DETERMINATION OF PREFERRED ORIENTATION OF CUBIC CRYSTALS IN THE EXPERIMENTALLY PRODUCED TECTONITES WITH A SPECIAL REFERENCE TO SPINEL TYPE MINERAL

AKIO FUJIMURA*, ICHIRO OHNO** and MINEO KUMAZAWA*

Abstract The basic problems are discussed on the determination of the preferred orientation of cubic crystallites in the experimentally produced tectonites by means of X-ray diffraction. The practical methods to acquire experimentally the orientation data by an X-ray pole figure goniometer are described, and the theoretical and numerical procedures to derive the reliable inverse pole figure and its error estimation are presented.

As an analogue to γ -(Mg, Fe)₂SiO₄, the major constituent of spinel layer in the upper mantle, trevorite (NiFe₂O₄, spinel structure) aggregates are deformed at high temperature and high pressure. The structural developments in the trevorite tectonite deformed uniaxially are studied by the pole figure inversion technique. The preliminary results indicated that at least two different types of preferred orientation ($\langle 110 \rangle$ //compression, and $\langle 111 \rangle$ //compression) were generated in the trevorite tectonite by syntectonic recrystallization.

1. Introduction

The preferred orientation (PO) or orientation density distribution (OD) of minerals in tectonites should bear the information on the states of stress, strain rate, total strain, pressure, temperature, and so on, to which the rocks have been subjected. In other words, the OD is regarded as a certain kind of fossils of the physical conditions. For this reason, many workers have made petrofabric observations in natural rocks since SANDER (1930) and SCHMIDT (1925). It is not easy to derive the relation between OD and the physical conditions only by the description of consequential OD. D. T. GRIGGS and his

co-workers have been trying to acquire the physical conditions and/or mechanisms of PO of minerals in the experimentally produced tectonites prepared under the controlled conditions. This approach has been now widely made, and some of the results up to date were well reviewed by CARTER (1976). In these studies, it is a basic problem to determine the complete orientation density distribution function (abbreviated to ODF) of crystallites in natural and experimental aggregates.

In the usual petrofabric study, the orientations of crystallites in a specimen are measured directly by means of a microscope with a universal stage. On the other hand, several X-ray methods (e.g., PHILLIPS & BRADSHAW, 1970; GOEMAN & SCHUMANN, 1975) are also employed to determine the ODF of crystallites for which optic method is difficult to apply. The typical examples are opaque materials such as metals, and optically isotropic or uniaxial minerals.

The pole figure or the plane normal ori-

Received Sept. 4, 1978

* Department of Earth Sciences, Nagoya University, Nagoya, 464 Japan.

** Department of Earth Sciences, Nagoya University, Nagoya, 464 Japan.

Present address; Department of Earth Sciences, Faculty of Science, Ehime University, Matsuyama, Ehime, 790 Japan.

Fig. 1. Representation of a unit vector $\mathbf{n}_i$ and of Euler angles ϕ , θ and ϕ . (a) A unit vector $\mathbf{n}_i$ is specified by colatitude χ_i and longitude η_i in specimen coordinates $o\text{-}xyz$. (b) The same unit vector $\mathbf{n}_i$ is specified by colatitude θ_i and longitude ϕ_i in crystallite coordinates $O\text{-}XYZ$. (c) The rotational relationship between $o\text{-}xyz$ and $O\text{-}XYZ$ is represented by three Euler angles ϕ , θ and ϕ . The Z -axis is specified with two Euler angles θ and ϕ , and the rotation around Z -axis is represented by an Euler angle ϕ in the system $o\text{-}xyz$.

entation density distribution for a certain crystallographic plane in cubic crystals can be measured as the relative intensity of X-ray diffraction. Therefore, X-ray pole figure technique is quite suitable to the study of spinel aggregates. Spinel type minerals are supposed to be an important cubic constituent in the mantle of the earth, and the PO of such minerals has some significance in global tectonics.

Since the deformation apparatus available at present is incapable of generating the stable pressure condition of $\gamma\text{-(Mg, Fe)}_2\text{SiO}_4$, we can not obtain the tectonite of $\gamma\text{-(Mg, Fe)}_2\text{SiO}_4$. We have started to study the experimental syntectonic recrystallization of trevorite NiFe_2O_4 ; an analogue of $\gamma\text{-(Mg, Fe)}_2\text{SiO}_4$, and to determine the ODF generated in the laboratory.

In this paper, the technical problems in measurements and analysis of OD of trevorite tectonites are discussed. First, the method of a pole figure inversion from X-ray data is described for cubic crystallites in the polycrystalline material having a uniaxial symmetry. The pole figure inversion is a mathematical method, by which a complete description of the OD in terms of ODF is derived from measured pole figures. The pole figure inversion technique has developed

in the textural study of polymer and metal (ROE & KRIGBAUM, 1964; KRIGBAUM & ROE, 1964; ROE, 1966; BUNGE, 1965), and was applied later to the study of trigonal minerals, such as quartz, calcite and dolomite in tectonites (BAKER, 1969; BAKER *et al.*, 1969; BAKER & WENK, 1972; WENK & WILD, 1972; RIEKELS & BAKER, 1977; WENK *et al.*, 1973; BAKER & RIEKELS, 1977; WENK & SHORE, 1975). Second, the practical method of deriving the complete description of trevorite fabric is presented together with some preliminary results on the preferred orientation types of spinel minerals. An emphasis is placed on the necessary procedure to derive reliable result and on the method of error estimation both in the measurement and data processing. Symbols and notations in this paper follow those employed by ROE & KRIGBAUM (1964).

2. X-ray analysis of preferred orientation

1 Coordinate systems to represent the crystallographic orientation

We set an orthogonal system $o\text{-}xyz$ on a polycrystalline sample and another orthogonal system $O\text{-}XYZ$ on a crystallite to describe the orientation of the crystallite in the sample. It is convenient, if possible,

Fig. 2. Relationship among a unit vector $\mathbf{n}_i$, specimen coordinates o-xyz and crystal-lite coordinates O-XYZ. Each relation connected with a tie line between two of the three mentioned above, is represented by the angular parameters in parentheses.

to make X-, Y- and Z-axis coincide with three crystallographic axes $\langle 100 \rangle$, $\langle 010 \rangle$ and $\langle 001 \rangle$, respectively. The orientation of O-XYZ with respect to o-xyz is represented by the Euler angles ϕ , θ and ψ (Fig. 1c). Let the z-axis coincide with the symmetric axis of the uniaxial sample. Then ξ is introduced to denote colatitude by $\xi = \cos \theta$.

A unit vector $\mathbf{n}_i$ (reciprocal lattice vector) is used to represent the i th crystallographic plane. The orientation of $\mathbf{n}_i$ with respect to O-XYZ is given by colatitude θ_i and longitude ϕ_i (Fig. 1b), and that with respect to o-xyz is described by colatitude χ_i and longitude η_i (Fig. 1a). Here, Ξ_i and ξ_i are introduced to denote colatitudes by $\Xi_i = \cos \theta$ and $\xi_i = \cos \chi_i$, respectively. The relationship among o-xyz, O-XYZ and $\mathbf{n}_i$ is represented by three sets of angles (θ_i, ϕ_i) , (χ_i, η_i) and (ψ, θ, ϕ) as shown in Fig. 2, and is given in Appendix A.

2 ODF and its representation with collected X-ray intensity data

For a sample of uniaxial symmetry around z-axis, as in the present case, OD is independent of ϕ , and X-ray diffraction in-

tensity is independent of η_i .

The OD in a polycrystalline sample is described by specifying the number of crystallites as a function of orientation. For this purpose, a function $w(\xi, \phi)$ is defined in such a way that the number of crystallites lying in the orientation range from (ξ, ϕ) to $(\xi + d\xi, \phi + d\phi)$, is proportional to $w(\xi, \phi) d\xi d\phi$. The orientation density distribution function $w(\xi, \phi)$ (abbreviated to ODF) shows the probability density of finding a crystallite with an orientation (ξ, ϕ) .

