Defect Structure Analysis of Polycrystalline Materials by Computer-Controlled Double-Crystal Diffractometer with Position-Sensitive Detector

BY R. YAZICI, W. MAYO, T. TAKEMOTO AND S. WEISSMANN*

Department of Mechanics and Materials Science, Rutgers University, Piscataway, NJ 08854, USA

(Received 29 March 1982; accepted 17 August 1982)

Abstract

The method represents an extension of a previously developed X-ray double-crystal diffractometer method when a film was used to record the crystallite reflections, each reflecting crystallite being regarded as the second crystal of a double-crystal diffractometer. By utilizing a position-sensitive detector (PSD) with interactive computer controls, the tedious and limiting task of data acquisition and analysis is greatly simplified. The specimen is irradiated with crystal-monochromated radiation and the numerous microscopic spots emanating from the reflecting crystallites are recorded separately by the position-sensitive detector and its associated multichannel analyzer at each increment of specimen rotation. An on-line minicomputer simultaneously collects these data and applies the necessary corrections. This process is then automatically repeated through the full rocking-curve range. The computer carries out the rocking-curve analysis of the individual crystallite reflections as well as that of the entire reflecting crystallite population. The instrument is provided with a specimen translation device which permits analysis of large sections of solid specimens. Thus, sites of local lattice defects induced either mechanically, chemically or by radiation can rapidly be established and quantitatively determined in terms of rocking-curve parameters as well as imaged by X-ray topography, by inserting a film in front of the PSD. The versatility and usefulness of the method is demonstrated by examples given from studies of fracture, fatigue and stress-corrosion cracking of commercial alloys.

Introduction

From a historic viewpoint the double-crystal diffractometer (spectrometer) was first used to answer theoretical questions associated with problems of X-ray intensities. Thus, it was used to measure the absolute integrated reflections (Compton, 1917; Bragg, James & Bosanquet, 1921; Wagner & Kulenkampff, 1922), the widths of the reflection curves (Davis & Stempel, 1921) and to test whether the intensities were proportional to $|F|$ or $|F|^2$ (Ehrenberg, Ewald & Mark, 1928). Many investigators pursued such studies (Allison, 1932; Parratt, 1932; Renninger, 1934) and it soon became apparent that the double-crystal diffractometer method (DCD), when employed in the parallel arrangement to eliminate the dispersion effect, could be used as a powerful research tool to elucidate lattice defects in single crystals (Renninger, 1934). Although the method soon found widespread application in imperfection studies of single crystals, it was not until 1951 that DCD was first employed to investigate the lattice defects in polycrystalline materials (Reis, Slade & Weissmann, 1951). Subsequently, DCD has been successfully used over the past few decades to study minute changes in the defect structure of polycrystalline metallic samples (Slade & Weissmann, 1952; Intrater & Weissmann, 1954; Weissmann, 1961; Garofalo, Zwell, Keh & Weissmann, 1963; Hida & Weissmann, 1975; Weissmann, Yazici, Takemoto, Tsakalakos & Kramer, 1979; Takemoto, Weissmann & Kramer, 1980; Pangborn, Weissmann & Kramer, 1981). The method is particularly useful when the amount of deformation is not too excessive and when the substructure of the material can be related to its properties. The study of the recrystallization behavior of cold-rolled low-carbon alloy steel (Slade & Weissmann, 1952) and aluminum (Weissmann, 1961; Weissmann, Imura & Hosokawa, 1963), the substructure formation in iron during creep (Garofalo, Zwell, Keh & Weissmann, 1961), and particle-dislocation interaction in age-hardened Ti–Al–Mo alloys (Hida & Weissmann, 1975) are examples of the diverse applications of the double-crystal method. More recently, the method has been utilized as a non-destructive tool to predict the onset of fatigue failure in an aluminum alloy in both noncorrosive (Pangborn, Weissmann & Kramer, 1981) and corrosive (Takemoto, Weissmann & Kramer, 1980) environments, and to study stress-corrosion cracking in austenitic stainless steel.
In spite of the success of the double-crystal technique in studying a number of problems of technological importance, the method has not enjoyed widespread usage. The major drawback of the method is the very tedious task of data collection and analysis. A typical experiment may require 2–3 d for exposure, film development and microdensitometer analysis of the film. Moreover, to provide reliable statistics for a meaningful analysis of structure/property relationships, many such experiments must be run. Even with efficient high-power X-ray sources, the data collection is very time consuming.

