4-D XRD FOR STRAIN IN MANY GRAINS USING TRIANGULATION

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ABSTRACT

Determination of the strains in a polycrystalline material using four-dimensional X-ray diffraction (4DXRD) reveals sub-grain and grain-to-grain behavior as a function of stress. Here 4DXRD involves an experimental procedure using polychromatic X-ray micro-beam diffraction (micro-Laue) to characterize polycrystalline materials in spatial location as well as with increasing stress. The *in-situ* tensile loading experiment measured strain in a model aluminum-sapphire metal matrix composite using Beamline 7.3.3 at the Advanced Light Source. Micro-Laue simultaneously resolves individual grains in the polycrystalline matrix. Results obtained from a list of grains sorted by crystallographic orientation depict the strain states within and among individual grains. Locating the grain positions in the plane perpendicular to the incident beam is trivial. However, determining the exact location of grains within a 3-D space is challenging. Determining the depth of the grains within the matrix (along the beam direction) involved a triangulation method tracing individual rays that produce spots on the CCD back to the point of origin. Triangulation was experimentally implemented by simulating a 3-D detector capturing multiple diffraction images while increasing the camera to sample distance. Hence by observing the intersection of rays from multiple spots belonging to the corresponding grain, depth is calculated. Depth resolution is a function of the number of images collected, grain-to-beam size ratio, crystal structure, and the pixel resolution of the CCD. The 4DXRD method provides grain morphologies, strain behavior of each grain, and interactions of the matrix grains with each other and the centrally located single crystal fiber.

INTRODUCTION

Microstructure controls mechanical properties of materials. X-ray diffraction has long been a probe into the nano-, micro-, and meso-scopic properties of materials. The three-dimensional (3-D) structure of grains in a polycrystalline material is an example of microstructure. Usually, individual grain behavior is averaged and properties considered on a macroscopic scale. However, the technique described here is one of a few capable of directly probing the microstructure. With a sub-grain resolution, the technique probes non-destructively in three-dimensions continuously over appreciable length scales including thousands of grains. The non-destructive nature of the method allows further manipulation of the material so that other independent variables such as temperature and stress can be probed as a function of time—the fourth dimension in "four-dimensional X-ray diffraction" (4DXRD).

Nearly all structural metals and ceramics are polycrystalline. Polycrystalline materials consist of a large number of small crystals, often randomly oriented, and may be of the same composition or of different composition or of different structures. Sizes of grains in such polycrystalline materials range from microscopic to several millimeters. The structure, size, orientation, and

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composition of the constituent grains affect the deformation characteristics, embrittlement, corrosion, and electronic properties of the bulk material. The deformation characteristics of most materials depend greatly on this sub-micron interaction of the grains. X-ray micro-diffraction using polychromatic X-ray radiation also known as micro-Laue is a new and powerful tool in probing non-destructively individual grains in polycrystalline materials and thus providing information such as strain, position, and shape of the grains. This information can be used to study individual grain behavior, grain-to-grain interactions and combined effects under conditions of applied load leading to the macroscopic mechanical properties. X-ray diffraction techniques that were employed previously [1, 2] in such studies utilized monochromatic X-ray - radiation. Even though these techniques have a high level of accuracy in strain measurements, their applicability is limited to a few grains due to the dependence of the technique on specific orientation of the material with the incident beam, or require sample rotation in order to obtain a set of usable reflections.

3-D diffraction measurements using monochromatic radiation coupled with sample rotation have been implemented for determination of stress states in grains and at grain boundaries on the 3-D XRD microscope at ESRF [2, 3] by tracking diffraction of focused X-rays. These experiments map spot shapes and back trace them into the sample to achieve the shape of the grains. Such techniques are successful in achieving reflection from mm deep grains due to the high energies. However, the method of rotation involves various limitations to the experimentation such as the addition of a sphere of confusion and limited reflections obtained from the grains due the monochromatic radiation. In 2002 the first strain experiments were published using polychromatic micro-X-ray diffraction were carried out on thin samples by B. C. Larson et al. [4]. Polychromatic X-rays have the advantage that they can be used at a single sample position while still providing statistics from many grains without rotation. This diminishes potential strain error due to displacement and complements the addition of auxiliary equipment such as a stress-rig or furnace. The technique also provides non-destructive mapping of grain shapes for both deformed and un-deformed grains.