The function $w(\xi, \phi)$ can be expanded by spherical harmonics;

$$w(\xi, \phi) = \sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} W_{\ell m} \tilde{P}_{\ell}^m(\xi) \exp(-im\phi) \quad (1)$$

where $\tilde{P}_{\ell}^m(\xi)$ ($\ell=0, 1, 2, \dots, \infty$, $m=-\ell, -\ell+1, \dots, \ell-1, \ell$) are the normalized associated Legendre polynomials, and $W_{\ell m} (=A_{\ell m} + iB_{\ell m})$ are the expansion coefficients (see Appendix B). Thus, $w(\xi, \phi)$ is described by the sets of $W_{\ell m}$ and the known functions of ξ and ϕ . In other words, $W_{\ell m}$ are the necessary and sufficient numerical parameters to describe ODF $w(\xi, \phi)$.

In the determination of OD by X-rays, the acquired data are the plane normal orientation distribution functions $q_i(\zeta_i)$ ($i=1, 2, 3, \dots$), which are proportional to the X-ray intensity diffracted by i th crystallographic planes orienting along ζ_i in a polycrystalline sample (see Appendix C). The $q_i(\zeta_i)$ is the normalized pole figure of one dimension and is expanded into a series of Legendre polynomials:

$$q_i(\zeta_i) = \sum_{\ell=0}^{\infty} Q_{\ell}^i \tilde{P}_{\ell}(\zeta_i) \quad (2)$$

where $\tilde{P}_{\ell}(\zeta_i)$ are the normalized Legendre polynomials, and Q_{ℓ}^i are the expansion coefficients (see Appendix C). Thus, $q_i(\zeta_i)$ is described by the sets of Q_{ℓ}^i and the known function of ζ_i . In other words, Q_{ℓ}^i are the necessary and sufficient numerical parameters describing the acquired data on the OD.

Therefore, we can determine $w(\xi, \phi)$ from $q_i(\zeta_i)$, once the relations between $W_{\ell m}$ and Q_{ℓ}^i are given. The relations have been given

by ROE & KRIGBAUM (1964) as follows (see Appendix D);

$$Q_i^t = 2\pi [2/(2t+1)]^{1/2} \{A_{t,0} \tilde{P}_t(\mathcal{E}_i) + 2 \sum_{m=1}^t (-1)^m [A_{t,m} \tilde{P}_t^m(\mathcal{E}_i) \cos m \Phi_i - B_{t,m} \tilde{P}_t^m(\mathcal{E}_i) \sin m \Phi_i]\} \quad (3)$$

(i = 1, 2, 3, ...)

Equation (3) which is applied to triclinic crystallites, is a set of linear equations with $(t+1)$ unknowns; $W_{t,m}$ ($m=0, 1, 2, \dots, t$), for given t . The angular parameters θ_i and Φ_i in eq. (3) are given by Miller indices of diffraction planes and lattice constants. By putting the numerical parameters Q_i^t into eq. (3), we obtain $W_{t,m}$ ($=A_{t,m} + iB_{t,m}$) as a function of Q_i^t (the $W_{t,m}$ are not affected by $Q_i^{t'}, (t' \neq t)$).

When it is necessary to incorporate such a diffraction peak that is made up with several overlapped diffractions (see section 3.4 for practical example) in order to obtain the ODF with better accuracy, we necessarily use

$$Q_i^t = 2\pi [2/(2t+1)]^{1/2} \times \{ \sum_j C_{ij} A_{t,0} \tilde{P}_t(\mathcal{E}_{ij}) + 2 \sum_{m=1}^t (-1)^m C_{ij} \times [A_{t,m} \tilde{P}_t^m(\mathcal{E}_{ij}) \cos m \Phi_{ij} - B_{t,m} \tilde{P}_t^m(\mathcal{E}_{ij}) \times \sin m \Phi_{ij}] \} \quad (4)$$

instead of eq. (3), though the relative weighting factors C_{ij} of all component peaks involved are needed. The C_{ij} denotes relative diffraction intensities of the j th diffraction peaks contributing to one overlapped i th diffraction.

Equation (4) is useful not only for a composite peak but also for a case where the integrated intensity data of two or more diffraction peaks lying close each other are not acquired independently as a consequence of the wide slit of pole figure goniometer.

3 Restriction on ODF due to symmetry of crystallite and their aggregate

Equations (3) and (4) are based only on the uniaxial symmetry around z-axis of the sample and also the symmetry due to Friedel's law. When the crystallites have higher symmetry than triclinic, further symmetric relations on $W_{t,m}$ arise from multiplicity.

Table 1

Symmetry requirements of $W_{t,m}$ for materials consisting of cubic crystals $B_{t,m} \equiv 0$			
t	m	$A_{t,m}$	
0	0	0.11253954	($=1/(\sqrt{8}\pi)$)
2	0	0.0	
4	0	linearly independent	
4	4	$0.59761430 \times A_{40}$	
6	0	linearly independent	
6	4	$-1.8708287 \times A_{60}$	
8	0	linearly independent	
8	4	$0.37605072 \times A_{80}$	
8	8	$0.57295971 \times A_{80}$	

Accordingly, all numerical parameters $W_{t,m}$ in eq. (3) are not independent.

In case of cubic crystallites, the following restrictions on $W_{t,m}$ are existing. The presence of three mutually orthogonal symmetry planes in cubic crystallites brings $B_{t,m} = 0$ for every t and m , $A_{t,m} = 0$ for odd t . A four-fold rotation symmetry around Z-axis brings $A_{t,m} = 0$ for $m \neq 4k$ where k is an integer $0, 1, 2, \dots, \leq t/4$. Further symmetric restriction on $W_{t,m}$, such as $A_{20} = 0$, is brought by a four-fold rotation symmetry around X- and/or Y-axis. The detailed procedures for obtaining the restriction on $W_{t,m}$ owing to cubic symmetry have been described by ROE (1966). The symmetry requirements on $W_{t,m}$ ($0 \leq m \leq t \leq 8$) are listed in Table 1.

The requirements on Q_i^t for the cubic crystallites in a uniaxial polycrystalline sample are given as

$$Q_0^t = 1/\sqrt{2} \quad (\text{normalization condition})$$

$$Q_2^t = 0$$

$$Q_i^t = 0 \quad \text{for odd } t \quad (\text{Friedel's law})$$

It is noted that the cut-off of the $W_{t,m}$ with larger t makes the data analysis quite practical without losing the useful information as is discussed in section 4.1, and it also makes the relation (3) very simple. When $t \leq 8$, we have

$$Q_i^t = 2\pi \sqrt{2/9} A_{40} [\tilde{P}_4(\mathcal{E}_i) + 2(A_{44}/A_{40}) \tilde{P}_4^4(\mathcal{E}_i) \cos 4\Phi_i] \quad (5a)$$

Fig. 3. Illustrations (in unit triangles of the crystallite coordinate) showing the relative contribution of the Q_i^1 to determine $A_{i,0}$, which depends on the orientation of the i th crystallographic plane.

(a) $[\tilde{P}_4(\mathcal{E}_i) + 2(A_{44}/A_{40})\tilde{P}_4^4(\mathcal{E}_i)\cos 4\Phi_i]$ in eq. (5a).

(b) $[\tilde{P}_6(\mathcal{E}_i) + 2(A_{64}/A_{60})\tilde{P}_6^4(\mathcal{E}_i)\cos 4\Phi_i]$ in eq. (5b).

(c) $[\tilde{P}_8(\mathcal{E}_i) + 2(A_{84}/A_{80})\tilde{P}_8^4(\mathcal{E}_i)\cos 4\Phi_i + 2(A_{88}/A_{80})\tilde{P}_8^8(\mathcal{E}_i)\cos 8\Phi_i]$ in eq. (5c).

$$Q_6^1 = 2\pi\sqrt{2/13} A_{60} [\tilde{P}_6(\mathcal{E}_i) + 2(A_{64}/A_{60})\tilde{P}_6^4(\mathcal{E}_i)\cos 4\Phi_i] \quad (5b)$$

$$Q_8^1 = 2\pi\sqrt{2/17} A_{80} [\tilde{P}_8(\mathcal{E}_i) + 2(A_{84}/A_{80})\tilde{P}_8^4(\mathcal{E}_i)\cos 4\Phi_i + 2(A_{88}/A_{80})\tilde{P}_8^8(\mathcal{E}_i)\cos 8\Phi_i] \quad (5c)$$

The important point in eq. (3) is that we can describe complete ODF through calculating only three independent parameters A_{40} , A_{60} and A_{80} for cubic crystallites in a sample with uniaxial symmetry (see Table 1).

