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Rapid measurement scheme for texture in cubic metallic materials using time-of-flight neutron diffraction at iMATERIA

Yusuke Onuki, Akinori Hoshikawa, Shigeo Sato, Pingguang Xu, Toru Ishigaki, Yoichi Saito, Hidekazu Todoroki and Makoto Hayashi

Rapid measurement scheme for texture in cubic metallic materials using time-of-flight neutron diffraction at iMATERIA

Yusuke Onuki,a,* Akinori Hoshikawa,a Shigeo Sato,a,b Pingguang Xu,c Toru Ishigaki,a Yoichi Saito,d Hidekazu Todorokid and Makoto Hayashie

aFrontier Research Center for Applied Atomic Sciences, Ibaraki University, 162-1 Shirakata, Tokai, Ibaraki 319-1106, Japan, bGraduate School of Science and Engineering, Ibaraki University, 4-12-1 Narusawa-cho, Hitachi, Ibaraki 316-8511, Japan, cJapan Atomic Energy Agency, 2-4 Shirakata, Tokai, Ibaraki 319-1106, Japan, dNippon Yakin Kogyo Co. Ltd, 4-2 Kojima-cho, Kawasaki, Kanagawa 210-8558, Japan, and eCROSS Tokai, 162-1 Shirakata, Tokai, Ibaraki 319-1106, Japan. *Correspondence e-mail: yusuke.onuki.0@vc.ibaraki.ac.jp

A rapid texture measurement system has been developed on the time-of-flight neutron diffractometer iMATERIA (beamline BL20, MLF/JPARC, Japan). Quantitative Rietveld texture analysis with a neutron beam exposure of several minutes without sample rotation was investigated using a duplex stainless steel, and the minimum number of diffraction spectra required for the analysis was determined experimentally. The rapid measurement scheme employs 132 spectra, and by this scheme the quantitative determination of volume fractions of texture components in ferrite and austenite cubic phases in a duplex stainless steel can be made in a short time. This quantitative and rapid measurement scheme is based on the salient features of iMATERIA as a powder diffractometer, i.e. a fairly high resolution in d spacing and numerous detectors covering a wide range of scattering angle.

1. Introduction

Crystallographic texture is recognized as an important factor that can control the properties of materials. The recent development and popularization of electron backscatter diffraction (EBSD) measurements have aided in the clarification of local and spatial crystal orientation distributions of materials and related phenomena (Onuki et al., 2013). However, to consider the texture as an overall property of a material, statistically sufficient information from a large enough volume is necessary. Pole figure measurements using X-rays have typically been used for this purpose, along with orientation distribution function (ODF) calculations (Kocks et al., 1998). However, the X-ray beam penetration depths in metallic materials are limited to a few tens of micrometres. Therefore, both X-ray and EBSD measurements can only represent the texture on the measured surface, while many engineering materials have inhomogeneous texture distributions through their thickness. For example, the layer on the surface and the thickness centre of a rolled sheet often have different textures (Truszkowski et al., 1982).

A beam of neutrons has a high transmittance in most metals, so it can be an ideal quantum beam for texture measurements to investigate the overall properties of materials. This advantage of neutron diffraction has been recognized in multiple published studies (Jensen & Kjems, 1983; Oles et al., 1985; Wenk, 1991). Additionally, the recent development of whole-spectrum analysis using time-of-flight (TOF) neutron diffraction has enabled the analysis of textures in multiphase materials (Matthies et al., 1997, 1999; Wenk et al., 2010).
The present authors have developed a neutron-beam texture measurement system based on the Ibaraki Materials Design Diffractometer, iMATERIA, on beamline BL20 at the Material and Life Science Facility of the Japan Proton Accelerator Research Complex (MLF/J-PARC, Japan). In addition to the above-mentioned advantages of TOF neutron diffraction, it has become possible to measure texture extremely rapidly with no sample rotation. In this paper, the details of the measurement and analysis methods are introduced.

2. Method
2.1. Instrument

The TOF diffractometer iMATERIA (BL20 at MLF/J-PARC, Japan) has a vacuum sample chamber and multiple detector banks covering a wide 2θ range. Detailed specifications can be found in a previous paper (Ishigaki et al., 2009).

