Polycrystal Scattering Topography

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Abstract

The concept of X-ray diffraction topography, which has been ordinarily applied to single crystals, has been extended to the observation of polycrystals and amorphous materials. This new method utilizes X-rays scattered elastically or inelastically from a specimen to observe polycrystals, and is called polycrystal scattering topography (PST). The principle and some PST techniques are described. The discussion is further extended to the potentiality of PST and an experimental method with synchrotron radiation. In order to demonstrate the capability of this method, several techniques of PST have been applied to metallurgical problems. It has been found that the PST provides a helpful tool in studying polycrystals in which neither X-ray radiography nor X-ray diffraction topography would be helpful.

1. Introduction

For the imaging of materials (including biological ones) by X-rays, two ways are known: X-ray radiography and X-ray diffraction topography. The principle of the former (Ong, 1967; Yoneda, Horiuchi & Hiramatsu, 1980) is based on mapping of differences in the X-ray absorption of materials, and has therefore widely been used in some medical examinations and also for material testing. This traditional method, however, gives little physical information about those materials which exhibit small differences in X-ray absorption. Moreover, this method provides, in principle, merely two-dimensional projection images. The other imaging method, X-ray diffraction topography, has given us a powerful tool for studying crystals because it reveals, in a non-destructive way, individual lattice defects in otherwise 'good' single crystals. However, X-ray diffraction topography presents a serious problem regarding the quality of crystals: it needs as specimens fairly perfect single crystals (the dislocation density, for example, must be smaller than of the order of $10^3 \text{ cm}^{-2}$). This condition has severely restricted the extent of the application to a limited number of fields, such as diffraction physics, crystal growth or defect study.

Polycrystal scattering topography (PST) has been developed to extend the applicability of X-ray diffraction topography, and to obtain more physical information than from X-ray radiography. In PST we use the scattered X-rays including elastically diffracted ones to image polycrystals or amorphous materials according to their (crystal) structure or material distribution. PST is able to give, in principle, three-dimensional images to record the differences of crystal structure, of fluorescence (absorption edge), of Compton scattering, of texture, of small-angle scattering and of any kinds of differences of scattering phenomena by atoms or crystals inside the specimen.

Historically, Horiuchi & Yoneda (1971) first discussed the concept of polycrystal scattering topography and demonstrated the applicability of PST to a preliminary research of diffusion processes between copper and aluminium. Later, Yoneda & Chikaura (1979) reported various techniques for PST using a Soller slit or a Soller-slit assembly, and discussed the feasibility of PST. Recently, a method similar to the above Soller-slit method has been devised, independently of ours, by Born (1979). Born & Schwarzbauer (1980) have also presented a simple technique using a diaphragm. However, their techniques, in principle, give only two-dimensional images, and have dis-
advantages such as long exposure times and poor resolution.

The aim of the present paper is to describe our PST methods and to discuss the potentiality of PST. In order to demonstrate the capability of PST we have examined commercially used aluminium plates, aluminium coins and aluminium polycrystalline ingots. The former two observations reveal the existence of a texture distribution and the latter reveals rotations of the orientations of individual grains depending on the degree of mechanical processing. Finally, we discuss a PST technique using synchrotron radiation.

2. Principle and experimental methods

2.1. Principle

The PST utilizes X-rays which are elastically or inelastically scattered from a specimen, whereas X-ray radiography uses transmitted radiation, as schematically shown in Fig. 1. An essential condition which PST imaging should fulfill is the one-to-one correspondence between the specimen and the image. In order to fulfill this condition a Seller slit (or a Seller slit assembly) and/or a specimen-scanning mechanism are used.

2.2. Cross-Seller-slit method

This method was used in the first PST equipment with which polycrystal copper–aluminium alloys were studied with Debye-Scherrer diffracted X-rays (Horiuchi & Yoneda, 1971). Fig. 2 shows a schematic principle of this technique in the transmission case. We call it the cross-Seller-slit method. A characteristic of this geometry is the use of two Seller slits rotated by 90° with respect to each other and the oscillating mechanism along a direction between the normals of the two Seller-slit planes as indicated in Fig. 2. The oscillation is performed to eliminate the shadows of the slit materials. The two combined Seller slits act as a collimator of scattered X-rays from the specimen.