Each equation in (5a, b, c) has only one unknown parameter: A_{40} , A_{60} or A_{80} . This fact indicates that a perfect ODF is theoretically determined uniquely from expansion coefficients Q_i of only one pole figure of any diffraction plane. For practical procedure, however, we have to employ the least-squares method to obtain reliable ODF by utilizing several pole figures. This method is to determine the best set of values $A_{i,0}$, which minimize the following sum;

$$S = \sum_i \rho_i \{ A_{i,0} \sum_j C_{ij} \tilde{P}_i(\mathcal{E}_{ij}) + \sum_m A_{i,m} \sum_j C_{ij} 2\tilde{P}_i^m(\mathcal{E}_{ij}) \cos m\Phi_{ij} - [1/\sqrt{(2i+1)/2}] [Q_i^1/(2\pi)] \}^2 \quad (6)$$

($i=4, 6, 8$)

where $A_{i,m}$ are separated into $A_{i,0}$ and $A_{i,m}$ ($m \neq 0$), and ρ_i are weighting factors of individual $q_i(\zeta_i)$ (see section 3.4 (b)).

The quantity in square brackets in eqs. (5a, b, c) is a function of the orientation

Fig. 4. A standard stereographic projection of a cubic crystal. (a) A unit triangle (an independent portion for cubic symmetry) is shown by a shaded area. (b) A unit triangle showing the poles of six crystallographic planes (open circles) on which intensity data were collected, and the poles of three crystallographic planes (solid circles) for which intensity profiles were synthesized from ODF (see Fig. 9).

$(\mathcal{E}_i, \Phi_i)$ of the i th diffraction plane, and gives us an information about the relative contribution of the Q_i^1 to determine $A_{i,0}$. The orientation dependency of the quantity is shown by the contour map in a unit triangle (Fig. 3). The orientations of diffraction planes whose X-ray intensity data are measured and/or calculated in this paper, are shown in a unit triangle (Fig. 4(b)). A unit triangle portion of a standard stereographic projection is sufficient to describe the orientation data of cubic crystals (Fig. 4(a)).

4 Representation of ODF by an inverse pole

Fig. 5. Illustration showing the relationship between ODF $w(\theta, \phi)$ and inverse pole figure $I(\alpha, \beta)$. (a) The coordinates O-XYZ of the crystallite is specified by θ and ϕ in the specimen coordinates o-xyz. (b) The same relation as described in (a) in the other coordinates rotated θ around y-axis. (c) The same relation as described in (a) and (b) in the crystallite coordinates O-XYZ.

figure

The conventional pole figure shows the OD of the poles of a particular crystallographic plane with respect to the specimen coordinates. A pole figure is the representation of a function of one parameter χ_1 (in general, of two parameters χ_1 and η_1), though ODF is a function of two parameters θ and ϕ (in general, of three parameters ψ , θ and ϕ). This is the reason why a pole figure gives us the comprehensible but deficient information about the OD. However, an inverse pole figure is a function of two angular parameters and provides the sufficient information. Representation of OD by an inverse pole figure gives us the information on the OD of an axis having a particular significance to the specimen (in the present case, the symmetrical axis of the deformed specimen, which is specified with colatitude α and longitude β) with respect to the crystallite coordinates. An inverse pole figure representation, therefore, provides a full information on the OD, in conformity with the two-parameter description of ODF.

The number of crystallites having the orientation in the angular range of $d\theta$ and $d\phi$ at (θ, ϕ) is proportional to $w(\theta, \phi) \sin\theta d\theta d\phi$. An inverse pole figure representation $I(\alpha, \beta)$ shows that the distribution probability of z-axis in an angular range of $d\alpha$ and $d\beta$ at (α, β) is proportional to $I(\alpha, \beta) \sin\alpha d\alpha d\beta$. We observe that $\alpha = -\theta$ and $\beta = -\phi$, and that the conversions of $-\theta$ to θ , and of $-\phi$

to ϕ , do not give rise to any change in the expression of $w(\theta, \phi)$ in cubic symmetry (see Fig. 5). Therefore, $w(\theta, \phi)$ with $\theta = \alpha$ and $\phi = \beta$ is identified as an inverse pole figure $I(\alpha, \beta)$.

Here, we introduce two functions $G(\xi, \phi)$ and $\Delta G(\xi, \phi)$ which are related to $w(\xi, \phi) = I(\alpha, \beta)$:

$$\begin{aligned} G(\xi, \phi) &= 4\pi w(\xi, \phi) \\ \Delta G(\xi, \phi) &= G(\xi, \phi) - 1 \end{aligned} \quad (7)$$

$G(\xi, \phi)$ and $\Delta G(\xi, \phi)$ are useful for comprehensive representation of OD, since $G(\xi, \phi) \equiv 1$ and $\Delta G(\xi, \phi) \equiv 0$ for the case of complete random orientation distribution. The $\Delta G(\xi, \phi)$ shows the degree of preferred orientation; positive value showing the concentration and negative value showing the de-concentration. Thus the negative values appeared in inverse pole figures represented by $\Delta G(\xi, \phi)$ do not mean the negative distribution density in this paper. In Fig. 10(a), the values of $\Delta G(\xi, \phi)$ are shown by contours at an 1% interval on the inverse pole figures. In order to obtain $\Delta G(\xi, \phi)$ in percent, we set $A_{00} \equiv 0$ and three independent coefficients A_{40} , A_{60} and A_{80} be multiplied by $(4\pi) \times 100$ instead of the values in Table 6.

3. Practical procedure of pole figure inversion on trevorite aggregates

In this paper we describe the actual procedure of pole figure inversion in the experi-

mentally produced tectonites consisted of spinel type minerals. We have to consider many technical problems originated from the nature of samples, the condition of preparing tectonites, X-ray measurements, data processing, estimation of the reliability in the final result, etc.

1 General comments on the axial symmetry and uniformity of OD

There should have been non-uniform distribution of stress and temperature in a specimen during the experimental deformation. Thus the non-uniformity of crystallographic OD and the deviation of OD from an expected axial symmetry are likely to take place throughout the deformed specimen. Such non-uniformity of textures exists certainly in the present specimens (see Fig. 11) and also in the deformed flint (BAKER *et al.*, 1969). However, the texture is supposed to be uniform at the central part of specimen and the OD is of axial symmetry as a consequence of the geometric setting of the specimen in the present experimental device.

The measurement on larger volume of the specimen enables us to make an efficient pole figure goniometry, while the smaller volume guarantees the better uniformity and axial symmetry of OD. As a compromise of the two opposing requirements in the above, we have chosen the central part 1/3 of the specimen along the specimen axis for the measurement of OD in this work. When we carefully examine the reported local variation of quartz orientation in a deformed flint (BAKER *et al.*, 1969) for example, our choice, a 1/3 fraction of the specimen, seems adequate.

2 Sample preparation

(a) Sample

In this paper, we describe the practical procedure of pole figure inversion for four specimens (HP 6, TD 8101 (annealed AHP), TW 8102 and F 1221) which are consisting of cubic crystallites and having uniaxial OD. Two of the four are experimentally deformed specimens at high temperature and the others are undeformed ones.

The material of the specimen is an aggregate of synthetic trevorite NiFe_2O_4 ;

a spinel type mineral. Two kinds of starting samples (HP 6 and AHP) which have been produced under the different conditions, are subjected to deformation experiments. The HP 6 having less than 5% porosity has been prepared by the usual hot-press technique. This polycrystalline material is prepared for the starting samples of the deformation experiments at atmospheric pressure and high temperature. Another type of aggregate (AHP) has been made by the activation hot-press technique by which both synthesizing and sintering of a trevorite aggregate are made at the same time. This material having about 10% porosity is prepared for the starting samples of the deformation experiments at high pressure and temperature. Both starting samples are produced under the uniaxially compressional condition, so the OD in each sample is expected to show a uniaxial symmetry. The grain size is less than $5\mu\text{m}$ and the grain shape is equant in both samples, according to the microscopic observation of polished and etched surface of the specimens.