A new technique developed at the Advanced Light Source (ALS), uses polychromatic X-ray Diffraction for determination of the 3-D positions of grains within a sample volume, and the associated strain tensor and orientation with sub-micron resolution as a function of stress (4DXRD). Developments in the detector technology and focusing optics allow precise focusing of the beam with controlled energy levels. High-resolution readouts [5] from the detectors coupled with powerful indexing software enable accurate Laue spot measurement corresponding to individual grains. The Micro-Laue diffraction experiment produces precise spots, which enables determination of the orientation of individual grains with respect to the incident beam. The deviatoric strain tensor is obtained directly from one indexation of the diffracted spots. The full tensor is obtained by including the dilatation strain found by determining the energy of a particular Laue reflection. These strains provide evidence for the testing of mechanical models.

METHOD

The experiment was conducted on the micro-diffraction station at Beamline 7.3.3 of the Advanced Light Source, which has the capability of producing a highly focused micro-beam of polychromatic X-rays with a beam cross section of $0.7 \times 0.8\ \mu m^2$ using Kirkpatrick-Baez mirrors [6, 7]. A model fiber matrix composite consisting of a single crystal central fiber approximately

140 μm in diameter, embedded within a polycrystalline aluminum A356 matrix with a millimeter cross sectional area was used.

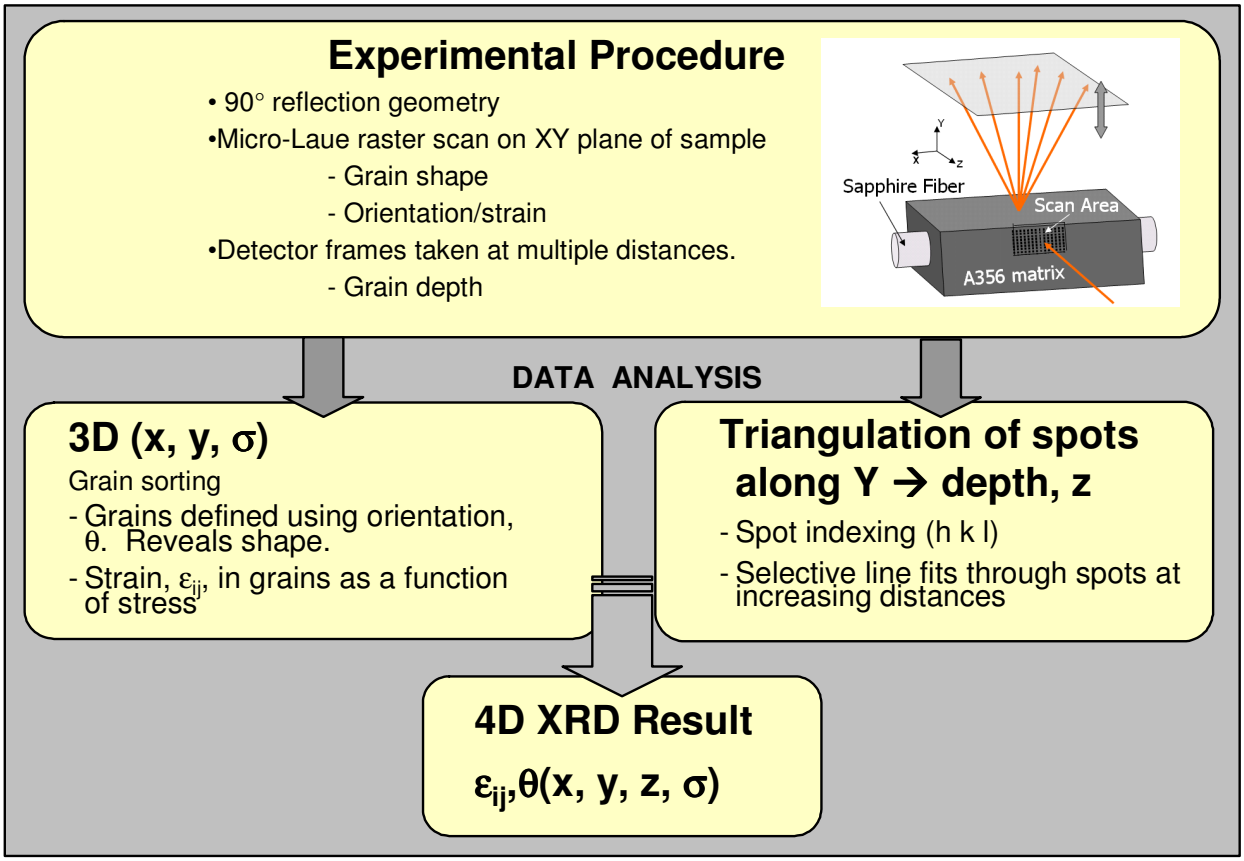

Figure 1. Diagram of 4DXRD analysis.