In order to observe a larger area of the specimen, an oblique setting of the specimen to the incident beam is made, as shown in Fig. 2, or diffracted X-rays from an asymmetrically cut monochromator are used as incident beam.

The spatial resolution depends on the parallelism of the Seller slits. If we tolerate an exposure time of up to several days with a normal sealed-off X-ray tube and sensitive industrial X-ray film, we attain a resolution of the order of 0.1 mm by using Seller slits with a parallelism of the order of 10^{-3}.

One of the merits of this system is simplicity of the equipment; we need no mechanism for scanning the specimen. Therefore, this system is suitable for the PST study of polycrystals under the influence of various external fields such as cooling, heating, magnetic field or mechanical stress. However, this cross-Seller-slit method gives merely two-dimensional images. Another disadvantage lies in less flexibility in specimen setting.

2.3. Seller-slit oscillating method

The schematic principle of a three-dimensional PST arrangement is shown in Fig. 3. Slits S1 and S2 and a Seller-slit assembly are initially aligned in such a way that X-rays scattered from the centre of the specimen in a wanted direction (i.e. at a given angle to the incident beam) are selected. To avoid shadows from the Seller-slit materials, the Seller-slit assembly should be oscillated vertically, the oscillation period being of the order of a few minutes. The slit combination together with the specimen and the photographic plate are mounted on a horizontal scanning mechanism such as is applied in a Lang camera (Lang, 1959, 1970). When using Debye–Scherrer diffraction of low-order reflections and a normal X-ray tube operated at 40 kV/25 mA, and allowing as a tolerable exposure time several tens of hours, the slits can be adjusted in such a way that the spatial resolution is practically of the same order as that in the cross-Seller-slit method. The present Seller-slit assembly consists of three Seller slits which are placed at particular intervals (Oki & Futagami, 1969). The assembly limits the vertical divergence of scattered X-rays to 4.0 × 10^{-3} rad. Needless to say we could obtain better resolution if a Seller slit with higher parallelism and slits S1 and S2 with narrow width were employed; but then a higher-output X-ray source like a rotating anode would be required. Merits of this system are the capability of three-dimensional observation and the
flexibility in specimen setting. This method is particularly suitable for a study of texture distribution because some textured materials reflect X-rays in a limited azimuthal angular range on a Debye–Scherrer ring. This method was applied for observing the internal structure of commercially used aluminium plates and aluminium coins (see § 3.1).

2.4. X-Y scanning method

A more sophisticated method is shown in Fig. 4 for the transmission case (it can also be modified for the reflection case). The method adopts electronic imaging procedures and two-dimensional scanning mechanism of the specimen synchronized with the electron scanning in a cathode ray tube (CRT). The X-rays enter the specimen as a pencil beam with white spectrum. X-rays scattered from one point are introduced through a cone-shaped slit into a solid-state detector, as shown in Fig. 4. A deformed cone-shaped slit can also be used when observing anisotropic materials such as heavily deformed metals, wood or other biological materials. The cone slit is sometimes partly closed to make the image contrasts of textured materials clearer. A solid-state detector and a multi-channel analyser resolve the specimen of the scattered X-rays coming through the cone slit to visualize the PST image in a multicolour cathode ray tube (Yoneda, Horiuchi & Hiramatsu, 1980). A detector D in Fig. 4 detects, if necessary, the influence of specimen thickness and is used to maintain a nearly constant background over the whole picture. In the present study we use this system only in single-colour mode without the detector D. We have studied polycrystals such as polycrystalline metals, wood and various other biological materials. Some observations will be described in § 3.

The exposure time for this system is typically 10 to 30 min under X-ray conditions 40 kV/15 mA. Although the spatial resolution depends on the preciseness of the cone slit, we have easily obtained better than 0.1 mm.