The sample AHP has larger porosity ($\sim 10\%$), and the OD in the sample has a possibility of change at elevated temperatures even under hydrostatic condition. Therefore, the AHP is annealed at $1,000^\circ\text{C}$ during 30 min. under the hydrostatic pressure of 6 kbar (0.6 GPa) in an apparatus described in the previous papers (IDA *et al.*, 1971; TSUKAHARA *et al.*, 1976; TSUKAHARA, 1974). This annealed specimen is named TD8101.

The ODF of the specimens, designated as HP 6 and TD 8101, are prepared for the comparison with ODF of experimentally deformed specimens.

Two experimentally deformed specimens TW 8102 and F 1221 are recrystallized AHP and HP 6, respectively, under uniaxial compression at high temperature by means of the same apparatus. These specimens show an axially symmetric cylinder with bulging.

The physical conditions for preparation of TW 8102, F 1221 and TD 8101 are shown in Table 2.

(b) Shaping and X-ray shielding

A thin section (oriented as shown in Fig. 6)

Table 2

Experimental conditions						
run no.	size (mm)		pressure (bar)	temperature (°C)	total strain (%)	strain rate (1/sec)
	l	d				
TD8101	8.85	9.15	6000	1000	0	0
TW8102	8.95	9.00	6000	1000	20.7	2.24×10^{-4}
F1221	12.69	10.06	1	1265	26.6	1.59×10^{-6}

l ; length, d ; diameter

Table 3

Run No.	l	Measured crystallographic planes (hkl)					
TW8102	8	(311)	(222)	(400)	(422)	(511.333)	(440)
F1221(A)	6	(311)	(222)	(400)	(422)	(511.333)	(440)
F1221(B)	6	(311)	(222)	(400)	(422)	(511.333)	(440)
TD8101	6	(311)	(222)	(400)	(422)	(511.333)	(440)
HP6	4	(311)	(222)	(400)	-	-	(440)

Fig. 6. Illustration showing the cylindrical specimen, the specimen coordinates o-xyz and the orientation of a thin section cutout for X-ray measurements.

is cut out of the polycrystalline specimen to measure the OD with an X-ray transmission method. The surfaces of the thin section were polished by abrasive powder to make uniform thickness. The thickness of the thin section is about 0.1 mm which was determined as a compromise between smaller absorption of X-ray and sufficient rigidity for easy handling.

A shielding plate (0.3 mm thick aluminum) with an aperture (2~4 mm in diameter) at the center, is employed in order to hold the thin section and also to restrict the X-ray irradiation to the central part of the specimen. Depending on the area to be subjected to pole figure goniometry, we have chosen the different diameter of an aperture on an aluminum plate.

3 Methods of measurements

(a) X-ray transmission method

A plane normal orientation distribution is determined usually by combining both transmission and reflection methods. However, we can obtain the plane normal orientation distribution only by a transmission method (DECKER *et al.*, 1948) with a thin section

sample oriented as shown in Fig. 6, because OD is axially symmetric around the longitudinal axis of the deformed cylindrical specimen in the present case.

The thin section is mounted with adhesive on the specimen holder which also works as a shielding and intercepting device for X-ray beam. We place the specimen normal at an angle of diffraction, placing the thin section perpendicular to the plane of an X-ray beam and an X-ray detector. Then we collect the integrated X-ray intensity data by a proportional counter as a function of the orientation (χ_i) of the specimen being rotated in its own plane around the specimen normal.

The absorption correction is not necessary in this study, since the length of the X-ray path in the specimen is kept constant during the intensity measurements on one crystallographic plane.

(b) Step scanning and fixed time modes

We have used step scanning (at intervals of 5 deg.) and fixed time modes (10 sec. duration for X-ray generating power of 40 Kv, 75 mA) instead of continuous acquisition of intensity data in order to make the

data processing simple.

We collected the X-ray intensity data (both diffraction intensity and background) covering the whole possible range $0^\circ \leq \chi_i \leq 360^\circ$ at every selected angle (2θ). The necessary data to yield a complete pole figure (plane normal orientation distribution) are the intensity profiles in the range of $0^\circ \leq \chi_i \leq 90^\circ$ (see Appendix C). The profiles are derived by superposing the four equivalent quadrants.

The overall error of profile data is statistically evaluated as less than 1.5% based on the acquired number ($\approx 5,000$) of photons in one step of scanning.

(c) Angular oscillation

For coarse-grained specimens (grain size $\geq 10 \mu\text{m}$), the number of crystallites having a particular orientation is so small that the satisfactory counting statistics are not provided. It is necessary to bring more than 10^4 grains into the diffraction orientation. In order to increase the effective number of grains to be measured, two techniques are known. One of the techniques employs vibrational translation of the specimen kept in a particular orientation on the goniometer. Although the device for this is standard in the commercial pole figure device, this technique can not be used for the small measuring area available in our specimens, but for a large homogeneous specimen. Therefore, we have introduced another procedure; an angular oscillation method (BAKER *et al.*, 1969). This is achieved by an angular oscillation of specimen orientation within a limited range (± 2.5 deg.), while the diffraction angle (2θ) is kept constant. In this method, the satisfactory counting statistics are obtained with the crystallites having the orientations slightly out of the diffraction condition at the particular specimen orientation.

(d) Pole figure goniometer

We used an X-ray power unit of a rotating anode type (Rigaku Rota Flex RU-3SH), and a commercial pole figure goniometer (Rigaku semi-automatic pole figure goniometer) on which an angular oscillation device developed in our laboratory is attached.

The angular oscillation mechanism is composed of two sets of electromagnetic vibra-

Fig. 7. An X-ray pole figure goniometer with the angular oscillation device.

tors driven by means of two independent triangle-shaped current supplies (~ 1 Hz and ~ 10 Hz). The triangular vibrations are transferred to the specimen holder with two-axis freedom through a couple of arms (see Fig. 7). The specimen vibration is swept in a square solid angle of $5^\circ \times 5^\circ$ by controlling the amplitudes of two electric power units.

(e) Selection of diffraction planes

All the crystallographic planes used for measurements are listed in Tables 3 and 4. The usual powder diffraction chart of the starting sample is shown in Fig. 8. The arrows in Fig. 8 indicate the positions where the background data are collected.

When several diffraction planes having the same crystallographic orientation are observed within an available range of 2θ , a more effective plane to secure the accurate ODF should be selected on a basis of the background levels. We selected (440) plane in a set of (220) and (440). Among (111), (222) and (333), we employed (222) and also (333) which is a composite peak (511-333) (see section 3.4 (b) and Table 4).

4 Data processing

(a) Effective intensity data

The effective intensity data for the i th diffraction peak $I_i(\zeta_i)$ is derived from the observed data of diffraction intensity $I_{i, \text{obs}}(\zeta_i)$ by subtracting background intensity $I_{i, \text{bg}}(\zeta_i)$.

$$I_i(\zeta_i) = I_{i, \text{obs}}(\zeta_i) - I_{i, \text{bg}}(\zeta_i)$$

The background intensity is evaluated by a linear interpolation of background data observed at just aside the diffraction angle.

Fig. 8. A powder diffraction chart of the starting material.

Table 4

Parameters for the plane normal orientation distributions measured and synthesized						
i	(hkl)	2θ Feka	θ_i (°)	ϕ_i (°)	weight	
					ρ_i	C_{ij}
1	311	45.31	72.45	18.43	1.0	
2	222	47.40	54.74	45.00	1.0	
3	400	55.32	90.00	0.0	1.0	
4	422	69.30	65.91	26.57	1.0	
5	511	74.18	78.90	11.31	1.0	0.7
	333	74.18	54.74	45.00		0.3
6	440	81.96	90.00	45.00	1.0	
7	210		90.00	26.57		
8	221		48.19	63.43		
9	321		36.70	63.43		

Table 5

Expansion coefficients Q_k^i of measured intensity data and the mean square deviation σ_{q_i} (eq. (10)) of sample TW8102						
k	(311)	(222)	(400)	(422)	(511·333)	(440)
0	1.414	1.414	1.414	1.414	1.414	1.414
2	6.938×10^{-3}	-3.451×10^{-3}	2.196×10^{-3}	1.127×10^{-2}	-2.797×10^{-3}	-1.123×10^{-2}
4	-2.767×10^{-3}	-5.906×10^{-3}	1.269×10^{-2}	1.229×10^{-2}	-1.471×10^{-3}	-8.857×10^{-3}
6	-6.852×10^{-3}	-7.770×10^{-2}	-3.908×10^{-2}	-4.448×10^{-2}	-2.512×10^{-2}	5.763×10^{-2}
8	-1.380×10^{-2}	1.308×10^{-2}	1.874×10^{-2}	-1.258×10^{-3}	-8.451×10^{-5}	1.531×10^{-2}
σ_{q_i}	1.7%	8.8%	3.5%	9.8%	2.5%	2.3%

The effective intensity data $I_i(\zeta_i)$ are normalized to get the plane normal orientation distribution function $q_i(\zeta_i)$ (see Appendix C).

