Determination of Texture From Individual Grain Orientation Measurements

John E. Blendell, Mark D. Vaudin and Edwin R. Fuller, Jr.
Ceramics Division
Materials Science and Engineering Laboratory
National Institute of Standards and Technology
Gaithersburg, MD 20899 USA

Abstract

We present a technique for determining the texture of a polycrystalline material based on the measurement of the orientation of a number of individual grains. We assumed that the sample has fiber (i.e. axisymmetric) texture and that the texture can be characterized by a function (the March-Dollase function) with a single parameter. We simulated a large number, N, of orientation data sets, using the March-Dollase function for a total of five different texture parameters, r^{init}. Using the maximum likelihood method we solved for the texture parameter, r′, that best fits each simulated data set in order to determine the distribution of r′ and evaluate the precision and accuracy with which r′ can be determined. The 90% confidence limits of the ratio r'/r^{init} varied as $N^{-½}$ but were independent of r^{init}. Using the texture of slightly textured alumina as determined by x-ray diffraction we calculated the 90% confidence limits for measurements of 131 grains. The orientations of 131 grains in textured alumina were measured by electron backscatter diffraction and the texture determined from those measurements lay within these 90% confidence limits.

Introduction

With the development of techniques for the rapid determination of the orientation of single grains in the surface of a polycrystalline sample[1] there is the potential for determining the preferred crystallographic orientation, or texture, of a sample. Typically the crystallographic texture of a polycrystalline sample is determined by diffraction techniques. Pole figure[2] and rocking curve analysis[3] are the most commonly used techniques, but for samples that exhibit

fiber texture, where the preferred orientation of the crystallites is axisymmetric about a sample axis (texture axis), Rietveld analysis of conventional theta - two theta scans can also be used to measure the texture[4,5]. All these techniques give the average texture over the area illuminated by the x-ray beam. There are cases where it may be required to measure texture over a smaller scale, either to characterize small specimens or to investigate local texture variations in a larger specimen, and in these cases, a smaller probe, such as an electron beam in a scanning electron microscope, can be used. The questions that arise are: how to compare the texture results from different techniques; and how many individual grains must be measured in order to achieve the desired accuracy and precision. In order to compare the measurements on individual grains to the results of Rietveld analysis of theta - two theta x-ray diffraction (XRD) scans, only samples with fiber texture will be considered.

Texture arises when the crystallites that make up a polycrystalline sample do not have a random arrangement of their crystallographic orientations. To describe axisymmetric texture, we define a crystallographic direction (the preferred orientation direction) that is preferentially aligned with the texture axis. The preferred orientation is typically specified as the normal, **n**, to a specific crystallographic plane, (hkl); the texture axis is usually a sample direction or processing axis. The conventional measure of the degree of texture is the ratio of the volume fraction of crystallites in a textured sample with **n** at a specific orientation to the texture axis to the same volume fraction for a random (or untextured) sample. This ratio is called the multiple of a random distribution, MRD. If we assume that the diffracted intensity for a specific Bragg reflection in a diffraction pattern is proportional to (among other parameters) the volume fraction of crystallites correctly oriented to diffract into the Bragg peak, then one measure of the MRD profile of a polycrystalline sample is to compare the intensity of the diffraction peaks to the intensity of the same diffraction peaks for a random or untextured sample. The functional relationship of MRD to the angle between the texture axis and the orientation of the crystallites can be modeled for the case of axisymmetric rod or disk shaped crystallites.

Texture Model

The model MRD function chosen for this work is the March-Dollase function[5] that has been incorporated in software packages (such as GSAS[6] - a Rietveld technique) used to analyze powder diffraction data. In these techniques, a number of functions which model the experimental and sample conditions are used to calculate a model diffraction pattern. The parameters in each model are adjusted to minimize the error between the experimental data and the pattern calculated from the model functions. The function, $P(r, \alpha)$, models the MRD for crystallites with orientation **n** at an angle α to the texture axis using a single parameter r:

$$P(r, \alpha) = [r^2 \cos^2\alpha + r^{-1} \sin^2 \alpha]^{-3/2} \qquad (1)$$

We set $M = P(r, 0) = r^{-3}$; M is the MRD at $\alpha = 0$, and it is frequently used as a parameter for describing crystallographic texture. For a random sample the probability of a given crystallite orientation is uniform over orientation space. The probability of a volume element with **n** lying at an angle α to the texture axis is proportional to $\sin \alpha$. Thus, for the textured specimen, the probability of a given crystallite orientation is $P(r,\alpha) \sin \alpha$.