2.5. Proposed equipment for PST with synchrotron radiation

Let us discuss a case of observation of metal or alloy polycrystals with Debye–Scherrer diffracted X-rays. Fig. 5 shows schematically the principle of PST equipment with synchrotron radiation. X-rays diffracted along a particular azimuthal angular range from a specimen enter a spatially resolving arrangement of two asymmetrically cut monochromators, M1 and M2, of perfect silicon or germanium crystals, two-dimensional position-sensitive proportional counter (PSPC) and CRT. The rotation axis of the monochromator M1 is placed parallel to the polarization direction of the synchrotron radiation. The reflection by the second crystal M2 is perpendicular to that of the first crystal M1. The two monochromators ensure the one-to-one geometrical correspondence between the specimen and the image in the CRT. They also act as an image magnifier; M1 and M2 enlarge the irradiated area of the specimen two-dimensionally.

The method of beam enlargement was first demonstrated in X-ray diffraction topography by Kohra (1962). Boettinger, Burdette & Kuriyama (1979) have also applied the technique to X-ray radiography. Spatial resolution obtained on the CRT is determined mainly by the resolution of PSPC and the
asymmetry factors of \( M_1 \) and \( M_2 \). The contribution of the angular divergence of incoming and outgoing beams of \( M_1 \) and \( M_2 \) to the resolution is negligible in comparison with the above factors, except in the case when a fine-grain emulsion-like nuclear plate is used instead of a PSPC and CRT.

If we use a PSPC with a resolution of 1 mm (Kaplan, Kaufman, Perez-Mendez & Valentine, 1973) and monochromatize with an asymmetry factor \( b \) of 0-1, we obtain 0-1 mm as spatial resolution \( \delta \) on the CRT (the factor \( b \) is defined in Fig. 6). The above resolution is obtainable under the following experimental parameters for a concrete example:

- wavelength \( \lambda = 0.71 \) Å,
- \( M_1 \) and \( M_2 \): silicon 220 reflection,
- \( \theta_0 = 88^\circ \)
- \( \alpha = 8^\circ 30' \).

When fine-grain emulsion plates are employed for image recording instead of the PSPC and CRT system, the spatial resolution \( \delta \) can be much improved, though the required time for recording one frame of the image may be longer. The value of \( \delta \) is determined mainly by the angular divergence \( \Delta \theta_0 \) of the incoming beams from \( M_1 \) and \( M_2 \), because of the smaller angular divergence \( \Delta \theta_h \) of the outgoing beams. Provided that the total distance \( l \) between the specimen and the emulsion plate is 300 mm under the experimental condition (A), we obtain

\[
\delta < l \Delta \theta_0 \approx 10 \mu m,
\]

where \( \Delta \theta_0 = \Delta \theta_h / \sqrt{|b|} = 1.1 \times 10^{-5} / \sqrt{0.1} = 3.4 \times 10^{-5} \) (\( \Delta \theta_h \) is the intrinsic angular width of Bragg reflection in the symmetric case).

Let us consider the contribution of higher harmonics of the synchrotron radiation. Since white synchrotron (SR) beams (Tanner, 1977) are used as the incident beam for this PST observation system, higher harmonics inevitably contribute to the imaging. However, two independent monochromators \( M_1 \) and \( M_2 \) attenuate the higher harmonics. Here we treat a case of 111 Debye-Scherrer diffraction of polycrystalline aluminium, for example, under the conditions (A). It may be reasonable to presume that the reflections at the specimen and at the perfect-crystal monochromators are kinematical and dynamical, respectively. Therefore, the ratio of the intensities \( I_{n\alpha} \) of \( n \)-th-order harmonics is given as

\[
I_{111} : I_{222} : I_{333} : I_{444} : ... = |F_{111}^{Al}|^2 |F_{220}^{Si}|^2 : |F_{222}^{Al}|^2 |F_{440}^{Si}|^2 : |F_{333}^{Al}|^2 |F_{660}^{Si}|^2 : |F_{444}^{Al}|^2 |F_{880}^{Si}|^2 : ...
= 100 : 24 : 4 : 1 : ... \tag{1}
\]

\((F_{hkl}^{Al} \) and \( F_{hkl}^{Si} \) are the structure factors of aluminium and silicon, respectively), where the spectral energy distribution of the synchrotron radiation and the absorption in the specimen for the relevant wavelength are neglected. When we take a condition of optimum thickness (nearly equal to the inverse value of the linear absorption for each wavelength) into consideration, the contribution of higher harmonics may become much smaller than that given in (1).