(b) *Weighting factors*

There are two kinds of conceivable weighting factors: One is related to the accuracy of an individual pole figure, i.e., ρ_i in eq. (6), and the other is a relative intensity factor of an individual diffraction peak in a composite peak, i.e., C_{ij} in eqs. (4) and (6). The weighting factors (ρ_i) relating to the accuracy are required in the step of calculating $A_{\epsilon m}$ in eq. (6), because the accuracy of measured pole figures ought to be different each other. KRIGBAUM & ROE (1964) made use of the subjectively assigned weighting factors which were estimated with background level. However, it is difficult to evaluate the objective weighting factors, because there are many kinds of factors (e.g., polarization, absorption, multiplicity, structure, and temperature factors) to be considered for estimating the exact intensity profile. After all, we considered the measured pole figures to have the same accuracy, and their weighting factors (ρ_i) are set unity.

Both (511) and (333) diffractions of trevorite take place as one composite peak on a diffraction profile (Fig. 8), because $h^2 + k^2 + l^2 = 5^2 + 1^2 + 1^2 = 3^2 + 3^2 + 3^2 = 27$. Therefore, we have to know fractional value of C_{ij} in eq. (4) for (511) and (333). The relative intensity factors of (511) and (333) are calculated by using the scattering factors (MAEGILLAVERG & RIECK, 1962) and u -parameter which describes the positions of oxygen atoms in a unit cell of a spinel type structure;

$$I_{hkl} = |\mathbf{F}|^2 P [(1 + \cos^2 2\theta) / (\sin^2 \theta \cos \theta)]$$

$$\mathbf{F}_{hkl} = \sum_{n=1}^N \mathbf{f}_n \exp [2\pi i (hu_n + kv_n + lw_n)]$$

where I_{hkl} is the diffraction intensity with Miller indices; hkl , $\mathbf{F}$ is structure factor, $\mathbf{f}_n$ is atomic structure factor, P is multiplicity, θ is Bragg angle, and u_n , v_n and w_n are position of n th atom in the lattice.

According to HASTINGS & CORLISS (1953), u -parameter of trevorite is 0.38 ± 0.003 . The calculated ratio of intensity value of (511)

diffraction to that of (333) diffraction $I_{511} : I_{333}$ is 0.69:0.31 at $u=0.38$ and is 0.68:0.32 at $u=0.37$. We employed the weighting factors 0.7 for (511) and 0.3 for (333) in solving eq. (4) (see Table 4).

(c) *Error estimation*

The two steps of reliability checks are made in the present work. As the first step, a check is made on a set of expansion coefficients Q_i^l , which are affected by errors originated from the measurements of the plane normal distribution functions $q_i(\zeta_i)$ and also by the termination effect of spherical harmonic series. We, therefore, compare $q_i(\zeta_i)$ with $q_i'(\zeta_i)$ which is synthesized by

$$q_i'(\zeta_i) = \sum_{l=0}^{\lambda} Q_i^l \tilde{P}_l(\zeta_i) \quad (l \leq \lambda) \quad (8)$$

where λ is the largest value of l in the actual harmonic expansion. An appropriate parameter indicating the quality of eq. (8) may be the mean square deviation $\sigma_{q_i'}$ of the series approximation of $q_i'(\zeta_i)$ from the measured $q_i(\zeta_i)$;

$$\sigma_{q_i'} = \int_{-1}^1 [q_i'(\zeta_i) - q_i(\zeta_i)]^2 d\zeta_i \quad (9)$$

An example of $\sigma_{q_i'}$ is shown in Table 5.

As the second step, reliability of $A_{\epsilon m}$ calculated from Q_i^l by eqs. (5) and (6) is made. The Q_i^l are calculated by eq. (3) from the values of $A_{\epsilon m}$ (Q_i^l thus calculated is denoted by $Q_i^{l'}$), and the comparison is made for the observed $q_i(\zeta_i)$ with $q_i''(\zeta_i)$ which are synthesized with $Q_i^{l'}$, by the same procedure as eq. (8). The mean square deviations $\sigma_{q_i''}$ between $q_i''(\zeta_i)$ and $q_i(\zeta_i)$;

$$\sigma_{q_i''} = \int_{-1}^1 [q_i''(\zeta_i) - q_i(\zeta_i)]^2 d\zeta_i$$

would provide a measure of the reliability of $A_{\epsilon m}$ in reference to the measured pole figures (Table 7).

The evaluation and representation of the overall errors in the ODF is made by utilizing the standard deviations $\sigma_{A_{\epsilon m}}$ of $A_{\epsilon m}$ in the least squares calculation by eq. (6). The $\sigma_{A_{\epsilon m}}$ is essentially a measure of the internal inconsistencies among the original X-ray pole figures, which are not independent each other.

When all the requirements on $W_{\ell m}$ (Table 1) are taken into account, eq. (1) is written simply by

$$w(\xi, \phi) = \sum_{\ell=0}^{\lambda} [A_{\ell 0} \tilde{P}_{\ell}(\xi) + 2 \sum_{m=4}^{\ell} A_{\ell m} \tilde{P}_{\ell}^m(\xi) \cos m\phi]$$

Thus the standard deviation σ_w of $w(\xi, \phi)$ is given by

$$\sigma_w^2(\xi, \phi) = \sum_{\ell=0}^{\lambda} [(\tilde{P}_{\ell}(\xi))^2 \sigma_{A_{\ell 0}}^2 + \sum_{m=4}^{\ell} (2\tilde{P}_{\ell}^m(\xi) \cos m\phi)^2 \sigma_{A_{\ell m}}^2]$$

which is a function of ξ and ϕ , and is represented on an inverse pole figure. In accordance with the graphical representation of pole figure by $G(\xi, \phi)$ or $\Delta G(\xi, \phi)$, the corresponding error estimation of $G(\xi, \phi)$ is given by

$$\sigma_G^2(\xi, \phi) = 4\pi \sigma_w^2(\xi, \phi) \quad (10)$$

(d) *Order of harmonic expansion of $q_i(\zeta_i)$ and $w(\xi, \phi)$*

The termination of spherical harmonic expansion of ODF to the limited order number is necessary for the practical application. The maximum value of ℓ of an expansion coefficients $A_{\ell m}$ for ODF is equal to or smaller than that of Q_i^{ℓ} for the plane normal orientation distribution $q_i(\zeta_i)$, as clarified in eq. (3).

Each $q_i(\zeta_i)$ usually includes the high order terms due to counting statistical fluctuation. In order to reduce the effect, we terminated the expansion of $q_i(\zeta_i)$ instead of smoothing the intensity data. For this purpose, we made use of $q_i(\zeta_i)$ expanded up to 8th order. We regarded $q_i(\zeta_i)$ to be specified sufficiently by the numerical parameters Q_i^{ℓ} resulting from harmonic expansion up to $\ell=8$ as shown in Fig. 9 and Table 5. There are nineteen data points to describe $q_i(\zeta_i)$ in the range $0 \sim 90^\circ$, since data acquisition is made by a step scanning mode at an interval of 5 deg. The wave number of the Legendre polynomial $P_{\ell}(\cos \chi_i)$ in the range $0 \sim 90^\circ$ is $\ell/4$. If it is necessary to have more than four data points in one wave, the maximum allowed integer for ℓ is equal to or less than 8.

For every specimen, we calculated ODF of three ranks of expansion, i. e., expansions

up to 4th, 6th and 8th order, to seek the most suitable ODF by comparing $q_i(\zeta_i)$ with $q_i''(\zeta_i)$.