Fig. 1 is a schematic diagram of the detector banks of iMATERIA. For texture measurements, the backscatter (BS, 145 ≤ 2θ ≤ 175°), special environment (SE, 79 ≤ 2θ ≤ 101°) and low-angle (LA, 12 ≤ 2θ ≤ 40°) detector banks are used. The banks are further separated into small regions called the ‘observation points’ (OPs). Each OP is assumed to behave as a point detector that measures the diffraction spectrum. The spectrum includes the Bragg diffraction peaks from various (hkl) crystallographic planes. Let \( \mathbf{y} \) be the normal direction of the (hkl) plane in the diffractometer coordinate system, \( \mathbf{X}_{D} = Y_D - Z_D, \) \( \mathbf{y} \) can be calculated from the positional vector of the OP, \( \mathbf{p} \), as

\[
\mathbf{y} = \mathbf{p}/|\mathbf{p}| - \mathbf{e}_{X_D},
\]

where \( \mathbf{e}_{X_D} \) is the unit vector along \( X_D \), which is parallel to the incident beam. The sample loader of iMATERIA can rotate the sample around the \( Y_D \) axis on the measurement position. Therefore, the coordinate conversion matrix \( \mathbf{\Omega}(\omega) \), a function of the rotation angle \( \omega \), should be applied in order to obtain the (hkl) normal in sample coordinates, \( \mathbf{y} \), as

\[
\mathbf{y} = \mathbf{\Omega} \mathbf{y}'.
\]

The sizes of the OPs were determined such that the angular tolerance of \( \mathbf{y} \) was smaller than 3°.

2.2. Analysis

Conventional texture analysis is based on pole figure measurements. In this method, the diffraction intensities from several planes \([hkl]\) are measured along ~1000 different \( \mathbf{y} \) vectors (Kocks et al., 1998). Rietveld texture analysis (Matthies et al., 1997, 1999; Wenk et al., 2010), which was applied in this study, uses diffraction spectra corresponding to various \( \mathbf{y} \) as the input for the ODF calculation. Fig. 2(a) shows some spectra used in the following analysis. Since the spectra include many diffraction peaks, the number of \( \mathbf{y} \) vectors required for quantitative analysis can be fewer than that for pole-figure-based ODF analysis. The diffraction intensities in the spectra are evaluated by Rietveld analysis or spectral fitting. At the first fitting, a uniform ODF corresponding to no texture is applied. Hence, the diffraction intensities of some peaks are overestimated while those of others are underestimated. The differences in the diffraction intensities between the measured and calculated spectra indicate the axis densities along \( \mathbf{y} \). Therefore, the spectra can be regarded as ‘input inverse pole figures’, which contain the data points at the red dots in Figs. 2(b) and 2(c). ODFs are calculated based on them and the results are passed back to the Rietveld analysis, resulting in better fitting. By repeating the paired Rietveld refinements and the ODF calculation several times, both the spectral fitting and the ODF converge to the most likely solutions. More details on Rietveld texture analysis can be found in the literature by the developers (Matthies et al., 1997; Wenk et al., 2010).

In this study, the MAUD (materials analysis using diffractometer) software package (Wenk et al., 2010) was used to conduct the above analysis. This software has a ‘HIPPO wizard’ function to treat spectra measured at HIPPO, a TOF neutron diffractometer at the Los Alamos Neutron Science Center (LANSCE), USA (Wenk et al., 2003). Because the structure of iMATERIA is somewhat similar to that of HIPPO, the HIPPO...
wizard could be used to import the spectra acquired at iMATERIA. The unique instrumental parameters of iMATERIA were supplied by a separate instrumental parameter file. However, the incident beam profile used at iMATERIA was too complex for the preset incident beam profile functions to reproduce the profile shape. For this reason, the input spectra were externally normalized (Izumi et al., 1987). The range for the analysis was set so that each spectrum contained ~20–30 diffraction peaks for each phase. For steel samples with cubic crystal structures, a range from 0.4 to 2.3 Å was used, as shown in Fig. 2(a). Since iMATERIA was designed as a powder diffractometer, the d-spacing resolution was high even for values below 1 Å. Therefore, iMATERIA could find many peaks with minimum overlapping ambiguity (Matthies et al., 1997).