Recently developed techniques allow the measurement of the crystallographic orientation of individual grains in the surface of a polycrystalline sample[1]. Such data can be used to determine the texture of the sample by fitting the data to Eq. (1). One method of doing this requires the data to first be put into bins to form a histogram. The March-Dollase equation is then fitted to the histogram data by allowing r to vary. This technique may lead to poor results if the number of grains measured is low; in addition, the results are dependent on the bin width.

An alternative technique for fitting the experimental data is the Maximum-Likelihood Method[7]. If we assume that the sample MRD distribution is given by the March-Dollase function, Eq. (1), then the probability of making any single grain measurement resulting in a orientation of α_i, is $P(r,\alpha_i)\sin(\alpha_i)$, which we will call the March-Dollase distribution. The measurements of the orientations, α_i, of the individual grains (the angle of the crystallographic axis to the texture axis) give a set of N orientations that are assumed to be randomly taken from the entire sample orientation distribution with parameter value r. We assume that the texture of the sample is homogeneous and that the grains on the polished surface examined in the SEM are representative of the bulk grains. While this may not be true for a fired surface, it will be the case for an internal

section. Given an experimentally measured (or simulated) set of orientations, taken from a population with an unknown degree of texture, r, the problem is to determine an estimate of the degree of texture of the entire sample orientation distribution. The estimate, r′, is the one that maximizes the likelihood that the sample set came from a population with degree of texture, r′. The relationship of r′ to r and the confidence limits on r′ will be investigated, and related to the number of orientation measurements in the set.

For the chosen estimate, r′, the probability of measuring a value of α_i is given by the probability function, $P(r', \alpha_i) \sin(\alpha_i)$. For the entire set of N orientation measurements the probability of getting that particular set, L(r′), (the likelihood function), is given by the product of the individual probability functions,

$$L(r') = \prod_{i=1}^{N} P(r', \alpha_i) \sin(\alpha_i) \qquad (2)$$

In order to solve Eq. (2) for r′, we take the logarithm of both sides to convert the product to a sum. This yields

$$\ln(L) = \sum_{i=1}^{N} \ln\left(P(r', \alpha_i) \sin(\alpha_i)\right) \qquad (3)$$

Substituting $M' = r'^{-3}$, Eq. (3) can be rewritten as:

$$\ln(L) = \sum_{i=1}^{N} \ln(M') - \frac{3}{2} \ln\left[M' - (M' - 1)\cos^2 \alpha_i\right] + \ln \sin \alpha_i \qquad (4)$$

In the method of maximum likelihood, the value of the estimator, M′, that has the highest probability is assumed to be the best value for M, the parameter for the whole population. In order to find the maximum of the likelihood function, the derivative of Eq. (4) with respect to M′ is set equal to zero and then solved for M′.

$$\sum_{i=1}^{N} \left[\frac{M'(1 - \cos^2 \alpha_i)}{\cos^2 \alpha_i + M'(1 - \cos^2 \alpha_i)} \right] - \frac{2}{3} N = 0 \qquad (5)$$

Thus the problem of determining the texture from a set of individual grain orientations, α_i, is reduced to finding the value of the texture parameter, M′, which solves Eq. (5).

Simulation

In order to determine the conditions where the maximum likelihood method yields a better estimate of the texture than fitting the data to a histogram, and to determine if there is any bias in the technique, we have simulated experimental results using a Monte Carlo method. First a random set of angles, α_i, is chosen from a March-Dollase distribution with a given texture parameter, r^{init} or M^{init}. Then the maximum likelihood technique is used to find an estimator of the texture parameter, r′, for that set of angles. This process is repeated a large number of times and the average and distribution of the texture estimator is compared to the initial texture parameter, r^{init}.

To find the probability of measuring an angle less than or equal to α_o for a sample with a texture parameter M, the function $P(M,\alpha)\sin(\alpha)$ can be integrated from 0 to α_0 to give the cumulative distribution:

$$C(M, \alpha_0) = \int_0^{\alpha_0} P(M, \alpha) \sin\alpha d\alpha \qquad (6)$$

which varies from 0 to 1 as α_o varies from 0 to 90°. In order to generate a set of angles, α_i with a March-Dollase distribution, we want to randomly sample probability space. To do this, random numbers, R_i, between 0 and 1 are generated and the value of α_o which results in the cumulative distribution, $C(M, \alpha_o) = R_i$ is taken as the value of α_i. Since $\frac{d}{d\alpha} C(M, \alpha) = P(M, \alpha_0) \sin \alpha_0$, this results in a set of angles that fit the March-Dollase distribution.