The sample was held with grips in a customized load frame,[†] which was mounted on the translation stage of the XRD microscope [8]. The sample was rastered across the micro-beam over an area of 0.5 x 0.3 mm^2. Up to 1000 points were selected for diffraction in the analyzed area. Each point produces a Laue spot pattern over the CCD, which is arranged in a 90° reflection geometry as shown in Figure 2. The Laue spot data acquisition was automated by the XMAS software developed by N. Tamura [9]. Mapping grain shapes perpendicular to the beam direction requires incremental translation of the sample stage in x and y. Here the triangulation procedure was carried out along a line coincident with the fiber axis, since behavior of grains lying close to the fiber were of significant interest. To maximize the efficient use of beam-time, data used for triangulating the depth of grains was taken at points spaced 0.05 mm apart along the x-direction. Triangulation is optional at any point throughout the rastered area. An optimal step size depends on the grain size and curvature expected in the depth direction.

[†] http://www.fullam.com

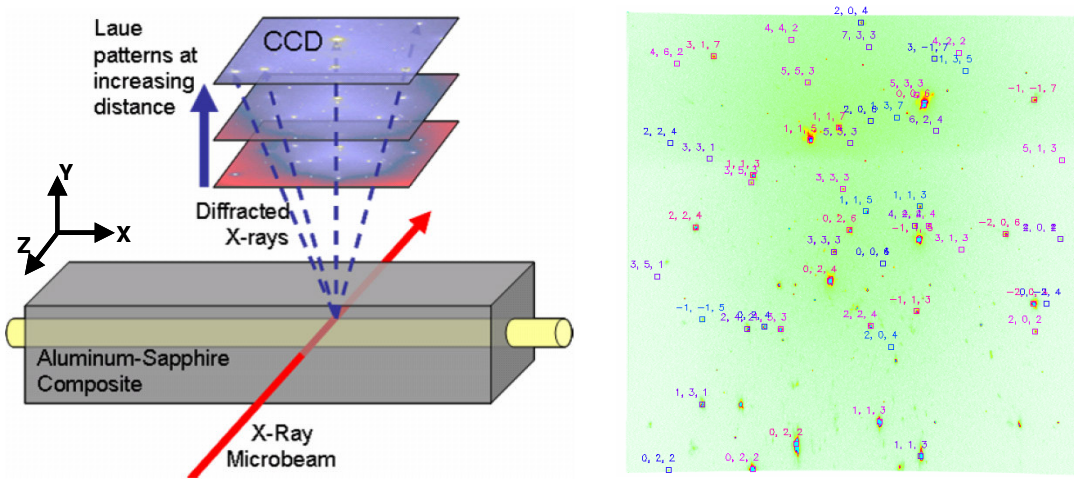

Figure 2. a) Schematic of the triangulation technique. The CCD was moved along Y in steps of 5 mm and corresponding reflections collected. b) 5 individual grains indexed at a single point from triangulation.

With sufficient count statistics, every scan point reveals the grain orientation and deviatoric strain tensor for the different indexed grains. As the detector is moved away from the sample the detector intersects a smaller fraction of reciprocal space and the number of diffraction spots decreases (their resolution increases).

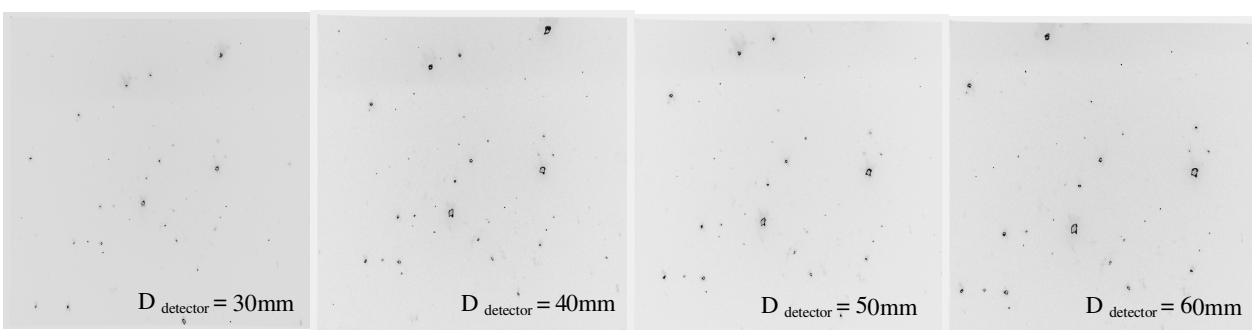

Figure 3. Laue spots of 5 different indexed grains as detector-to-sample distance is increased.