4. Experimental results

1 Results on the specimen TW 8102

For the specimen TW 8102, six sets of plane normal orientation distributions $q_i(\zeta_i)$ were determined from intensity measurements consisting of both six sets of diffraction intensity data and four sets of background data. The six sets of $q_i(\zeta_i)$ are plotted with open circles in Fig. 9 (a~f). Expansion coefficients Q_i^{ℓ} determined by eq. (A8) from $q_i(\zeta_i)$ are listed in Table 5.

Then the functions $q_i'(\zeta_i)$ were calculated from Q_i^{ℓ} by eq. (2) including terms up to 8 (with $Q_i^0=0$). (The reconstructed $q_i'(\zeta_i)$ are not plotted in Fig. 9.) The standard deviations $\sigma_{q_i'}$ (eq.(9)) of the series approximations $q_i'(\zeta_i)$ from the original data are calculated and shown in Table 5. These values $\sigma_{q_i'}$ suggest that the five-term series (i.e., including ℓ up to 8) is sufficient to represent all the measured data.

The values Q_i^{ℓ} can not be affected by the different ℓ th order coefficients, thereby we regarded the other ℓ th order coefficients (Q_i^0 , Q_i^4 and Q_i^8) as available.

The necessary and sufficient numerical parameters $A_{\ell m}$ to represent ODF are determined from Q_i^{ℓ} with the least squares method by eq. (3), and are listed in Table 6. The values of standard deviation for each coefficient $A_{\ell m}$ are also shown in order to know the reliability of ODF.

Once $A_{\ell m}$ are determined, we can calculate $Q_i^{\ell'}$ from $A_{\ell m}$ with eq. (3) and/or eq. (4) (Tables 7 and 8), and synthesize the plane normal orientation distribution $q_i''(\zeta_i)$ for any crystallographic orientation by using eq. (2). The ten sets of $q_i''(\zeta_i)$ are calculated for the nine crystallographic orientations listed in Table 4 ($i=1 \sim 9$), and for one composite peak (511·333). The solid lines in Fig. 9 (a~j) show the $q_i''(\zeta_i)$.

The values of synthesized $q_i''(\zeta_i)$ include all the contribution from the measurements on six diffraction peaks. Judging from both the values of $\sigma_{q_i''}$ and the degree of fit in

Fig. 9. Plane normal orientation distributions of TW8102 plotted against χ_i . Open circles (a~f); measured diffraction intensity data $q_i(\zeta_i)$ used in the analysis. Solid lines (a~j); synthesized plane normal orientation distribution $q'_i(\zeta_i)$ from ODF. Both the values of $q_i(\zeta_i)$ and $q'_i(\zeta_i)$ are normalized to unity when the orientation distribution is random or uniform.

Fig. 9, we can regard the measurements for all diffraction peaks as adequate. In other words, resulting ODF is reliable.

Using eq. (1) and the coefficients A_{i0} obtained from the data for all six diffraction

peaks, we calculated the value of $\Delta G(\xi, \phi)$ which retains the terms with ϵ up to 8. The error estimation of the resulting $\Delta G(\xi, \phi)$ is made from the values of standard deviation for A_{i0} . In Fig. 10, the $\Delta G(\xi, \phi)$ and its

estimated standard deviation are shown. Figure 10(a) shows that $\langle 110 \rangle$ -axis prefers to be oriented parallel to the z -axis (the maximum compression axis), while the other two $\langle 111 \rangle$ - and $\langle 100 \rangle$ -axes do not. These remarkable patterns of OD for three typical crystallographic orientations are considered to be real nature of preferred orientation (PO).

2 Results on the other specimens

The PO in the samples HP 6 and TD 8101 are so weak that the presence of any valid degree of OD is not claimed on the basis of comparison with their accuracy of ODF (Figs.

12 and 13). On the other hand, a considerable degree of PO are certainly observed in the samples F1221 and TW 8102, which are experimentally recrystallized HP 6 and AHP, respectively. The ODF thus determined are certainly regarded as those generated by the syntectonic recrystallization.

In order to know the extent of the non-uniformity of crystallographic OD in a specimen, we have determined the ODF of the specimen F1221 at the two portions (Fig. 11); (A) the central portion and (B) the extreme end portion of the specimen. As shown in Fig. 11, we can easily recognize that the OD for each portion are completely different each other; the $\langle 111 \rangle$ -axis is statistically parallel to z -axis of the specimen at the central portion (A), while at the end portion (B) $\langle 110 \rangle$ -axis tends to be parallel to z -axis. This fact indicates that trevorite has at least two types of PO depending on stress, temperature, strain rate, total strain, or some other factors.

Table 6

Expansion coefficients of ODF of sample TW8102		
l	A_{l0}	Standard deviation
0	1.13×10^{-1}	0.0
2	0.0	6.80×10^{-4}
4	-1.44×10^{-4}	9.29×10^{-4}
6	-3.81×10^{-3}	6.59×10^{-4}
8	1.54×10^{-3}	5.39×10^{-4}

Table 7

Expansion coefficients $Q_l^{i'}$ of synthesized intensity and the mean square deviation σ_{Q_i}'' (eq. (12)) of sample TW8102						
l	(311)	(222)	(400)	(422)	(511+333)	(440)
0	1.41	1.41	1.41	1.41	1.41	1.41
2	0.0	0.0	0.0	0.0	0.0	0.0
4	-3.88×10^{-4}	1.20×10^{-3}	-1.81×10^{-3}	4.52×10^{-4}	-4.59×10^{-4}	4.52×10^{-4}
6	-6.33×10^{-3}	5.74×10^{-3}	-4.79×10^{-2}	-2.46×10^{-2}	-1.21×10^{-2}	7.78×10^{-2}
8	-1.11×10^{-2}	5.74×10^{-3}	1.94×10^{-2}	-7.76×10^{-3}	1.62×10^{-3}	1.09×10^{-2}
σ_{Q_i}''	2.4%	8.9%	5.8%	12%	4.1%	6.1%

Table 8

Expansion coefficients $Q_l^{i'}$ of synthesized intensity of Sample TW8102				
l	(210)	(221)	(321)	(511)
0	1.41	1.41	1.41	1.41
2	0.0	0.0	0.0	0.0
4	-3.61×10^{-4}	8.70×10^{-4}	4.52×10^{-4}	-1.17×10^{-4}
6	3.25×10^{-2}	-2.03×10^{-2}	5.25×10^{-3}	-1.97×10^{-2}
8	-4.18×10^{-3}	4.49×10^{-3}	-2.32×10^{-3}	-1.39×10^{-4}

Fig. 10. The inverse pole figures represented by $\Delta G(\xi, \phi)$ of TW 8102 and its standard deviation $\sigma_G(\xi, \phi)$ (eq. (10)). (a) The contours of $\Delta G(\xi, \phi)$ show the deviation in percent of orientation density from the random or uniform orientation. (b) The contours show the standard deviation in the same unit as $\Delta G(\xi, \phi)$.

Fig. 11. The inverse pole figures represented by $\Delta G(\xi, \phi)$ of F1221 and its standard deviation. (A) $\Delta G(\xi, \phi)$ at the central portion of F1221 and (B) $\Delta G(\xi, \phi)$ at the end portion of F1221. $\Delta G(\xi, \phi)$ are described as the same way as in Fig. 10(a). (a) $\sigma_G(\xi, \phi)$ at the central portion and (b) $\sigma_G(\xi, \phi)$ at the end portion.

5. Summary and discussion

A practical method has been established for the complete determination and representation of OD in the uniaxial tectonites consisting of cubic crystallites by the X-ray pole figure inversion. The technical details including

Fig. 12. The inverse pole figures representing (a) $\Delta G(\xi, \phi)$ of the sample HP6, and (b) the standard deviation.

Fig. 13. The inverse pole figures representing (a) $\Delta G(\xi, \phi)$ of the sample TD 8101 and (b) the standard deviation.

experimental device and data precessing are described in this paper.

One problem remained for further study is the preparation of the really good specimen having an axial symmetry appropriate for the present pole figure inversion technique. The acquired numerical coefficients Q_i^j by the harmonic analysis of X-ray diffraction data were not zero in the present case, while they are required to be zero for cubic crystallites in a specimen of axial symmetry.