The entropy-modified Williams–Imhof–Matthies–Vinel (E-WIMV) method (Lutterotti et al., 2004) was employed as the ODF calculation method. Although MAUD also provides the conventional harmonic expansion method, discrete ODF calculation methods such as E-WIMV are more suitable for strongly textured engineering samples, e.g. electrical steel (Matthies et al., 2005).

2.3. Samples

A rolled sheet of NAS64 duplex stainless steel (JIS SUS329J4L, ASTM A240) was supplied by the Nippon Yakin Kogyo Co. Ltd. The chemical composition is given in Table 1. The supplied sheet was additionally cold-rolled to a thickness reduction of 20% in the laboratory. The steel contained two phases, ~60 mass% of body-centred cubic (b.c.c.) ferrite and ~40 mass% of face-centred cubic (f.c.c.) austenite. Using Rietveld texture analysis, the textures in multiphase materials can be determined even with overlapping dominant peaks.

2.4. Measurement schemes

As stated in §2.2, Rietveld texture analysis uses diffraction spectra along various y as the input. However, the number of spectra necessary to calculate a quantitative ODF with a resolution of 5° was unclear. In order to determine the optimal number of spectra, we firstly set 36 OPs. Therefore, 36 spectra were obtained by one exposure of the neutron beam without sample rotation. A stereographic projection of the distribution of y is shown in Fig. 3(a). By repeating this measurement at different sample angles \( \omega \) (rotation around \( Y_D \)), the total

<table>
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<th>Table 1</th>
<th>Chemical composition of the NAS64 duplex stainless steel (mass%).</th>
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<tr>
<td>C</td>
<td>Si</td>
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<tr>
<td>0.01</td>
<td>0.4</td>
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Since we focused on texture analyses for metallic engineering materials, the resolution of the ODF (voxel size in Euler space) was set to 5°. This is the same resolution as that usually constructed by monochromatic X-ray measurements (Kocks et al., 1998).
number of spectra ($N$) can be multiplied. Figs. 3(b)–3(e) depict
the distribution of $y$ in the sample coordinate system achieved
by multiple measurements. In this way, $N$ can be increased
indefinitely, while the total measurement time is also
increased. Additionally, calculations using MAUD become
time consuming when hundreds of spectra are provided.

In order to realize fast measurement and analysis, an
alternative measurement scheme is suggested using the 132
OPs shown in Fig. 4. In this case, the 132 spectra can be
obtained simultaneously without sample rotation. This
enables rapid texture measurement, requiring only 1–3 min of
data acquisition without sample rotation. Hereinafter,
the results from the above schemes are referred to by $N$, where $N = n \times 36$ ($n = 1, 2, \ldots, 5$) indicates results from the former
scheme and $N = 132$ corresponds to the latter.

The measurements were conducted at a beam power of
500 kW. The receiving frequency for the incident pulse
neutron was 25 Hz.

3. Results and discussion

3.1. Optimal number of spectra for texture analysis

Figs. 5 and 6 show the [110] pole figures for the ferrite and
austenite phases in the 20% rolled duplex stainless steel
analysed with different numbers of input spectra. All show
the well known conventional pole distributions for rolled
textures (Hamada et al., 2003; Hirsch & Lücke, 1988).
However, the maximum intensity is somewhat low for cases
where $N < 108$. The contour lines are rough with insufficient $N$,
implying a lack of input data to construct the ODF. With $N \geq 108$, these problems are suppressed and the features of the
pole figures are not affected by $N$. Hence, it can be said that
the quality of the pole figure is saturated for $N > 100$.

3.2. Rapid measurement scheme

From the above discussion, the number of spectra $N$ for
texture analysis must exceed 100. This indicates that the rapid
measurement scheme using 132 OPs (Fig. 4) can also provide a
quantitative texture analysis.

Fig. 7 shows the calculated pole figures for the ferrite and
austenite phases using the rapid measurement scheme, $N =
132$. The figures agree well with the results shown in Figs. 5 and
6. It seems that the homogeneity of the distribution of OPs is
not as important as the number of OPs, by considering the
differences between Figs. 3 and 4.

