three-dimensional orientation distribution function of a given sample, several pole figures are required for different *hkl* planes.[5-8]

In this article we are focused on the Rietveld refinement of the highly textured tungsten (W) sample with the pole density for each reflection obtained from the inverse pole figure as a correction parameter for the preferred orientation effect.

II. Experimental Procedure

The sample used in this study was a conical shaped tungsten liner prepared by the thermo-mechanical forming process. It was 12×12×4 mm in size. The specimen was mounted on the four-circle diffractometer. The pole figure data of three reflections, (110) (200) and (211) were mea-

sured over an entire hemisphere with 1.525 Å neutrons at the National Institute Standards and Technology (NIST) research reactor. The ODF's of two different directions (normal direction (ND) and radial direction (RD)) were calculated by the Williams-Imhof-Matthies-Vinel (WIMV) method, using popLA program.[9] Orthorhombic sample symmetry was used for the texture analysis. The inverse pole figures of the ND and RD were obtained from their ODFs.

The neutron diffraction patterns of W sample were measured in the ND and RD over scattering angle 35°~104° using 1.525 Å neutrons. The diffraction data were measured with the fixed orientation mode, θ-2θ mode, at two different sample orientations (ND and RD). The Rietveld refinement was run with the RIETveld ANalysis (RIETAN) program, which was modified for the description of texture. The RIETAN program is a FORTRAN program developed by F. Izumi for the Rietveld refinement for X-ray, neutron, and synchrotron powder diffraction data.[10,11]

III. Results and Discussion

Fig. 1 shows that the observed neutron diffraction patterns of W sample at two different sample orientations (ND and RD), and the calculated one of W polycrystalline sample with an ideal random orientation distribution, respectively. Compared with the calculated pattern of an ideally random oriented sample, the intensity ratio between the observed and calculated patterns was very different. The intensities of 200 and 110 reflections for two different directions (ND and RD) in the observed diffraction patterns were higher than those in the calculated one. This indicates that W sample has the texture or preferred orientation. The direction of preferred orientation is (200) for the ND and (110) for the RD, respectively.

In the case of the refinement procedure without the preferred orientation parameter as a variable parameter, it was not possible to obtain good refined patterns due to the texture as shown in Fig. 2. The diffracted intensity of randomly oriented polycrystalline samples is influenced only by their crystal structure, whereas the intensity of materials with texture is subjected to the crystal structure and the texture effect. The structural information, therefore, cannot be accurately obtained from textured samples by means of the least-squares method, such as Rietveld refinement and pattern decomposition method.

In order to correct the effect of texture in the samples, most of Rietveld programs use some empirical and semi-empirical functions.[2] Among these functions, March-Dollase function based on the concept of rigid platelets or needles in a viscous medium is widely used to correct the preferred orientation effect. It contains one variable parameter and needs to select the preferred orientation direction. The variable parameter, March-Dollase coefficient, represents the effective sample compression or extension due to the preferred orientation.

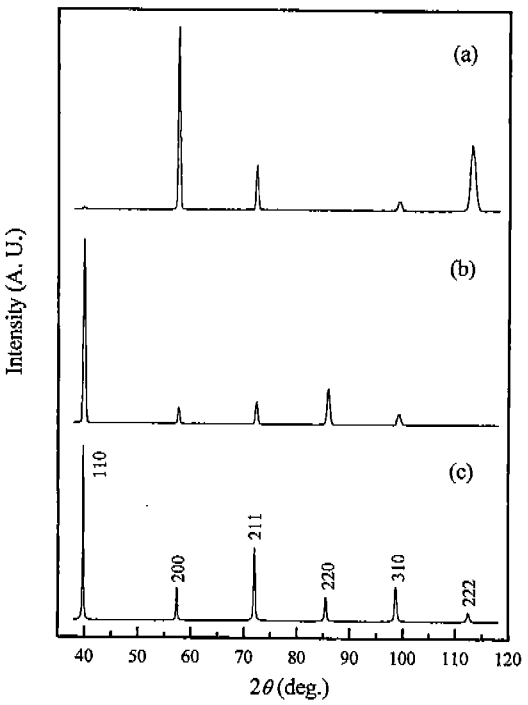

Fig. 1. Neutron diffraction patterns of tungsten: (a) observed neutron diffraction pattern at the normal direction, (b) observed one at the radial direction and (c) calculated one of tungsten polycrystalline sample with an ideal random orientation distribution.

