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Application of Inverse Pole Figure to Rietveld Refinement: I. Rietveld Refinement of Copper Sheet using X-ray Diffraction Data

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Both the X-ray diffraction data of the normal direction in the sample orientation and the pole figure data of three reflections, (111), (200) and (220), were used to do the Rietveld refinement for the copper sheet prepared by a cold rolling process. The agreement between calculated and observed patterns was not satisfactory, which was attributed to the preferred orientation effect of the copper sheet. The Rietveld refinement for the copper sheet could be done successfully by applying the pole density of each reflection obtained from the corresponding inverse pole figure to the X-ray diffraction data of the normal direction. The *R*-weighted pattern, R_{wp}, was 12.99% and the goodness-of-fit indicator, *S*, was 3.68.

Key words: Rietveld refinement, Texture, Preferred orientation, Cu-sheet, Inverse pole figure

I. Introduction

Recently the Rietveld refinement technique is probably the most popular and widely used technique for refining structure and lattice parameters directly from whole X-ray or neutron powder diffraction patterns without separating reflections.[1] It can be applied only for the randomly orientated powder samples, not for the polycrystalline ones with the effect of strong texture. Many kinds of engineering materials, however, are manufactured by rolling processes that produce a preferred orientation or texture effect of each crystal grain. If a polycrystalline sample is textured, the observed intensity of diffraction pattern depends on the texture effect as well as the crystal structure, and can be no longer directly related to structural parameters. If the Rietveld refinement is performed on the diffraction data showing the preferred orientation, it is not possible to obtain a good fitting result because of the preferred orientation or texture effect. The results of structural parameters are also not available.

To do the Rietveld refinement for the textured polycrystalline, the correction procedure for the effect of preferred orientation is applied to data exhibiting the effect of preferred orientation. The spherical harmonic[2,3] and semi-empirical function models[4] are used to solve the problem. In the case of strongly textured samples, the former is not useful for low symmetry system due to a number of free parameters in some cases. The latter needs only one or two free parameters, but it is valid only both for fiber or disk textures and for the Bragg-Brentano diffraction method. In addition to the above models, the texture effect of a sample can be corrected by means of the inverse pole figure obtained from the three-dimensional orientation distribution function of the sample because the effect of texture on the diffraction pattern of a unique sample direction in the sample is directly proportional to the pole densities of the corresponding inverse pole figure.[5,6]

The purpose of this paper is focused on the Rietveld refinement of the cold rolled copper (Cu) sheet 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 diffraction and preferred orientation studies were performed by conventional powder X-ray diffraction, back reflection Laue diffraction, and pole figure analysis using X-ray diffraction techniques.

The sample used in the present experiment was prepared by the general process which makes sheet types of steels. It was approximately 20×20×1.5 mm in size. The Laue diffraction method was used to determine the texture or preferred orientation for the sample qualitatively.

The sample was mounted on the pole figure attachment of the conventional powder diffractometery (RIGAKU Dmax/2200V). The sample orientation was kept making the normal direction (ND) of the Cu sheet surface to be the hemispheric pole. The pole figure data for three reflections, (111), (200) and (220), were measured with CuK_α radiation in reflection geometry. The pole figure data were measured over the orientation hemisphere within the α angles ranging from 0° to 70°, and the β angles ranging from 0° to 360°. The step interval was 5° for each α and β angles. The background intensities were measured at off-Bragg positions. The orientation distribution function (ODF) of the ND was

calculated by the Williams-Imhof-Matthies-Vinel (WIMV) algorithm, using Preferred Orientation Package-Los Alamos (popLA) program.[7] WIMV algorithm uses all available experimental information and tries to find among all possible solutions for the orientation distribution that one which has the smallest discrepancy between experimental and recalculated pole figures and has the sharpest texture. Orthorhombic sample symmetry was used for the texture analysis. The inverse pole figure of the normal direction was obtained from its ODF.

The X-ray diffraction data were obtained at room temperature in the ND over scattering angle 35°~140° at a 2θ step of 0.02° using CuK_α radiation with graphite monochromator. The diffraction data were collected with the θ-2θ scanning mode at the fixed normal direction of the sample orientation. The sample was rotated during the measurement of diffraction data. 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.[8,9]

III. Results and Discussion

There are several ways to determine the effect of texture or preferred orientation of polycrystalline samples. Of them, the back reflection Laue diffraction method was chosen to know the degree of texture in Cu-sheet sample qualitatively. Fig. 1 shows the Laue diffraction patterns for Cu powder and sheet sample, respectively. As shown in Fig. 1, the Debye rings of Cu sheet exhibits non-uniform intensities around their circumstances, whereas the rings of Cu powder sample show smooth and uniform intensities. From these diffraction patterns, we could know that the Cu sheet has texture or preferred orientation. The diffraction pattern patterns for Cu powder and sheet samples with the conventional X-ray diffractometry are shown in Fig. 2. We can see substantial difference between two diffraction patterns as expected from Fig. 1.