Eq. (6) can be solved and inverted to give $\cos(\alpha_i)$ for an initial texture parameter, M^{init}.

$$\cos^2 \alpha_i = \frac{(1 - R_i)^2 M^{init}}{1 + (1 - R_i)^2 (M^{init} - 1)} \qquad (7)$$

Using the maximum likelihood method, we want to solve for the M′ value for the generated set of α_i values. Substituting Eq. (7) into Eq. (5) gives

$$\sum_{i=1}^{N}\left[1+\left(\frac{M^{init}}{M'}\right)\left(\frac{(1-R_i)^2}{1-(1-R_i)^2}\right)\right]^{-1}-\frac{2}{3}N=0 \qquad (8)$$

The variable in Eq. 8 is the ratio M'/M^{init}, therefore, the results are expected to fall on a master curve independent of N or M^{init}. We also fitted the simulated data by putting the data in bins and fitting the resultant histogram. Bin widths were varied from 15° to 3.75° and the March-Dollase distribution was fitted to the data using a least squares analysis.

Results

Sets of α_i were generated for M=8, 27, 64 and 125 (corresponding to r = $1/2, 1/3, 1/4, 1/5$). The sets contained 10, 20, 50, 100, 200, 500, 1000 or 10000 grain orientations. For each set, M'/M^{init} was found using Newton's method to solve Eq. (8) and this was repeated 10000 times. The mean of M'/M^{init} and the 5% and 95% limits were determined and are shown in Figure 1. It is seen that the data normalized by the initial M^{init} of the simulation lie on a single curve. Also, there is a bias in the calculated values of $\overline{M'}/M^{init}$ for small numbers of grain orientations measured. For 10 orientations measured, the maximum likelihood method gave an estimate of $\overline{M'}/M^{init}$ that is 4% too small but the error decreases to 2% for 20 orientations measured and decreases to less than 1% when more than 40 orientations are measured.

The 90% confidence limits are seen to decrease as the number of orientations measured increases. The accuracy (standard deviation) of M'/M^{init} was found to vary approximately as $N^{-½}$ which suggests that the main source of error is random error and not a systematic error due to the maximum likelihood technique. The slope of the standard deviation as a function of N was -0.518. When only the values for more than 100 orientations are used, the slope decreased to -0.503.

For fitting the March-Dollase distribution to a histogram of the data, the results were found to be dependent on the bin width chosen. The value of M'/M^{init} was always smaller than the corresponding value from the maximum likelihood method except for the case of a 15° bin size and an M^{init} value of 125 (r^{init} =0.2). Thus, the results were dependent on the initial texture value chosen for the simulation and did not fall on a single curve when normalized by M^{init}. When both the bin size and data set are small, it is likely that some bins will have no orientations which makes the fitting inaccurate. When analyzing highly textured samples, the orientation density may change significantly across the bin so that the center of the bin will not accurately represent the average value of the data in the bin. While there are methods for adjusting the bin size and location based upon the data, these methods may introduce artefacts which can be avoided by using the maximum likelihood method.

The maximum likelihood method was used to calculate the degree of texture for a set of 131 grain orientations measured using backscattered Kukuchi patterns (EBSP) generated in a SEM[8]. The sample was a polycrystalline Al_2O_3 substrate (SRM 1976[9]) and the fired surface was examined. The grain size was from 1 μm to 10 μm and the orientation measurements were taken every 100 μm, so that the sampling position was chosen at random, no grain was sampled twice and there was no knowledge of the size of each sampled grain. The EBSP data was analyzed using both the maximum likelihood method, and by putting the data into bins and fitting the March-Dollase distribution to the resulting histogram. From the histogram data, M′=6.81 (r=0.5277); and from the maximum likelihood method, M′=6.20 (r=0.5443).

For comparison, the sample texture was also measured using an x-ray technique. Measurement of weak [0001] texture in Al_2O_3 cannot simply be performed by measuring the intensity diffracted by the basal planes using the 0006 or 000.12 peaks since both those peaks have low structure factors and are extremely weak; therefore they can only be used in rocking curve or single pole figure measurements on alumina samples with considerable texture. Therefore, the texture was measured by performing a Rietveld refinement of standard theta-two theta x-ray diffraction data using GSAS. From the Rietveld refinement, the texture parameter M was found to be 4.28 (r=0.616) indicating that the sample has some small degree of [0001] texture for a texture axis normal to the surface of the substrate.