Grain shapes are obtained by grouping reflections according to grain orientation. Reflections from a particular grain orientation and located in the same vicinity (contiguity criterion) are considered to be from the same grain. The spatial resolution of the information over any grain along the plane perpendicular to the beam direction can be improved by increasing the number of points in the raster scan. Knowledge of the depth of the grains (Z) within the sample is determined by triangulating the center of Laue spots observed from the CCD translation. To eliminate potential strain error, the triangulation is done immediately after the XY scan so that strains are not affected by CCD translation.

In the 4DXRD method, the three dimensions correspond to the spatial information of any grain and the fourth dimension is dependent on time. Here, stress was increased after each scan to observe the resulting strain response. The composite was observed under tensile stresses until failure. The current paper presents results under globally elastic deformation. Examination of the remaining stresses requires further analysis.

Strain Analysis

Strains are calculated based on deviations of spot positions from the unstrained position. To determine the absolute lattice spacing of the reflections, a 4-crystal monochromator is inserted into the beam and the energy is scanned while monitoring the reflection intensity on the CCD.[10, 11] This is particularly necessary to determine the dilatational component of the strain, which in combination with the deviatoric strains from the micro-Laue experiment completes the whole strain tensor, calculated with respect to the unstrained unit cell parameters (literature values are used). Strain accuracies are significantly dominated by factors such as number of reflections used for indexing, the quality of peak fit, geometrical calibration of the CCD-to-sample distance and measurement of the absolute d-spacing.

By definition of the strain tensor,

$$\varepsilon_{ij} = \varepsilon = \begin{pmatrix} \varepsilon_{11} - \Delta/3 & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{12} & \varepsilon_{22} - \Delta/3 & \varepsilon_{23} \\ \varepsilon_{13} & \varepsilon_{23} & \varepsilon_{33} - \Delta/3 \end{pmatrix} + \begin{pmatrix} \Delta/3 & 0 & 0 \\ 0 & \Delta/3 & 0 \\ 0 & 0 & \Delta/3 \end{pmatrix} \quad (1)$$

$$\text{With } \Delta = \varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33} \quad (2)$$

The first term represents the deviatoric strain tensor (ε'_{ij}) and the second is called the dilatational strain term. The matrix can be expressed in six independent terms. If the dilatational term is ignored, there are 8 unknowns in total, including the unknown crystal orientation (3 parameters) and deviatoric strain parameters (5 parameters). Knowing the unstrained unit cell parameters and using at least four reflections, the unknowns can be solved to determine the orientation and deviatoric strain parameters. The determination of unit cell parameters and orientation, in addition to the measurement of the energy of a single reflection to obtain the *d*-spacing and the corresponding magnitude of the reciprocal lattice point, leads to the determination of the cell volume and total strain tensor. Of the several possible methods for energy discrimination, insertion of a monochromator crystal has been implemented at 7.3.3. A method of non-linear least-squares refinement of Laue patterns from a calibration sample (we use here the unstrained sapphire fiber) is used to refine the geometrical parameters. Hence average local strain tensors are determined at every scan point on the sample from the unit cell parameters determined earlier. Spread out spots from aluminum make exact location of the centre of spots difficult. To limit strain error, additional reflections at every scan point are included in the calculation of the deviatoric strain tensor. Strain accuracies are on the order of 2×10^{-4}. [8]

RESULTS

3D Grain morphologies and strain

The A356 matrix sample consists of a large number of grains with varying sizes. Hence, the preliminary step in analyzing the experimental output was to sort out and subject the analysis to a subset of large grains among the whole range of grains that show up due to polychromatic radiation. Sizes of the grains were sorted by histogram of the number of coordinates for each grain. Based on the histogram, 16 grains were sorted out of almost 800 grains that were indexed by the software based on the sectional area of the grain (>100 μm^2) within the illuminated sample volume of 0.5 x 0.3 x 1.1 mm^3. Approximately 1200 orientations representing grains and sub-grains were observed.