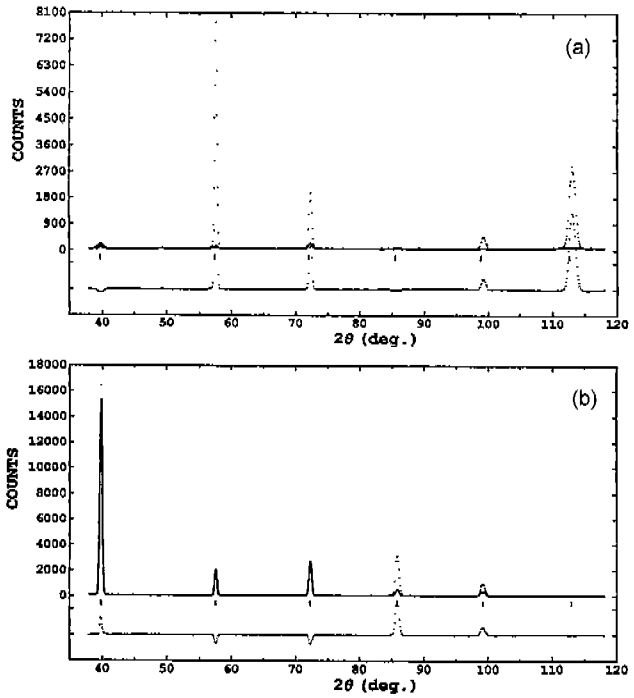

Fig. 2. Rietveld refinement patterns without the preferred orientation parameters at two different sample orientations: (a) normal direction and (b) radial direction. Dots represent the observed intensities, and the solid line is calculated ones. A difference (obs.-cal.) plot is shown beneath. Vertical bars are the reflection position markers.

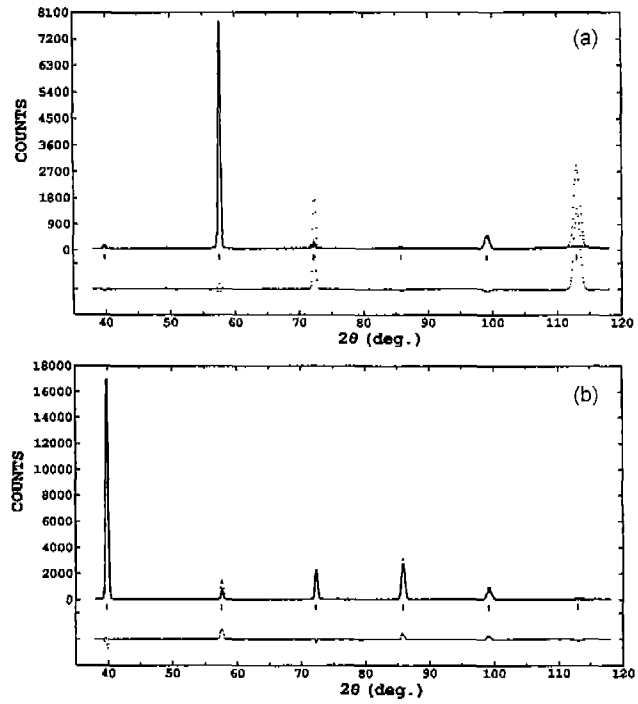

Fig. 3. Rietveld refinement patterns corrected with March-Dollase function at two different sample orientations: (a) normal direction and (b) radial direction. Dots represent the observed intensities, and the solid line is calculated ones. A difference (obs.-cal.) plot is shown beneath. Vertical bars are the reflection position markers.