In order to estimate the confidence limits, the simulation was run for the case of 131 orientations with a M^{init} of 6.20. The average of 10,000 iterations was M = 6.14 (r=0.5460) and the normalized 90% confidence limits for 131 orientations are $M'(5\%)/M^{init}$= 0.723 and $M'(95\%)/M^{init}$= 1.373. This implies that based on the measured M′, the true M of the sample, with a 90% confidence limit, would lie between 4.46 and 8.00. It is seen that the M value determined from the measurement of the orientation of individual grains is above the measured x-ray M value. For an M^{init} of 4.28 (r = 0.616) the simulation yielded an M of 4.07 (r = 0.626) with 90% confidence limits of M = +2.23, -1.29 (r = +0.1504, -0.1278, respectively). Thus for measurements of 131 grains from a sample with a texture parameter of 4.28, 90% of the time, the measurements would lie between M=2.78 and M=6.30.

It is seen that the single measurements we have made is within the range of expected values of the texture parameter for the sample, but it is at the high end of the range. This reflects the errors associated with the small number of grains and also may be due to the differences between the surface grains measured by EBSP and the grains measured by x-ray diffraction. Thus the discrepancy between the x-ray and EBSP measurements of the texture parameter is within the expected variation of the techniques and does not indicate a difference in texture.

The simulation we have performed allows an estimation of how different the texture parameters must be in different areas of a sample, or between two samples, in order for the texture of the regions to be considered distinct. As the above measurements show, different values of M may just reflect the statistical variations for small sample size. Any technique based on individual grain orientation measurements will be subject to these uncertainties. Clearly, if x-ray diffraction techniques can be used, the results will be a more accurate measure of the texture of the whole sample than to the individual grain orientation measurements. The advantage of individual grain orientation measurements is that the technique used to make the measurements has a probe which is smaller than the scale of the microstructure. This allow texture to be determined on a much finer scale as compared to x-ray techniques. Thus local variations in texture can be measured. The advantage of the maximum likelihood method of analyzing the orientation data,

compared to a histogram, is that the results do not depend on the initial conditions, such as bin width for fitting a histogram.

Summary

We have developed a method for evaluating the accuracy of the measurement of crystallographic texture of polycrystalline materials from individual grain orientation measurements. We assume that for samples that have fiber texture, the March-Dollase distribution describes the orientation distribution of the crystallites. By fitting the orientation measurements to the March-Dollase distribution, the texture can be determined. The fitting is based on the maximum likelihood method which yields a better fit for small numbers of measured grains than the techniques of binning the data into a histogram and fitting the texture function to a histogram. In order to determine the accuracy and precision of the technique, we have simulated grain orientation distributions and fitted them to the March-Dollase distribution using the maximum likelihood method. It was found the there was a bias in the calculated texture for small numbers of grains measured, but if more than 100 orientations were measured, the error was less than 1%. The 90% confidence limits decreased as $N^{-½}$ where N is the number of orientations measured.

In order to compare the method to standard x-ray diffraction techniques, measurements were made on a polycrystalline alumina sample. The method was used to compare the results from the measurement of the 131 grains on the fired surface of the sample and the texture parameters from the two techniques were found to be within the 90% confidence limits. Thus, the maximum likelihood method is found to be a good method of fitting a model texture function to a data set of orientations to determine the texture of a polycrystalline sample.

Figure Captions

Figure 1. Maximum likelihood texture results ($\overline{M'}/M^{init}$) from simulations of N individual grain orientations with varying texture levels (M^{init}).