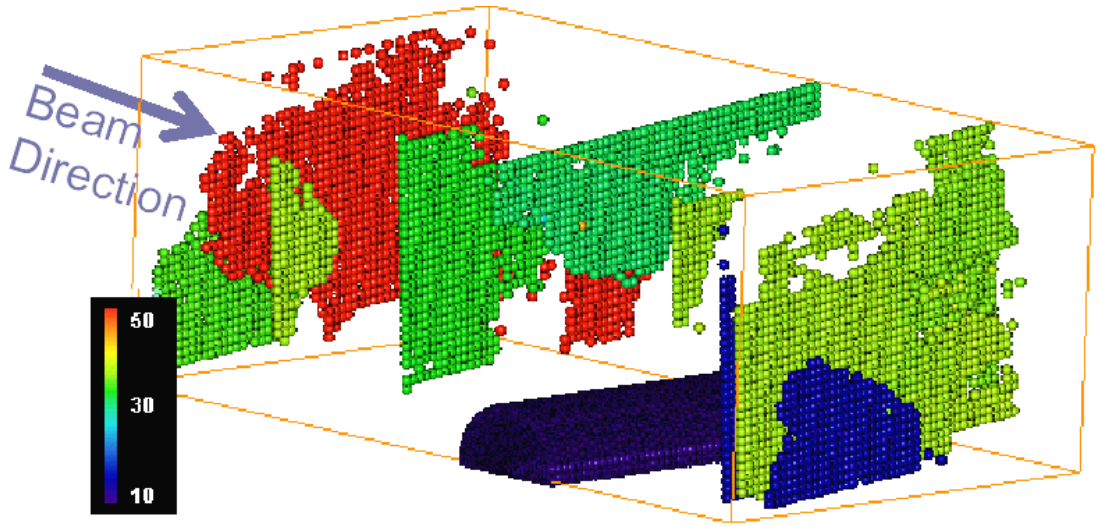

Figure 4. Grain morphologies arranged in accordance with the depth determined by triangulation. Each slice demonstrates the cross section of grains with color showing the orientation angle. The c-axis of the fiber is along the cylindrical axis.

Figure 4 shows grain slices in the XY plane, each slice depicts the cross section of the grain found at a particular location along Z, the colors on the slices indicating the orientation angle with respect to the beam at no applied stress. Corresponding to change in applied load from 0 to 31 N the change in orientation angles for all the grains were less than 1°. The orientation angle is used to identify reflections from particular grains across space. In addition, the consistency of orientation angles with increase in load aid in identifying individual grains as a function of stress.

Results of grain depth profiles using triangulation

Figure 5 summarizes the depth profiles of grains within the sample volume. The measured positions of individual grains along the beam direction are shown with numbered labels.

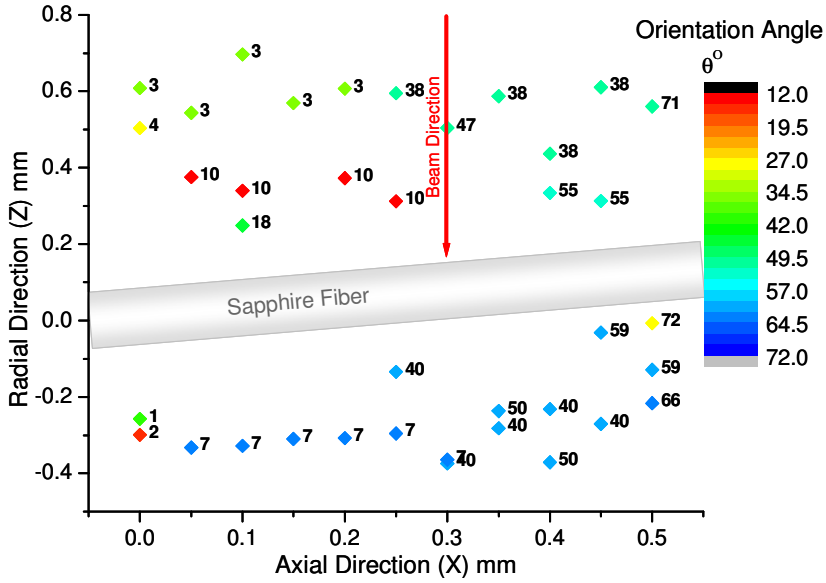

Figure 5. XZ Plot. Result of triangulation at 10 points along the fiber axis. Every point with a number indicates local depth of that grain in the sample along the beam direction.