We selected (200) for the ND and (110) for the RD as a direction of preferred orientation, respectively. The March-Dollase coefficient values for two directions were 0.221 for the ND and 0.389 for the RD, respectively. These values suggest that many crystallites in W sample are oriented to (200) for the ND and (110) for the RD. Fig. 3 shows the Rietveld refinement patterns for two sample orientations (ND and RD). By applying the March-Dollase function to 200 reflection in the ND and 110 reflection in the RD, respectively, the overall agreement between the observed and calculated patterns was not satisfactory. All *R*-factors obtained from the March-Dollase function are listed in Table 2. From these results, we could know that the March-Dollase function as a correction factor of the preferred orientation effect may not be suitable for the sample exhibiting the highly textured effect, and it is necessary for an accurate description of texture effect to get a complete Rietveld refinement pattern for a strongly textured sample. We, therefore, carried out the texture analysis for this sample to describe the preferred orientation accurately.

Generally the texture analysis is the procedure for determining orientation characteristics of a polycrystalline sample from diffraction experiments. A commonly used description of texture is by the three-dimensional orientation distribution function. The ODF analysis requires several diffraction data for each pole distance and azimuthal angle. A projection of the ODF along an axis, inverse pole figure, describes the orientation density of all the *hkl* poles oriented to the particular sample direction (ex. ND or RD). The pole density of a given *hkl* plane in the inverse pole figure of ND is proportional to the volume fraction of all the *hkl* planes oriented to the plane normal parallel to the ND direction.

The pole density is expressed in terms of the multiplies random distribution (m.r.d.) unit by normalizing the observed intensities to the random distribution intensity. The random distribution intensity, *P*, which is equivalent to the powder diffraction intensity, is calculated from the experimental pole figure data:[12]

$$P=\frac{\left\{\sum_{i=1}^{m} R(\chi_i)\cos(\chi_i)+R(90)/n\right\}}{\left\{\sum_{i=1}^{m} n\cos(\chi_i)+1\right\}}$$

where $m=90/\Delta\chi$, $R(\chi_i)=\Sigma F(\varphi_i, \chi_i)$, $n=360/\Delta\varphi$, and $F(\varphi_i, \chi_i)$ is the observed intensity at the goniometer coordinate (φ_i, χ_i). The center of the pole figure corresponds to the hemispheric pole ($\chi=90°$, $\varphi=\varphi$), and the periphery to the equator of the hemisphere ($\chi=0°$, $\varphi=\varphi$).

The pole density for each *hkl* plane in the ND and RD was obtained from their inverse pole figures. Fig. 4 shows the inverse pole figure for the ND and the RD, respectively. Table 1 shows the pole density for each *hkl* plane in the ND and RD, respectively. The pole density value of (200) in ND and (110) in RD was significantly higher than that of other ones, as shown in Table 1. It shows that the strongest texture plane for ND and RD in all reflection planes is (200) and (110), respectively. The value of pole densities for each *hkl* plane is in agreement with the intensity ratio between the observed and calculated intensities as shown in Fig. 1.

Completely to correct the texture effect in the Rietveld refinement for W sample, we used the pole density for each *hkl* plane obtained from the inverse pole figure instead of the semi-empirical function as a preferred orientation parameter. The pole densities for all reflection planes were applied to the calculated diffraction patterns for the ND and the RD. Fig. 5 presents the calculated diffraction patterns corrected by means of the pole densities obtained from the inverse pole figure. The calculated diffraction patterns were very nearly identical with that in the observed ones as shown in Fig. 1. These results suggest that the texture effect correction technique in terms of the inverse pole figure can be suitable for the heavily textured sample.