The purpose of this paper is to present an updated computerized version of the double-crystal diffractometer which greatly reduces the data acquisition and analysis time. By utilizing a position-sensitive detector to replace the X-ray film in the previous technique, and employing an on-line computer for control and subsequent analysis, the time required for collecting data is often reduced by a factor of 50. An additional benefit of the new method is the ability to discriminate rapidly between grains which are strongly deformed from those which are not. This feature appears to be of great interest since strongly deformed sites may turn out to be precursors of microcracks and, therefore, warrant more detailed analysis. The subsequent X-ray topographic imaging of these disclosed sites may provide then the experimental link for detailed TEM and SEM studies of the dislocation structure.

System description

1. Operating principle

A sample is held stationary while being irradiated by a crystal-monochromated parallel X-ray beam. Several grains in the specimen will be in reflecting positions which result in individual microscopic spots along the appropriate Debye–Scherrer arc. These spots are detected by a position-sensitive detector oriented parallel to the Debye–Scherrer arc as shown in Fig. 1. The intensity of each spot and its location are then stored in a digital memory device for numerical analysis. Subsequently, a small discrete change in the angular setting of the sample is made and the X-ray measurements are repeated. In Fig. 1, the change in angular specimen setting was carried out in discrete intervals of 6 min. By changing the angular setting of the sample, the individual crystallites are stepwise rotated through the range of reflecting positions. The reflected intensities, recorded as a function of crystallite rotation, represent rocking curves. In the detail of Fig. 1, the reflection of grain D, as recorded by the PSD and shown on the display screen, exhibits a complete rocking curve. At the position of angular setting 0 the intensity signal is small. It increases with specimen rotation of 6 min of arc ('), passes through a maximum at 12' rotation and is about to disappear after 18' rotation. For grains A, B, C the reflecting range is much smaller. For these grains the ascent and descent of the reflected intensities in the rocking curve become much more apparent when the specimen is rotated through smaller, discrete angular intervals (not shown in Fig. 1). The width, β, at half the maximum intensity of the rocking curve provides a measure of the angular lattice misalignment introduced during the deformation of the crystallite. Thus, the β values provide information about the local densities of the accumulated excess dislocations of one sign.

Some relationships between the distribution of excess dislocations and the resultant rocking-curve profiles are schematically shown in Fig. 2. If the profile...
is smooth, a homogeneous distribution of excess dislocations would exist (grain A). If, on the other hand, a multipeaked profile is produced, the excess dislocations are heterogeneously distributed, giving rise to distinct lattice tilts as would be encountered, for example, in grains with subgrain boundaries (grain B). The angle between the subpeaks of such rocking curves would then represent the relative misorientation of adjacent lattice domains or subgrains, whereas the width of well resolved subpeaks indicate the degree of internal distortion in these domains. Annealed grains with low excess dislocation densities (viz grain C) give rise to smooth rocking curves with small $\beta$ values.

Assuming that the excess dislocations are randomly located in the grains and thus can be represented by a Gaussian distribution, the excess dislocation density $D = \beta^2/\pi b^2$, where $b$ is the magnitude of the Burgers vector. If the excess dislocations are aligned in the subgrain boundary and the tilt angle of adjacent subgrains, $\epsilon$, is measured, the excess dislocation density in the subgrain boundary $D_{SB} = \epsilon/3bt$, where $t$ is the subgrain size (Hirsch, 1956). If resolvable, the subgrain size can be measured from the images of X-ray reflection topographs (Berg–Barrett topography).

2. Hardware

A schematic diagram of the entire CARCA (Computer Aided Rocking Curve Analyzer) system is shown in Fig. 3. The X-ray beam, emerging from the X-ray window, is monochromatized by diffraction from the (111) planes of a Si or Ge crystal. The resultant beam has a very small horizontal convergence and is very nearly parallel, while the vertical divergence of the incident beam is unaffected. This reflected beam passes through a slit system before entering the X-ray camera as shown in the detailed diagram of the camera in Fig. 4. The sample under investigation is mounted on a two-dimensional microscope stage (detail 4) which enables precise selection of the region of interest. The diffracted beam from the sample is detected by the one-dimensional position-sensitive detector (detail 6) which is located parallel to the Debye arc. The X-ray intensity spectra are collected and displayed by the multichannel analyzer (MCA) before being transmitted to the on-line minicomputer. When this serial data transmission is completed, the computer issues a command pulse to the sample position device to rotate the sample incrementally before the next data-aquisition sequence is initiated. When the defect structure of a large population of crystallites had to be surveyed by the previous film method, for example surveying the grain population of the gage length of a metal specimen, equatorial as well as nonequatorial reflections had to be analyzed. Owing to the effect of the vertical divergence of the incident beam, time-consuming corrections had to be made for the broadening of the rocking curves pertaining to nonequatorial reflections (Slade & Weissmann, 1952). Applying CARCA, a large grain population can readily be analyzed using only equatorial reflections and the entire gage length can quickly be analyzed by moving the specimen on the microstage relative to the incident monochromated beam. This aspect of the use of CARCA is one of the most attractive and time-saving innovations.