In the case of an emulsion recording system, the film emulsion again weakens the effect of higher harmonics. Roughly estimated values of a blackening factor for a nuclear emulsion (Kodak R) have been given (Stephenson, Tuomi & Kelhä, 1980) as 0.6, 0.4, 0.02, ~0, ... for wavelengths of 0.71 Å \((n=1)\), 0.36 Å \((n=2)\), 0.24 Å \((n=3)\), 0.18 Å \((n=4)\), ..., respectively. Therefore, the relative contribution of the higher harmonics becomes (under unfavourable conditions)
Accordingly, the higher harmonics contribute negligibly to the PST images. In the case of employing the PSPC and CRT system, the final contribution depends on the characteristics of the pulse-height analyser and PSPC within a maximum value given in (1).

Here we note again that the monochromators $M_1$ and $M_2$ have three functions; the first is to ensure the one-to-one correspondence between the specimen and the PST image, the second to magnify the specimen images and the third to attenuate higher harmonics. Although we have discussed here an experimental setup and its condition mainly for a metallurgical application, the basic principle may be applicable to other research fields with different kinds of scattered X-rays.

3. Experiments with polycrystals

3.1. Observation of texture distribution in commercially used aluminium plates and in aluminium coins

Fig. 7(a) represents the 111 reflection Debye-Scherrer ring of a commercially used aluminium plate with a thickness of 1 mm. The inhomogeneous intensity distribution implies the existence of texture, which is explicitly represented in a set of pole figures (Taylor, 1961, for example). The plate was impressed with three metal types of 'P', 'S' and 'T' to change the local texture. Figs. 7(b), (c), (d), (e) and (f) represent PST images observed with X-rays diffracted along different directions by means of the Soller-slit oscillating method described in § 2.3. The experimental conditions were Mo Kα, 40 kV/25 mA, 80 h exposure. The topographs and Debye-Scherrer photograph are positive prints, i.e. white regions are higher radiation areas. We indicate the observing direction with an azimuthal angle $\phi$ measured clockwise from a reference line in Fig. 7(a). Since in this Soller-slit oscillating method reflection vectors maintain their physical sense, they are given in the relevant topographs.

All the topographs reveal light and dark patterns parallel to the rolling direction. The irregular pattern indicates that the mechanical plastic deformation is not homogeneous over the plate. In other words, the texture of the aluminium plate caused by the rolling varies from point to point. Accordingly, we find that the texture of the commercially used aluminium plate cannot be represented with a set of pole figures. The spatial period of the texture irregularity is found to be approximately 1 mm. Such structure inhomogeneities sometimes influence seriously the mechanical and chemical properties, which are closely linked with a distribution of microscopic stress. The investigation on a relationship between such structure irregularities and physical properties is in progress.

The behaviour of the image contrast caused by the impressions $P$, $S$ and $T$ is rather odd. In the topographs of Figs. 7(b) and (d) taken at azimuthal angle $\phi = 0^\circ$ and $180^\circ$, respectively, the traces of the types $P$, $S$ and $T$ are almost invisible, although the letter $T$ may be deciphered in Fig. 7(d). On the contrary, topographs of azimuthal angle $\phi = 90^\circ$, 255 and 287° give clear contrast at the traces of $P$, $S$ and $T$, as shown in Figs. 7(c), (e) and (f). This contrast extinction suggests that impressing with typing tools changes the texture parallel to the rolling direction. We note that the contrast of the letter images is different in Figs. 7(c), (e) and (f). The contrast behaviour including the above image extinction in Figs. 7(b) and (d) gives information on features of the local deformations and an interaction between the mechanical processing and the existing structure. [Sharp fringe-like patterns parallel to the reflection vectors in Figs. 7(d) and (f) are due to shadows of Soller blades.]