The purpose of the Rietveld refinement is to refine the crystal structure of a material under the assumption that crystallites used in the Rietveld refinement have a random orientation distribution (Fig. 1). It, however, is difficult to obtain a sample with ideally random orientation. If the polycrystalline sample is textured as shown in Fig. 1b and Fig. 2b, the observed intensities of diffraction pattern may depend both on the crystal structure and on the texture effect. They can be no longer directly related to structural parameters. If the Rietveld refinement is performed on such diffraction data, it is not possible to obtain a good fitting result due to the texture effect of sample. Therefore, most Rietveld programs often use the semi-empirical texture correction functions, such as March-Dollase and Toraya functions, to correct the texture effect in the powder samples.[4] These functions are useful for a weak and one dimension-

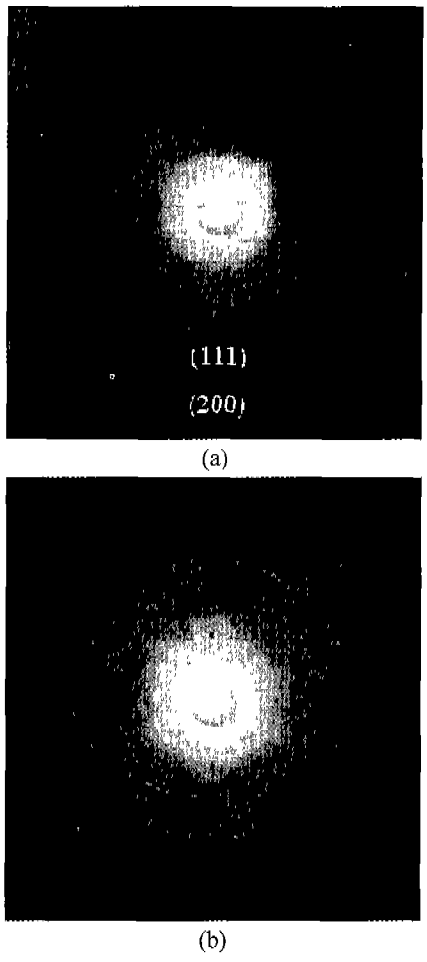

Fig. 1. Laue diffraction patterns: (a) Cu powder and (b) Cu sheet samples.

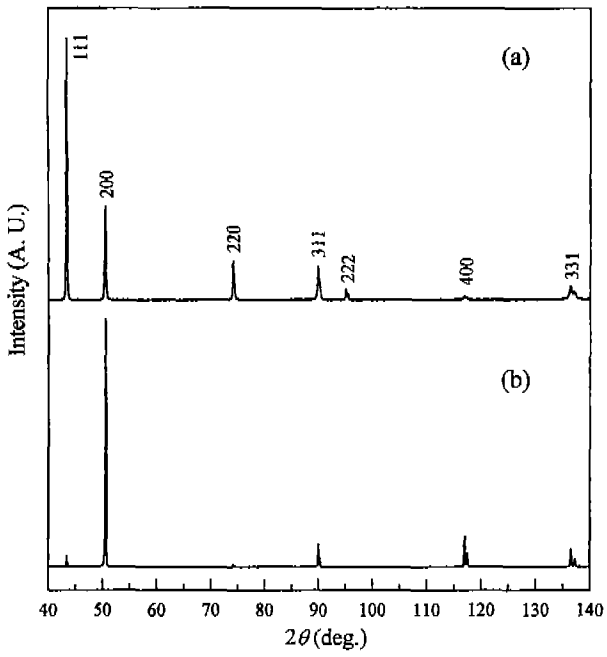

Fig. 2. X-ray diffraction patterns: (a) Cu powder and (b) Cu sheet samples.