The preferred orientation of spinel type minerals has scarcely been studied. In their studies of mechanical properties of cubic ferrite $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ (spinel type structure), NISHIKAWA *et al.* (1974) mentioned on the preferred orientation in polycrystalline specimens which were uniaxially deformed by 45% at 1,470°C. They claimed that OD was not recognized on the basis of peak-height comparison of X-ray diffraction intensities measured on the surfaces parallel and perpendicular to the compression axis of the deformed specimen. In the present case, however, the noticeable OD is generated in

the specimen deformed only by 20—30%. One interpretation on the difference between $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ and NiFe_2O_4 is that both materials are really different in the properties of textural development under stressed condition. However, we suppose that the degree of OD was so slight in $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ of NISHIKAWA *et al.* (1974) and their simple technique did not allow them to detect the OD which has actually been generated. Since the pole figure inversion technique is a kind of harmonic analysis, it enables us to detect the preferred orientation having very low degree. In other words, we can deduce a useful information from the data with a small S/N by means of X-ray pole figure inversion technique. Actually the degree of the OD detected in the present work is several percents.

The present results on the preferred orientation of trevorite developed in the experimental tectonites are quite preliminary. However,

we can claim that there are at least two different preferred orientation types; $\langle 111 \rangle //$ compression, and $\langle 110 \rangle //$ compression. The occurrence of these two types is certainly depending on the physical condition under which the OD is generated. The cause of OD and its dependency on the physical conditions are the next problem to be studied (FUJIMURA, in preparation) in order to clarify the structural development in the spinel layer in the upper mantle.

Acknowledgement

We would like to thank Mr. Tatsuo YAMAZAKI, Ceramic Research Institute, Aichi Prefecture, for offering us the samples HP6. The numerical computations were carried out with a FACOM 230—60 computer at the Computation Center of Nagoya University (Problem Nos. 4001DI1299, 4001 EC 8°652 and 4001 FC 8°637).

Appendix

A) Relationship among $(\chi_i, \eta_i), (\Theta_i, \Phi_i)$ and (ϕ, θ, ϕ) (χ_i, η_i) is related to (Θ_i, Φ_i) through

$$(\phi, \theta, \phi) \text{ as } \begin{pmatrix} \sin \chi_i \cos \eta_i \\ \sin \chi_i \sin \eta_i \\ \cos \chi_i \end{pmatrix} = T(\phi, \theta, \phi) \begin{pmatrix} \sin \Theta_i \cos \Phi_i \\ \sin \Theta_i \sin \Phi_i \\ \cos \Theta_i \end{pmatrix} \quad (\text{A1})$$

where

$$T(\phi, \theta, \phi) = \begin{pmatrix} \cos \phi \cos \theta \cos \phi - \sin \phi \sin \phi & -\cos \phi \cos \theta \sin \phi - \sin \phi \cos \phi & \cos \phi \sin \theta \\ \sin \phi \cos \theta \cos \phi + \cos \phi \sin \phi & -\sin \phi \cos \theta \sin \phi + \cos \phi \cos \phi & \sin \phi \sin \theta \\ -\sin \theta \cos \phi & \sin \theta \sin \phi & \cos \theta \end{pmatrix}$$

The vectors in both side of eq. (A1) are components of the unit vector $\mathbf{n}_i$ with respect to o-xyz and O-XYZ, respectively. $T(\phi, \theta, \phi)$ is the matrix which represents the transformation between coordinate systems o-xyz and O-XYZ.

If the specimen has a cylindrical symmetry around z-axis and the preferred orientation is independent of ϕ as in the present case, it is sufficient to reduce eq.(A1) to

$$\begin{aligned} \cos \chi_i &= \cos \theta \cos \Theta_i \\ &+ \sin \theta \sin \Theta_i \cos [\phi - (\pi - \Phi_i)] \end{aligned} \quad (\text{A2})$$

B) Representation of ODF and spherical harmonic expansion

Since the function $w(\xi, \phi)$ is defined as a density distribution function, it satisfies the normalization condition;

$$\int_0^{2\pi} \int_{-1}^1 w(\xi, \phi) d\xi d\phi \equiv 1 \quad (\text{A3})$$

The function $w(\xi, \phi)$ can be expanded by spherical harmonics;

$$w(\xi, \phi) = \sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} W_{\ell m} \tilde{P}_{\ell}^m(\xi) \exp(-im\phi) \quad (1)$$

where $\tilde{P}_{\ell}^m(\xi)$ ($\ell=0, 1, 2, \dots, \infty, m=-\ell, -\ell+1, \dots, \ell-1, \ell$) are the normalized associated Legendre polynomials;

$$\tilde{P}_\ell^m(\xi) = \{[(2\ell+1)(\ell-m)!] / [2(\ell+m)!]\}^{1/2} P_\ell^m(\xi) \quad (A4)$$

and $W_{\ell m}$ are the expansion coefficients;

$$W_{\ell m} = [1/(2\pi)] \int_0^{2\pi} \int_{-1}^1 w(\xi, \phi) \tilde{P}_\ell^m(\xi) \times \exp(im\phi) d\xi d\phi \quad (A5)$$

C) *Normalization of the collected X-ray intensity data and harmonic expansion*

Let the intensity data of X-ray diffraction of the i th crystallographic plane be $I_i(\zeta_i)$ which are collected in the all possible range $0^\circ \leq \chi_i \leq 360^\circ$ on the sample coordinates o -xyz. In the data analysis, however, it is necessary to use a plane normal orientation distribution function $q_i(\zeta_i)$ (i.e., a normalized pole figure of one dimension) in the range $0^\circ \leq \chi_i \leq 180^\circ$ or $-1 \leq \zeta_i \leq 1$;

$$q_i(\zeta_i) = I_i(\zeta_i) / \int_{-1}^1 I_i(\zeta_i) d\zeta_i \quad (A6)$$

when $q_i(\zeta_i)$ is defined by eq.(A6),

$$\int_{-1}^1 q_i(\zeta_i) d\zeta_i = 1$$

and

$$Q_0^i = 1/\sqrt{2}$$

In this case, the random or uniform orientation distribution is represented by

$$q_i(\zeta_i) = 1/2$$

The normalization by eq.(A6) is adopted in eqs. (3), (4), (5), (6) and (A10).

Another type of normalization is comprehensible for the practical use;

$$q_i(\zeta_i) = I_i(\zeta_i) / \int_0^1 I_i(\zeta_i) d\zeta_i \quad (A6')$$

because eq.(A6') yields

$$q_i(\zeta_i) = 1$$

for the random or uniform orientation distribution. When $q_i(\zeta_i)$ is defined by eq.(A6'),

$$\int_{-1}^1 q_i(\zeta_i) d\zeta_i = 2$$

and

$$Q_0^i = \sqrt{2}$$

When the normalization by eq.(A6') is used, the factor $2\pi [2/(2\ell+1)]^{1/2}$ in eqs. (3), (4), (5), (6) and (A10) should be replaced by $4\pi [2/(2\ell+1)]^{1/2}$.

Although ROE & KRIGBAUM(1964) has employed (A6), the use of (A6') is recommended for the practical application from the following rea-

son. A measured X-ray profile has a mirror symmetry at $\chi_i = 0^\circ (180^\circ)$ and $\chi_i = 90^\circ (270^\circ)$, because of the Friedel's law ($q_i(\zeta_i)$ is equal to $q_i(-\zeta_i)$) and axial symmetry of OD. Therefore, the independent data of the X-ray intensity profiles actually required to yield a plane normal orientation distribution function $q_i(\zeta_i)$ are those in the range $0^\circ \leq \chi_i \leq 90^\circ$ or $0 \leq \zeta_i \leq 1$. Therefore, we use the normalization by eq.(A6') in section 3 and later in this paper except Appendix D.

We can expand $q_i(\zeta_i)$ in a series of normalized Legendre polynomials;

$$q_i(\zeta_i) = \sum_{\ell=0}^{\infty} Q_\ell^i \tilde{P}_\ell(\zeta_i) \quad (2)$$

where

$$\tilde{P}_\ell(\zeta_i) = [(2\ell+1)/2]^{1/2} P_\ell(\zeta_i) \quad (\ell=0, 1, 2, \dots, \infty) \quad (A7)$$

is a series of normalized Legendre polynomials, and

$$Q_\ell^i = \int_{-1}^1 q_i(\zeta_i) \tilde{P}_\ell(\zeta_i) d\zeta_i \quad (\ell=0, 1, 2, \dots, \infty) \quad (A8)$$

are numerical coefficients. The coefficients Q_ℓ^i are equal zero for odd ℓ , because $q_i(\zeta_i)$ is an even function.