The grain depths were measured with an accuracy of ±25 microns along the beam direction (see Figure 7). The fiber gave the highest depth resolution. Grain depth in general is indicative of a "diffraction center of mass" which is nominally identical to the true center of mass for grains which are small relative to X-ray attenuation. For the range of energies here, 5 keV penetrates 20 µm of Al ($G_{50\%}$) and 14 keV 500 µm ($G_{50\%}$). Using the analyzed depth related information from Figure 5, a 3D representation of the entire sample volume is constructed. The three dimensions are shown in Figure 6 containing spatial data and the color the deviatoric strain component , ε'_{xx} (along the fiber axis).

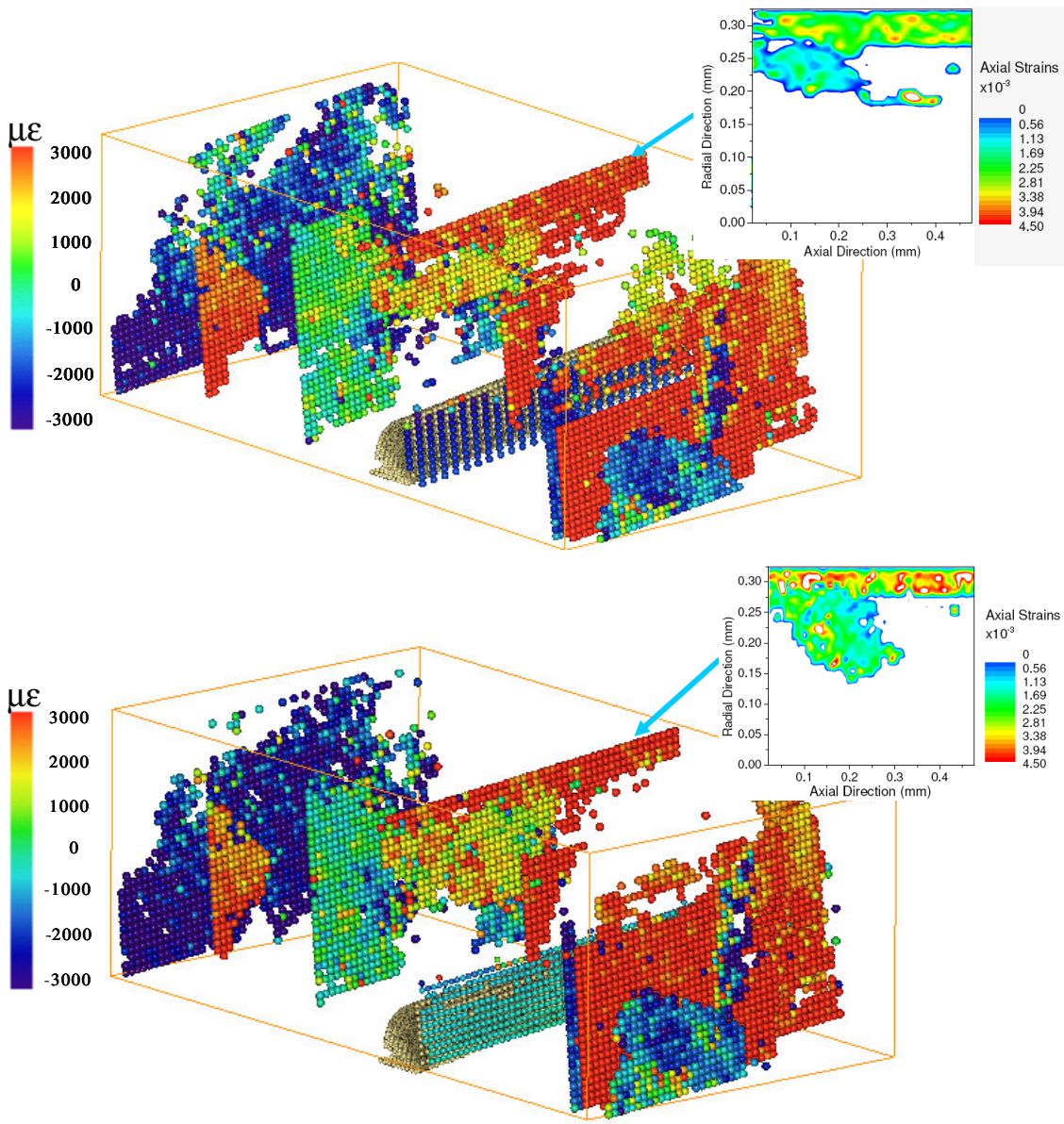

Figure 6. 4D(x,y,z,θ) strains from the first and second applied stress on the composite. The inset shows the change in strain for a single grain.