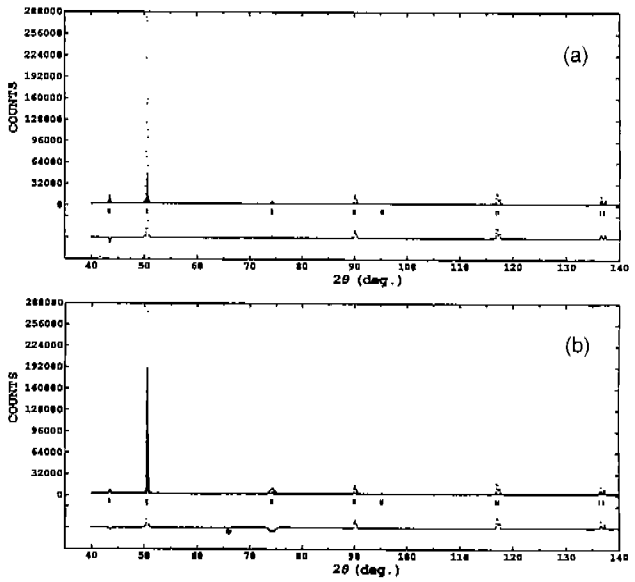

Fig. 3. Rietveld refinement pattern of Cu sheet: (a) without preferred orientation function and (b) with March-Dollase function. Dots represent the observed intensities and the solid line is calculated ones. A difference (obs.-calc.) plot is shown beneath. Vertical bars are the reflection position markers.

ally textured sample. Among them, the March-Dollase function based on the concept of rigid platy or acicular in a viscous medium has been widely used as a technique for correcting the preferred orientation effect.

The initial refinement of Cu sheet in the Rietveld refinement was done by instrumental and lattice parameters. The isotropic thermal factor obtained from Cu powder, B=0.236 Å^2, was used as the initial value of temperature factor. We could not obtain a good refined pattern due to the strong texture effect of Cu sheet as shown in Fig. 3a. We further added preferred orientation parameters as a refinable parameter to take into account the texture effect. The March-Dollase function was applied to the Cu sheet to correct the effect of preferred orientation. However, the agreement between calculated and observed patterns was not very good (Fig. 3b), and showed the high R-factors (R_{wp}= 29.93%, R_p=17.53%, R_I=48.57%, R_F=32.60% and S(=R_{wp}/R_e) =8.48). From the above results, we knew that the use of March-Dollase function for weakly textured samples is very useful but may be limited for heavily textured ones.

Choi *et al.*[10] reported that the texture effect on the diffraction pattern of a certain sample direction is directly proportional to the pole density of the corresponding inverse pole figure, which can be obtained from the three-dimensional orientation distribution function of the material. In order to apply this approach to Cu sheet sample, the pole density of each reflection is required. We measured the pole figure data of three reflections (111, 200, and 220) with the pole figure attachment using conventional powder diffractometer. The ODF of Cu sheet was obtained by the WIMV method, using popLA program.[7] The experimental and

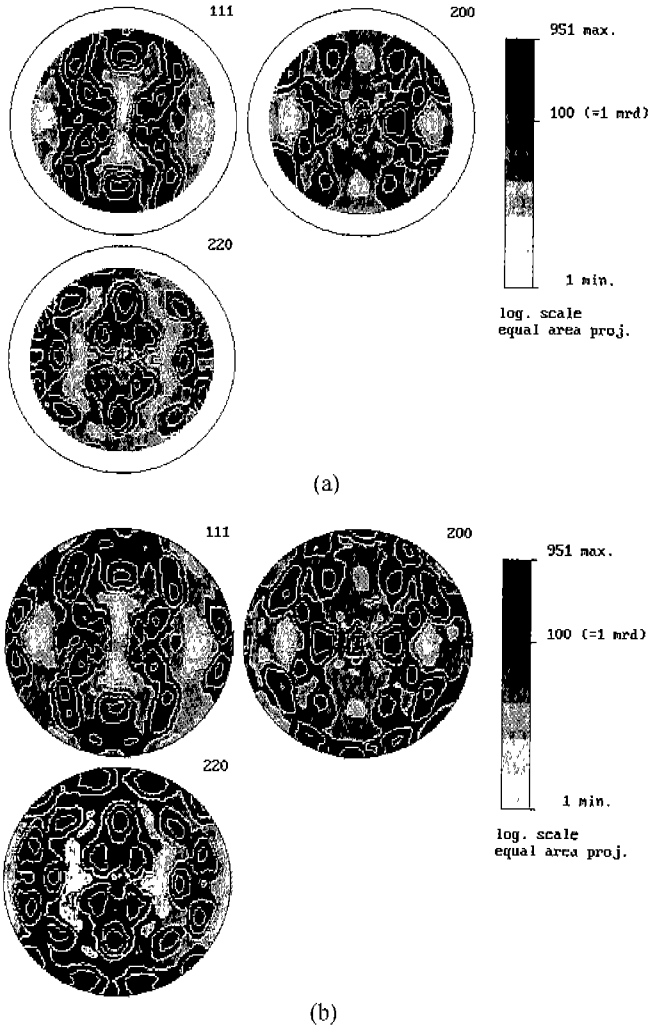

Fig. 4. Pole figures of Cu-sheet: (a) experimental and (b) recalculated pole figures.

recalculated pole figures for three reflections (111, 200, and 220) are shown in Fig. 4. The agreement between the experimental and the recalculated pole figures was satisfactory. Table 1 and Fig. 5 give the pole densities and the inverse pole figure of the normal direction obtained from the ODF, respectively. As listed in Table 1, the pole density value of 200 reflection is significantly higher than that of other one, which is in agreement with the observed diffraction pattern (Fig. 1b).