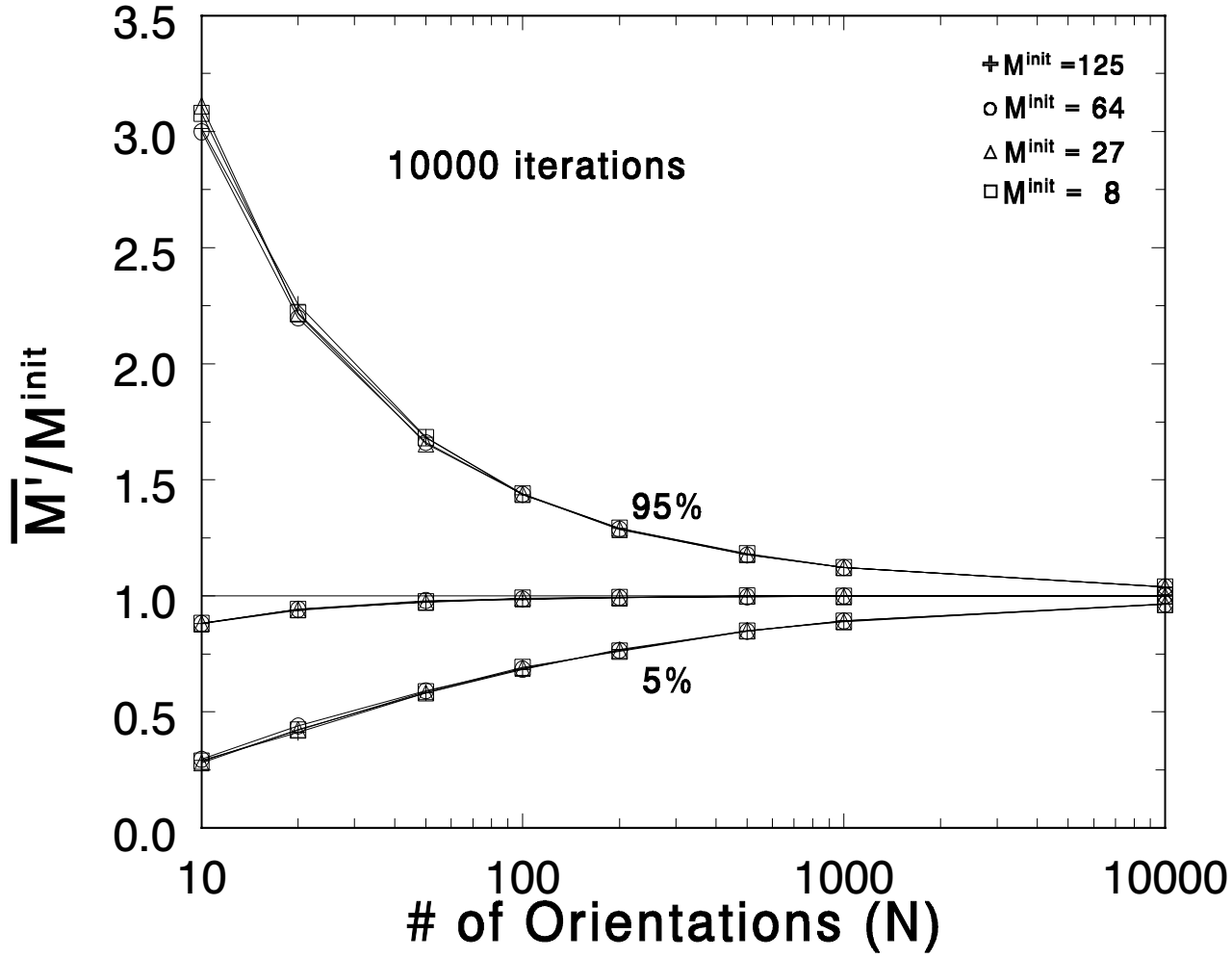
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
10
100
1000
10000
# of Orientations (N)
$\overline{M'}/M^{init}$
10000 iterations
+ M^{init} =125
○ M^{init} = 64
△ M^{init} = 27
□ M^{init} = 8
95%
5%

Figure 1

References

1. B.L. Adams, S.I. Wright and K Kunze, "Orientation Imaging: The Emergence of a New Microscopy", Met. Trans., 24A, 819-831, 1993.

2. H.P. Klug and L.E. Alexander, X-Ray Diffraction Procedures, J. Wiley, New York, 1974.

3. M.D. Vaudin, M.W. Rupich, M. Jowett, G.N. Riley, Jr., and J.F. Bingert, "A Method For Crystallographic Texture Investigations Using Standard x-ray Equipment", in press, J. Materials Research, 1998.

4. J.P. Cline, M.D. Vaudin, J.E. Blendell, C.A. Handwerker, R. Jiggetts, K.J. Bowman and N. Medendorp, "Texture Measurement Of Sintered Alumina Using The March-Dollase Function", Advances in X-Ray Analysis, 37, 473-478, 1994.

5. W.A. Dollase, "Correction of Intensities for Preferred Orientation in Powder Diffractometry: Application of the March Model", J. Appl. Cryst. 19, 267-272, 1986.

6. A.C. Larsen and R.B. Von Dreele, General Structure Analysis System, Rietveld Analysis Software, LANSCE, MS-H805, Los Alamos National Laboratory, Los Alamos, NM 87545.

7. P.R. Bevington and D.K. Robinson, Data Reduction and Error Analysis for the Physical Sciences, McGraw-Hill, Inc, New York 1992.

8. M.D. Vaudin and W.C. Carter, "Studies of Ceramics by use of Backscatter Diffraction Patterns in the Scanning Electron Microscope", Microbeam Analysis, 152, 1991.

9. Standard Reference Material 1976, National Institute of Standards and Technology, Gaithersburg MD 20899.