The initial refinement was conducted by zero point shift and lattice parameters after the correction of texture effect. We added other parameters such as background and thermal parameters. Fig. 6 and Table 2 show the Rietveld refinement patterns and all *R*-factors, respectively. The agreement between observed and calculated patterns was very satisfactory as shown in Fig. 6. Compared with the refinement patterns as shown in Fig. 3, this result shows

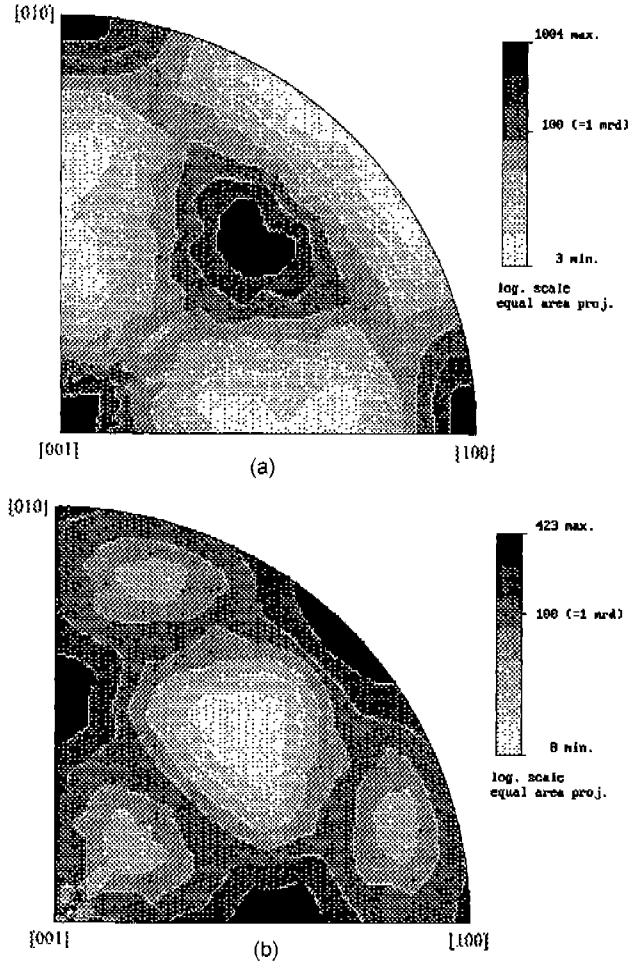

Fig. 4. Inverse pole figures of tungsten: (a) normal direction and (b) radial direction. Equal-area projection; levels are in multiples of random distribution.

Table 1. Pole Densities Obtained from the Inverse Pole Figures of the Normal Direction (ND) and the Radial Direction (RD)

h k l	ND	RD
110	0.04[1)]	4.37[1)]
200	8.42	2.18
211	0.84	0.53
310	0.88	1.40
222	6.35	0.14

[1)]multiplies of random distribution

that the texture effect correction method using pole density for each reflection much better than that using March-Dollase function. Also, all R-factors obtained with the application of the pole density to the diffraction data were far lower than those with the March-Dollase function as listed in Fig. 3. The weight R-factor, R_{wp}, was rapidly decreased from 54.06% to 10.78% for the ND diffraction pattern and 18.39% to 7.83 for one for the RD diffraction pattern, respectively. The profile factor, R_p, which is a true quantity based on the discrepancies between observed and calculated intensities,