We also show polycrystal topographs of an alum-
The aluminium coin in Figs. 8(b) and (c), and for comparison the optical photograph in Fig. 8(a). The topographs are taken with the Soller-slit oscillating method with Mo Kα radiation, 40 kV/20 mA, 111 reflection. The exposure time was 72 h. The relevant reflection vectors are indicated by an arrow. These topographs also reveal clearly the existence of texture distribution with a wider period than that in Fig. 7 and a special contrast behaviour of the image. An impressed trace in the central part of the coin becomes invisible in a topograph with a certain reflection vector, as shown in Fig. 8(c). The contrast behaviour is qualitatively similar to that observed in Figs. 7(b) to (f).

3.2. Observation of the developing stages of rolled texture in an aluminium ingot

Figs. 9(a) to (d) represent topographs which indicate the structural change in an aluminium polycrystalline ingot after some degree of rolling. The topographs were taken by the transmission X–Y scanning method (see § 2.4) under the following conditions; Mo Kα, 40 kV/17 mA, beam diameter 0.5 mm, 111 Debye–Scherrer diffracted beams. All the topographs are positive prints. The initial state shown in Fig. 8(a) is an as-grown polycrystal slowly cooled from its melt in a furnace. After every rolling process a PST observation was made. From these topographs it is clearly revealed that orientations of individual grains rotate gradually in the rolling direction, as a whole, as the specimen is rolled. The rolling texture has been clearly formed in the final state (d). The quantitative analysis of such a series of observations should be performed with the help of mechanical metallurgy. This kind of PST observation may enable us to interpret the origin and mechanism of rolling texture or recrystallization.

4. Conclusion

The concept and method of a new kind of topography have been established, where X-ray images are constructed by recording scattered X-rays — polycrystal scattering topography (PST).

Three experimental methods for PST have been described and compared: (1) the cross-Soller-slit method, (2) the Soller-slit oscillating method, (3) the X–Y scanning method. The cross-Soller-slit method may be helpful for studying polycrystals under the influence of various kinds of external fields. This method can be employed in synchrotron radiation experiments. The Soller-slit oscillating method is found to be suitable particularly for studying the texture of polycrystals or the influence of the texture on some physical properties. The X–Y scanning method is apparently the most sophisticated technique in the sense of potentiality and capability. An electronic imaging system decreases the required exposure time down to 10 min (with a normal sealed-off X-ray tube), compared with 100 h in the cross-Soller-slit or the Soller-slit oscillating methods. This method is a powerful tool for studying biological materials.

At present there is certainly room for improving the image quality. The spatial resolution, for example, is more or less in all the above techniques counter-
balanced with the required exposure time. Therefore, higher-output X-ray sources such as a rotating-anode generator or a synchrotron storage ring may be helpful to obtain clearer PST images. The PST technique which we discussed in § 2.5 may then give smaller values than 10 µm for the resolution. This value is of the same order as that of the ordinary X-ray diffraction topography or X-ray radiography. An alternative is to employ an electronic imaging system, as described in § 2.4.

As a demonstration we have made a preliminary PST experiment in the field of metallurgy. The observation has proved that a texture distribution of commercially used aluminium plates exists and that a mechanical process like impressing changes the existing texture in a particular direction. Also, developing stages of rolling texture of an aluminium ingot have explicitly been observed as a function of the degree of rolling. We have confirmed by these experiments that PST is indeed a helpful tool for studying polycrystals in a way which neither X-ray radiography nor X-ray diffraction topography would provide.

Although, in the present paper, we have been concerned mainly with some metallurgical problems for the application, the basic principle may maintain its availability also for other research fields. In fact, we have recently succeeded in observing internal structures of eggs, wood and other biological materials by the X–Y scanning PST method. These will be published elsewhere. We note here that it will be possible to observe polycrystals or biological materials on a multicolour CRT.

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