3. Software

The software package consists of three Fortran programs and a data file stored on magnetic disk. These programs are used for data acquisition, data analysis and data transfer. The acquisition program controls the operation of the PSD/MCA system and the sample positioning device. It is also responsible for the data collection and creation of the data file. The management program is used for transferring the data file from the operating disk onto the permanent storage disk.

Fig. 3. Schematic diagram of the CARCA X-ray system.

Fig. 4. Close-up photograph of double-crystal X-ray camera. (1) X-ray tower; (2) slit systems; (3) monochromator; (4) sample and X–Y positioner; (5) film holder; (6) position-sensitive detector; (7) stepping motor; (8) film shifter; (9) rotational knob and counter.
A third program opens the data file created by the acquisition program and performs the rocking-curve analysis. The input data are reconstructed to simulate a two-dimensional picture of the grain reflections. A curve-smoothing routine is utilized to minimize counting errors before the background intensity is determined and subtracted. The curve smoothing consists of averaging the intensity readings of three adjacent MCA channels. No attempt is made to curve smooth a single channel through the sample rotation increments. The present method of background determination consists of searching each MCA channel for background present in highly deformed samples with small particle size. This value is then subtracted from all intensity readings on that channel before the procedure is repeated for the next channel.

The bulk of the analysis program consists of a process of pattern recognition by scanning the two-dimensional field to locate local peaks and assigning these to the appropriate grain reflections. The array consisting of these local peaks is grouped into individual grain reflections and then scanned to determine if multiple peaks are present. If found, the multi-peaked array is separated into the individual peaks before the rocking-curve halfwidths are determined.

The halfwidths are calculated by examining each group of reflections to find the width of the distribution at one-half of the peak intensity. Both sides of the peak are examined and, if necessary, a linear interpolation between data points is made. In the case of multiple peaks, however, only one side of the curve is searched and a temporary halfwidth for that portion is determined. The intensity distribution is then assumed to be symmetric and the temporary halfwidth is doubled to obtain the reported value.

An option is available to suppress the reporting of any reflecting grains which have very low intensities or very narrow halfwidths. This is a very powerful feature of the analysis, since it permits the isolation of those highly deformed grains from among the potentially large population of undeformed grains.

A greatly simplified computer printout of sample test results is shown in Fig. 5. The filtered intensity map is shown after curve smoothing, removal of the background and identification of the local maxima. Also shown are the numerical results of the rocking-curve analysis for each grain which exceeds the threshold. If a multiple peak is found, the \( \beta \) value is followed by \( (M) \) for identification.

4. Error sources of CARCA

The principal source of error in the CARCA method arises from the determination of the background level. Significant errors in measurements can result from overestimating the background and these errors are most severe for low signal-to-noise ratios \( (S/N) \). There are two primary sources of error in determining the background. When the background is low, the curve-smoothing process will introduce large errors of the order of 20–30%. It is fortunate in this case that the \( S/N \) ratio is always large. A more serious problem arises when the background level is high. Then the tail of a peak may be misinterpreted for the background and errors in the 10–30% range are possible. Based on calculations and experiment, it was found that a setting of the intensity threshold which was at least three times the background gave satisfactory results. In practice, it is found that a large number of low-intensity reflections have very small \( \beta \) values. Unless these data are eliminated based on the above criterion, the overall grain distribution is artificially distorted.

If sufficient computer memory is available, the problems cited above can be avoided or minimized by a more sophisticated numerical analysis. The method used employs a two-dimensional fast Fourier transform (FFT) to analyze and separate the background noise. This method markedly improves the \( S/N \) ratio, but at a premium of computational time. Such techniques are presently being implemented in a second-generation CARCA system in this laboratory and will be reported separately.
Applications

1. Quantitative mapping of deformation gradients

The problem of stress raisers and their interactions have always been of great concern in technologically applied polycrystalline materials. A nondestructive analysis of long-range plastic strains and associated deformation gradients of a tensile deformed notched specimen has been carried out to illustrate the usefulness of this technique (Mayo, Yazici, Takemoto & Weissmann, 1981). A solution-annealed 304 stainless steel (18Cr, 8Ni) notched tensile specimen with the geometry shown in Fig. 6 was stressed near the yield point. The area surrounding the notch was scanned by the CARCA system to determine the deformation gradient as measured by the rocking-curve halfwidth. For such an application of CARCA where a high degree of spatial resolution was required, the sample was mounted on a microscope stage. A sample movement of 100 μm could be achieved in two dimensions, and the contour map of the deformed region was obtained with fine detail. The shape of the contour map and the corresponding micrograph are shown in Fig. 6. The region immediately adjacent to the notch tip (region 2 of Fig. 6) was shown by the present computerized method to have a relatively low excess dislocation density (β = 15–17⁰). The reason for this effect is revealed by the photomicrograph. It was found that the region near the notch tip exhibited a high density of slip traces indicating a high dislocation mobility. No such slip traces were found more than 3–4 grain diameters from the immediate vicinity of the notch tip. It is rationalized that in grains close to the notch dislocations were free to move to the surface, while in grains further from the notch interactions between the activated slip systems caused a local accumulation of excess dislocations with concomitant workhardening. This gives rise to regions 3 and 4 in the contour map with β values as high as 21⁰. Conventional micrographic analysis and scanning X-ray diffractometry analysis show only the rough contour of region 4 whereas the CARCA method reveals the full detail of the strained region.