The change in strains or the two loads is small compared to the span of residual strains initially in the matrix grains; thus, strain changes do not appear distinctly in the 3-D plots (Figure 6). The 2D strain plots shown on the right in the figure uses a finer strain scale for the specific grain

indicated. Then, the change in strain due to stress is clear. However, even in 3-D, the effect of applied load in the fiber is readily observed. This is an indication of the strong bond at the fiber interface leading to load partitioning between the phases.

Triangulation of the spots originating from a particular grain result in the depth measurement of the grain within the sample volume. Besides the dependence on goodness of fit for the individual indexed spots, the depth measurement error is dependant on the number of exposures and the distance between individual exposures. More triangulation exposures on the CCD result in a reduced error in the grain depth measurement. Figure 7 shows the error variation with increased number of exposures. In addition, a comparison is made concerning the detector travel distance between exposures. Though initial error in case of the closely spaced exposures is high, it converges to 25 μm as the number of exposures exceed 8. Apart from the number of exposures, the measurement is dependant on spot distribution over the CCD. As the CCD is moved away from the sample, the number of spots on the CCD decrease, which leads to inaccuracies in the fit. This is the source, evident in Figure 7, of the increase in error in Grain #1 after 8 exposures in comparison with the other two grains and of the more significant error in Grain #1 observed for the two exposures taken at 40 mm steps.

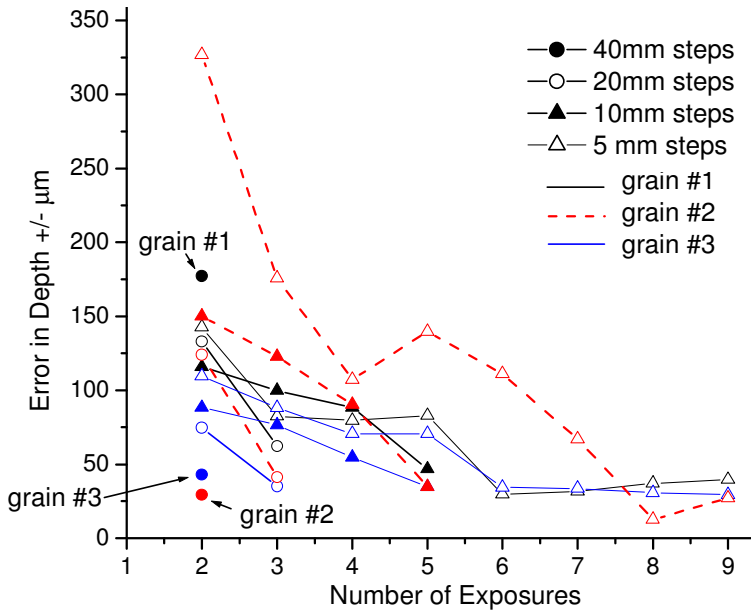

Figure 7. Decrease in depth measurement error by increasing the number of triangulation exposures. The error trend for three different grains indexed at a single scan point is shown.

DISCUSSION

The depth resolved method of locating grains with respective local strain data is useful in probing bulk samples. The method can be successfully applied to polycrystalline materials with grain sizes larger than 1μm. A profile of the shape of the grain is obtained from its set of reflections throughout the raster scan. With the current beam size, the spatial resolution of the grains in the plane perpendicular to the beam can be a micron. The spatial resolution in the depth direction is limited by grain size. Here the grain position was identified to within 2% of the specimen width.

An alternative depth profiling method, the differential aperture X-ray diffraction method (DAXM), which also utilizes polychromatic radiation can potentially offer greater depth resolution for large grains, but for the same number of exposures, it is limited to shorter depth spans along the beam. Here only two exposures are necessary to obtain 30 μm depth resolution with increasing exposures significantly increasing the precision only for grains with few reflections. However, the high depth resolution with DAXM (1 μm) offers the potential resolution of grain boundaries in the depth direction. For this sample, 44 exposures would be required to give the same depth resolution as triangulation.

Currently much of the experiment and analysis, such as spot indexation and strain refinement, is automated by the XMAS software. However, development remains for automation in the depth profiling procedure. The individual indexed grains are manually selected, depending upon number of reflections, and then triangulated for depth measurement. Some grains have weaker and fewer reflections, which results in higher uncertainty or error in the triangulated depth value.