For a more detailed texture analysis, metallurgists often
examine textures using the ODF in Euler space rather than
pole figures. The ODFs in Bunge’s Euler space ($\varphi_1, \Phi, \varphi_2$)
were constructed from the pole figures exported by MAUD
using the LaboTex software (Pawlik & Ozga, 1999). Because
six complete pole figures were used as input for calculating the
ODF for each phase, the ambiguity introduced in this process
is negligible. In the reconstruction, orthorhombic sample
symmetry was applied, resulting in $\varphi_1$ values ranging between
0 and 90°.
In this study, the tolerance angles \( \phi_2 = 45^\circ \) cross sections of the ODFs for both the ferrite and austenite phases are shown in Fig. 8. The results of the scheme used in the previous section \((N = 5 \times 36\) OP\) are also indicated. The results of the rapid scheme with \(N = 132\) (Figs. 8b and 8d) show good agreement with those from the \(N = 5 \times 36\) scheme. In addition, both phases indicate familiar rolled textures. The texture of the ferrite phase consists of a \((hkh)(110)\) and \(y(111)uvw\) fibres (Hamada et al., 2003), while that of the austenite phase has brass \((110)(112)\), Goss \((110)(001)\) and copper \((112)(111)\) components (Hirsch & Lücke, 1988)

In order to verify that quantitative texture component analysis is possible using the new method, the volume fractions of some common texture components were calculated. The volume fraction \(V(g)\) of a texture component at an orientation \(g(\psi_1, \Phi, \phi_2)\) can be calculated as

\[
V(g) = \frac{1}{8\pi^2} \int f(g) \, dg
\]

\[
= \frac{1}{8\pi^2} \int_{\psi_2-\Delta\psi_2}^{\psi_2+\Delta\psi_2} \int_{\phi-\Delta\phi}^{\phi+\Delta\phi} \int_{\phi_1-\Delta\phi_1}^{\phi_1+\Delta\phi_1} f(\psi_1, \Phi, \phi_2) \sin \Phi \, d\phi_1 \, d\Phi \, d\psi_2.
\]

In this study, the tolerance angles \(\Delta\psi_1 = \Delta\Phi = \Delta\phi_2 = 10^\circ\) were applied. The results are listed in Tables 2 and 3 for the ferrite and austenite phases, respectively. It is seen that the volume fractions of the texture components are almost constant above \(N = 72\) \((= 2 \times 36)\). The differences fall within 0.8%. Further-

more, the orders of volume fractions in both phases are also constant above \(N = 72\), including \(N = 132\).

Table 2 shows the analysed weight fractions of the austenite phase with different \(N\). Again, the values obtained with \(N \geq 72\) are similar, whereas the phase fraction with \(N = 36\) is a little smaller than the others. Since the phase fraction is determined from the diffraction intensities, the correct values can be obtained when accurate texture analyses are conducted for both phases. Therefore, these results also imply convergence of the Rietveld texture analysis with the data from iMATERIA with \(N \geq 72\).

Thus, the TOF neutron diffractometer iMATERIA is capable of detailed texture measurement with ODF analysis, which is often conducted on the basis of the results of conventional X-ray texture measurements. The most striking feature of the present method is the very short time required for the measurement. The complete ODF for steel can be

![Figure 7](image)

Figure 7

(a) The [110] pole figure of the ferrite phase and (b) the [100] pole figure of the austenite phase for the 20% rolled duplex stainless steel, calculated by the optimized method using 132 OPs.
obtained within a few minutes of exposure. This is especially beneficial for the in situ measurement of textures during deformation and annealing. The authors are now developing suitable deformation and heating environments for such in situ measurements.

4. Conclusions

A texture measurement system using Rietveld texture analysis has been developed using iMATERIA, a TOF neutron diffractometer at MLF/J-PARC, Japan. By optimizing the analysis method, a very fast texture measurement system has been developed. The main results of this study are as follows.

1. Rapid quantitative texture analysis is possible within a few minutes for duplex stainless steels using iMATERIA, which is equipped with numerous detectors covering a wide range of scattering angle and has a high d-spacing resolution at a beam power of 500 kW.

2. Rietveld analysis has been employed to analyse the texture of the material by deducing the ODF from many TOF diffraction spectra having a high d-spacing resolution.

3. The number of diffraction spectra required for a reliable and quantitative texture analysis was determined experimentally at about 100. The rapid texture measurement scheme thus developed consists of 132 spectra acquired in a few minutes without any rotation of the sample.

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References