2. Statistical determination of overall structural integrity

A reliable and practical means of predicting failure due to fatigue has recently been demonstrated (Pangborn, Weissmann & Kramer, 1981). In the present experiments a commercial aluminum alloy, Al 2024-T4, was fatigued (R = -1) in air at 80% of yield and analyzed at various fractions of the number of cycles to failure. As shown in Fig. 7, the CARCA results indicate that most surface grains underwent plastic deformation although the distribution became increasingly asymmetric as failure was approached. The mean β value increased almost linearly with the number of cycles, offering a nondestructive means of measuring the remaining useful life. Such aggregate information of the defect structure of a large grain population is of obvious technological importance, especially in view of the fact that this information can be rapidly obtained by application of the CARCA system.

3. Isolation of regions of intense deformation

One of the greatest challenges to today's technology is the nondestructive failure prediction in stress-corrosion cracking (SCC). This task is made especially difficult if failure occurs at low stress levels. Under
such conditions, relatively few grains in a large grain population undergo appreciable deformations. In the present experiment, solution-annealed 304 stainless steel (18Cr, 8Ni) was stressed at 55% of the yield strength and exposed to a boiling MgCl₂ solution. The samples were analyzed at various deformation times. The selectivity of grain deformation in SCC is well demonstrated in Fig. 8 which shows rocking-curve measurements of this sample carried out photographically. The crystal-monochromated incident beam was reflected by a few grains and the reflected spots were recorded along the (111) and (200) Debye–Scherrer arc. Some of these spot reflections are denoted by the numbers 1, 2, 3, 4, 5. To obtain rocking curves from the grains, which gave rise to these spot reflections, the specimen was rotated in discrete intervals of 3° and between each angular setting the film was shifted so that the successive reflections could be spatially separated. This procedure gave rise to the array of spots shown in Fig. 8. Because of the dependence of spot intensity on angular rotation, the arrays of spots represent rocking curves. It will be seen that in the stress-corroded condition (Fig. 8b) only the grain reflections denoted by 1, 3 and 5 exhibited an increase in the rocking curve while the rest of the grain reflections, such as 2 and 4, remained virtually unaltered. The identification of the grains responsible for the specific spot reflections was established by X-ray topographic imaging of the grain reflections on a photographic film taken at the surface (Berg–Barrett X-ray topography) and spatial tracing of the reflections from the specimen surface to the Debye–Scherrer arc (Weissmann et al., 1979).

If one uses conventional scanning diffractometry in SCC, the minimal contribution of the deformed grains to line broadening remains undetected. By contrast, using CARCA, the selectivity of the deformation process in SCC can be successfully disclosed by eliminating all the rocking curves of those reflecting grains which are below the mean of the distribution of the annealed sample, denoted by the arrow in Fig. 9. Thus, when the attention is focused only on those grains exhibiting increases in β values the broadening effect in SCC is substantial and makes failure prediction possible.

It is evident that the photographic technique of detecting and isolating sites of intense deformation (Fig. 8) is tedious and very time consuming. CARCA, on the other hand, can perform this operation with great speed and efficiency and the examples cited here may serve to demonstrate the special versatility and usefulness of the method in deformation studies of polycrystalline materials. In technologically important deformation processes, such as fracture, fatigue, creep, stress-corrosion cracking, the grains undergo considerable deformation. The intense interactions of dislocations give rise to local accumulation of dislocations resulting in slip and persistent slip bands in deformation bands and dislocation substructure. Such sites are important centers from which long-range strains emanate and they are invariably associated with lattice curvature. It is this local lattice rotation and induced curvature in grains which CARCA can measure quantitatively. Hence the method can be instrumental in elucidating the various stages of the deformation processes and even predict catastrophic failure (Pangborn et al., 1981; Mayo, 1982; Takemoto, 1982; Yazici, 1982).
The support of this work by the Division of Materials Research, Ceramics Program, Metallurgy, Polymer and Ceramics Section of the National Science Foundation under Grant No. DMR 81-04985 is deeply appreciated.

References