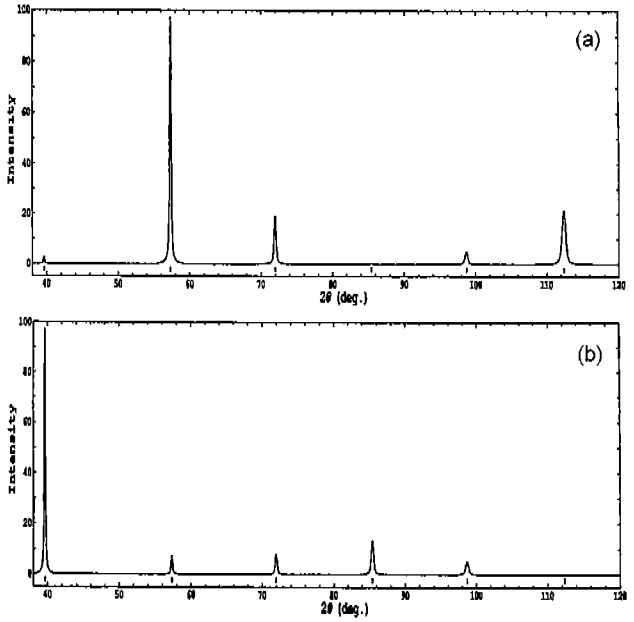

Fig. 5. Neutron diffraction patterns weighted with the pole density obtained from the inverse pole figure: (a) normal direction and (b) radial direction. Vertical bars are the reflection position markers.

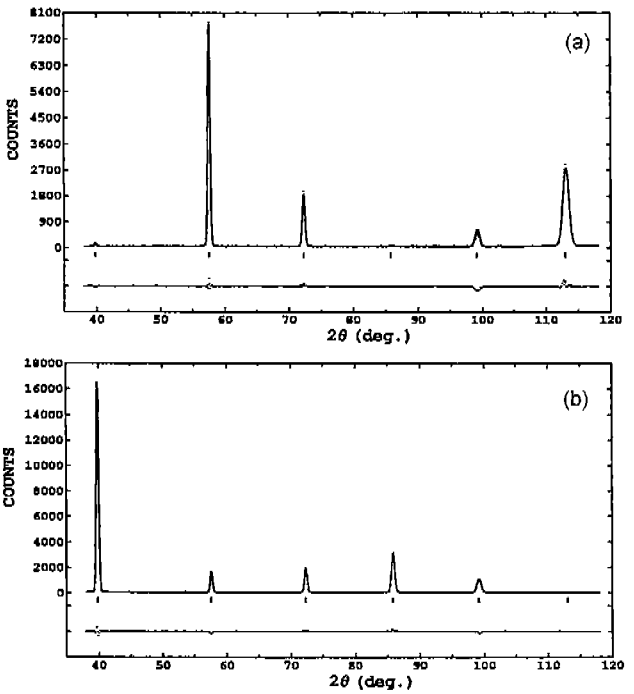

Fig. 6. Rietveld refinement patterns of tungsten using the pole densities obtained from the inverse pole figure: (a) normal direction and (b) radial direction. Dots represent the observed intensities and the solid line is calculated ones. A difference (obs.-calc.) plot is shown beneath. Vertical bars are the reflection position markers.

was far lower than those in the previous refinements (Fig. 3). The goodness-of-fit value, S (=R_{wp}/R_e), was lowered from 7.03 to 1.40 for ND, 2.71 to 1.29 for RD, respectively.

Table 2. *R*-factors Obtained with the Application of March-Dollase Function (A) and the Pole Density (B) to the Observed Neutron Diffraction Data at Two Different Sample Orientations

	R_{wp}	R_P	R_I	R_F	S $(=R_{wp}/R_e)$
A[i)]	54.06%	37.43%	49.48%	42.88%	7.03
B[i)]	10.78%	7.05%	4.17%	3.23%	1.40
A[ii)]	18.39%	10.48%	8.00%	6.90%	2.71
B[ii)]	8.73%	5.82%	2.70%	2.59%	1.29

[i)]Normal direction
[ii)]Radial direction

IV. Conclusion

By applying the pole densities of each reflection obtained from the inverse pole figure to the calculated diffraction pattern to correct the texture effect, we could successfully carry out the Rietveld refinements for the strongly textured W sample. The texture effect correction based on the inverse pole figures can be very useful to do the structure refinement for the highly textured samples, and is a good improvement over the semi-empirical texture correction based on the direct usage of simple empirical functions. Also, this can be extended to the kinds of polycrystalline samples showing the strong texture effect.

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