Visualization of the internal structural arrangement with high resolution is also possible with other methods such as serial sectioning. Similar to other 3-D XRD work [12], comparison of these results with a sectioning technique can provide validation for the spatial information obtained from 4DXRD experiments. However, serial sectioning is destructive and cannot be extended to 4D (in-situ determination of local strain as a function of stress).

4DXRD provides valuable non-destructive in-situ determination of the intra-granular as well as inter-granular strain, which provide experimental evidence for the development of micro-mechanical models. The current experiment has results pertaining to residual strains within the grains and strains at applied load in the elastic limit. Grains persist in a compressive strain state well into the applied tensile stress.

CONCLUSION

The sub-grain strain, grain-grain strain, and grain orientations in a metal matrix composite were observed as a function of stress in 3-D space. Depth information was obtained using triangulation of polychromatic micro-Laue X-ray diffraction spots. Only a few exposures with detector translation are necessary to add depth resolution to the data with error decreasing with the increase in exposures. Two exposures 40 mm apart gave similar resolution to ten exposures 5 mm apart when many indexed diffraction spots were available. Results show orientation and strain maps of grains that have been located through the depth of the sample. Orientation changed little as a function of applied stress—providing a means to identify grains across stress as well as space. However, grain boundaries were resolved only in the plane perpendicular to the beam. The results aid our understanding of deformation in aluminum and characteristics of metal-matrix composites.

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REFERENCES

[1] Böhm, J. A.; Wanner.; Kampmann, R.; Franz, H.; Liss, K.-D.; Schreyer, A.; Clemens, H.; "Internal stress measurements by high-energy synchrotron X-ray diffraction at increased specimen-detector distance." Nuclear Instruments and Methods in Physics Research B 2003, 200, 315–322.

[2] Poulsen, H.F.; Nielsen, S. F.; Lauridsen, E. M.; Schmidt, S.; Suter, R.M.; Lienert, U.; Margulies, L.; Lorentzen, T. D.; Juul Jensen; "Three-dimensional maps of grain boundaries and the stress state of individual grains in polycrystals and powders." J. Appl. Cryst. 2001, 34, 751–756.

[3] Margulies, L.; Winther, G.; Poulsen, H. F.; "In situ measurement of grain rotation during deformation of polycrystals," Science, 2001, 291 (5512), 2392-2394.

[4] Larson, B.C.; Yang, W.; Ice, G.E.; Budai, J.D.; Tischler, J.Z.; "Three-dimensional X-ray structural microscopy with submicrometre resolution," Nature 2002, 415, 887–890.

[5] Bruker SMART 6000 CCD at http://www.bruker-axs.de/index.php?id=158

[6] Tamura, N.; MacDowell, A. A.; Spolenak, R.; Valek, B. C.; Bravman, J. C.; Brown, W. L.; Celestre, R. S.; Padmore, H. A.; Batterman B. W.; Patel, J. R.; "Scanning X-ray microdiffraction with sub-micrometer white beam for strain/stress and orientation mapping in thin films", J., Synchrotron Rad., 2003, 10, 137-143.

[7] Tamura, N.; Spolenak, R., Valek, B.C.; Manceau, A.; Meier Chang, M.; Celestre, R.S.; MacDowell, A.A.; Padmore, H.A.; Patel, J.R.; "Submicron x-ray diffraction and its applications to problems in materials and environmental science" Review of Scientific Instruments, 2002, 73, 1369-1372.

[8] Bale, H. A., Hanan, J. C., Tamura, N., "Average And Grain Specific Strain Of A Composite Under Stress Using Polychromatic Microbeam X-Rays," Adv. X-Ray Anal., 2005, 49.

[9] http://xraysweb.lbl.gov/microdif/user_resources.htm

[10] Chung, J.-S.; Ice, G. E.;. "Automated indexing for texture and strain measurement with broad-band-pass X-ray microbeams," J., Appl. Phys. 1999, 86, 5249–5255.

[11] Yang, W.; Larson, B.C.; Ice, G.E.; Tischler, J.Z.; Budai, J.D.; Chung, K.S.; Lowe, W.P.; "Spatially resolved Poisson strain and anticlastic curvature measurements in Si under large deflection bending", Appl. Phys. Lett., 2003, 82, 3856–3858.

[12] Chawla, N.; Ganesh, V. V.; Wunsch, B.; "Three-dimensional (3D) microstructure visualization and finite element modeling of the mechanical behavior of SiC particle reinforced aluminum composites," Scripta Materialia, 2004, 51, 161–165.