Table 1. Pole Densities Obtained from the Inverse Pole Figure of the Normal Direction (ND)

h k l	ND
1 1 1	0.46[1)]
2 0 0	7.08
2 2 0	0.31
3 1 1	0.30
3 3 1	4.99

[1)]mulitiples of random distribution

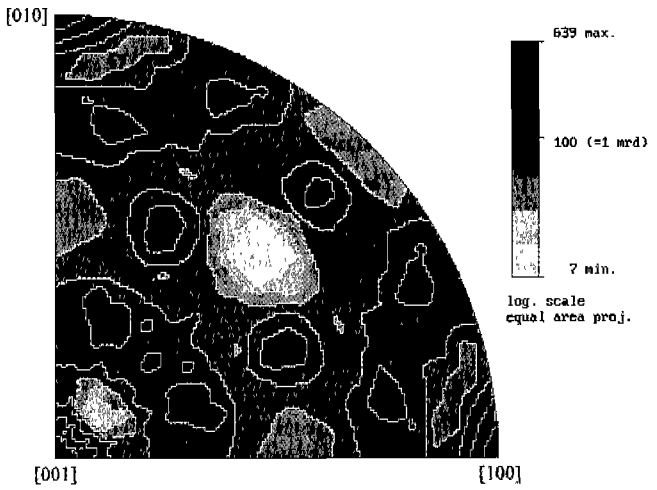

Fig. 5. Inverse pole figure of the normal direction. Equal-area projection; levels are in multiples of random distribution.

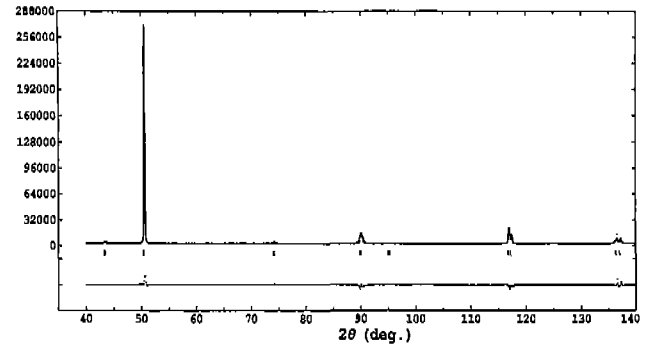

Fig. 6. Rietveld refinement pattern of Cu sheet using the pole densities obtained from the inverse pole figure. Dots represent the observed intensities and the solid line is calculated ones. A difference (obs.-calc.) plot is shown beneath. Vertical bars are the reflection position markers.

In the application of the pole densities of three reflections (111, 200, and 220) obtained from the inverse pole figure of Cu sheet (Fig. 5), R_{wp} was rapidly decreased from 29.93% to 12.99%, but the isotropic thermal parameter, B, converged to negative value, which is physically unreasonable. B was then fixed at 0.236 A^2, which was obtained from the Rietveld refinement of Cu powder. Fig. 6 shows the Rietveld refinement pattern of Cu sheet. Other R-factors (R_p=5.55 %, R_I=14.18%, and R_F=9.54%) were much lower than those in the previous refinement (Fig. 3b) . Also, the goodness-of-fit value, S, was lowered from 8.48 to 3.68. The agreement of the Rietveld refinement was satisfactory. Evidently, this result indicates that the intensity correction technique based on the pole figures could be very useful to do the Rietveld refinement for the highly textured samples.

IV. Conclusion

When the March-Dollase function as a correction factor of the preferred orientation was used to do the Rietveld refinement for the Cu sheet showing (200) preferred orientation, the final R-factors were very high. However, by applying the pole density for each reflection as a preferred orientation factor to the X-ray powder diffraction data, the R-factors were decreased dramatically. The approach using the pole density distribution for each reflection as a correction factor of the preferred orientation may be an alternative to the March-Dollase function in the Rietveld refinement for the highly textured samples if the pole figure data were measured accurately. Also, this work suggests that it can be applied to the kinds of polycrystalline samples, such as fiber, pressed, and extrusion type objects.

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