D) *Relationship between X-ray diffraction data $q_i(\zeta_i)$ and ODF $w(\xi, \phi)$*

We make ODF be represented by X-ray diffraction measurements, i.e., plane normal orientation distributions $q_i(\zeta_i)$, using the relationship among three sets of angles (θ_i, Φ_i, χ_i and (θ, ϕ)).

Let the Legendre addition law be applied to eq.(A2) for the description of $\tilde{P}_\ell(\zeta_i)$ in eq.(2) and eq.(A8) as a function of θ, θ_i and Φ_i . Then we have

$$\begin{aligned} \tilde{P}_\ell(\cos \chi_i) &= [2/(2\ell+1)]^{1/2} \\ &\times \sum_{m=-\ell}^{\ell} \tilde{P}_\ell^m(\cos \theta) \tilde{P}_\ell^m(\cos \theta_i) \exp[i m(\phi + \Phi_i - \pi)] \end{aligned} \quad (A9)$$

When we multiply both sides of eq.(A9) by $q_i(\zeta_i) w(\xi, \phi)$ and integrate over whole range of ζ_i, ξ and ϕ , we obtain the following form;

$$\begin{aligned} Q_\ell^i &= 2\pi [2/(2\ell+1)]^{1/2} \\ &\times \sum_{m=-\ell}^{\ell} (-1)^m w_{\ell m} \tilde{P}_\ell^m(\xi) \exp(im\Phi_i) \end{aligned} \quad (A10)$$

As $\tilde{P}_\ell^m(\xi) = (-1)^m \tilde{P}_\ell^{\bar{m}}(\xi)$ ($-m = \bar{m}$; m : positive), and $W_{\ell \bar{m}}$ equals to a conjugate of $W_{\ell m}$, we have

$$Q_i^1 = 2\pi [2/(2i+1)]^{1/2} \\ \times \{A_{i,m} \tilde{P}_i(\mathcal{E}_i) + 2 \sum_{m=1}^i (-1)^m [A_{i,m} \tilde{P}_i^m(\mathcal{E}_i) \cos m\Phi_i \\ - B_{i,m} \tilde{P}_i^m(\mathcal{E}_i) \sin m\Phi_i]\} \quad (3)$$

References

- BAKER, D.W., 1969: X-ray analysis and representation of preferred orientations in crystal aggregates. Ph.D. dissertation, Univ. California, Los Angeles (Univ. Microfilms, Ann Arbor, Mich., no. 70—14257), 160p.
- and RIEKELS, L. M., 1977: Dauphiné twinning in quartzite mylonite. *Jour. Geol.*, **85**, 15—26.
- , WENK, H. R. and CHRISTIE, J. M., 1969: X-ray analysis of preferred orientation in fine grained quartz aggregates. *Ibid.*, **77**, 144—172.
- and ———, 1972: Preferred orientation in a low-symmetry quartz mylonite. *Ibid.*, **80**, 81—105.
- BUNGE, H.J., 1965: Zur Barstellung allgemeiner Texturen. *Zeitschr. Metallkunde*, **56**, 872—874.
- CARTER, N. L., 1976: Steady state flow of rocks. *Rev. Geophys. Space Physics*, **14**, 301—360.
- DECKER, B.F., ASP, E.T. and HARKER, D., 1948: Preferred orientation using a geiger counter X-ray diffraction goniometer. *Jour. Appl. Phys.*, **19**, 388—392.
- GOEMAN, U. and SCHUMANN, H., 1975: Roentgenographische Messungen zur Teilbarkeit des Granat. *Neues Jahrb. Min. Abhdl.*, **125**, 91—105.
- HASTINGS, J.M. and CORLISS, L.M., 1953: Neutron diffraction studies of zinc ferrite and nickel ferrite. *Rev. Mod. Phys.*, **25**, 114—121.
- IDA, K., TSUKAHARA, H., KUMAZAWA, M., KOBAYASHI, Y., SUZUKI, I. and KASAHARA, J., 1971: Deformation of dunite at slow strain rates under high temperature and pressure. *Jour. Soc. Material Sci. Japan*, **20**, 179—184. (in Japanese with English abstract)
- KRIGBAUM, W.R. and ROE, R.J., 1964: Crystal-lite orientation in materials having fiber texture. II. A study of strained samples of crosslinked polyethylene. *Jour. Chem. Phys.*, **41**, 737—748.
- MAEGILLAVRY, C.H. and PIECK, G.D. eds., 1962: *International tables for X-ray crystallography*. **3**, 362p., The Kynoch Press, Birmingham, England.
- NISHIKAWA, T., NISHIDA, T., ISHIKAWA, M., UEGAKI, T. and UEI, I., 1974: Hot gorging of cubic polycrystallite ferrite. *Yogyo-Kyokai-shi*, **82**, 25—30. (in Japanese with English abstract)
- PHILLIPS, F.C. and BRADSHAW, R., 1970: The use of X-rays in petrofabric studies. In PAULITSCH, P. ed.: *Experimental and natural rock deformation*. 75—97. Springer-Verlag Berlin.
- RIEKELS, L. M. and BAKER, D.W., 1977: The origin of the double maximum pattern of optic axes in quartzite mylonite. *Jour. Geol.* **85**, 1—14.
- ROE, R. J., 1966: Inversion of pole figures for materials having cubic symmetry. *Jour. Appl. Phys.*, **37**, 2069—2072.
- and KRIGBAUM, W. R., 1964: Description of crystallite orientation in polycrystalline materials having fiber texture. *Jour. Chem. Phys.*, **40**, 2608—2615.
- SANDER, B., 1930: *Gefuegekunde der Gesteine*. Springer, Berlin, 352p.
- SCHMIDT, W., 1925: Gefuegestatistik. *Tchermaks Mineral. Petrol. Mitt.*, **38**, 392—423.
- TSUKAHARA, H., 1974: Flow properties in rocks and silicates. In UEDA, S. ed.: *Flow in solids*. 109—132. Tokai Univ. Press, Tokyo. (in Japanese)
- , YAMAZAKI, T. and OHNO, I., 1976: Preferred lattice orientation produced by deformation in natural olivine aggregate and synthetic fayalite under stress. *Jour. Geol. Soc. Japan*, **82**, 713—727.
- and WILD, W. R., 1972: Orientation distribution diagrams for three Yule marble fabrics. In HEARD, H.C. et al., eds.: *Flow and fracture of rocks*. 83—94. Geophysical monograph 16, Amer. Geophys. Union, Washington, D.C.
- , VENKITASUBRAMANYAN, C. S. and BAKER, D. W., 1975: Preferred orientation in experimentally deformed dolomite. *Contrib. Mineral. Petrol.*, **50**, 115—126.

実験的に変形された岩石中の立方晶系結晶の選択配向の決定
特にスピネル型鉱物について

藤村 彰夫・大野 一郎・熊沢 峰夫

要 旨

変形した多結晶体中における立方晶系結晶の選択配向を、X線回折によって決定する方法に関する基本的な問題について研究した。すなわち、(1) X線極点図形 (ポールフィギュア)、ゴニオメーターを使用して、結晶方位のデータを得る方法、(2) インバース・ポール・フィギュアを得るための理論的・数値的手段、(3) それらの誤差の見積り方法である。

上部マントルのスピネル層中の主要成分である γ -(Mg,

Fe)₂SiO₄ の類似物質として、trevorite (NiFe₂O₄; スピネル構造を持つ) の多結晶体を、高温高圧下で一軸変形した。変形された trevorite の多結晶中に生じた選択配向を、ポール・フィギュア・インバージョンで解析した。予備的な結果では、trevorite 多結晶には、syntectonic な再結晶によって、少なくとも2つのタイプの選択配向 ($\langle 110 \rangle //$ 圧縮軸, および $\langle 111 \rangle //$ 圧縮軸) が生ずることが明らかになった。