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ULTRASONIC CHARACTERIZATION OF TEXTURE IN PURE AND ALLOYED ZIRCONIUM

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INTRODUCTION

Recently, we have developed a method to relate the angular dependence of ultrasonic velocities to the five expansion coefficients of the crystallographic orientation distribution function (CODF) of hexagonal materials with orthorhombic macroscopic symmetry.¹ The ultrasonic velocity measurements are performed with an acoustic microscope on one, two, or three of the principal planes of symmetry using one or more ultrasonic modes. In this paper, the theory of ultrasonic measurement of texture is reviewed, and we present data showing that it can be applied to two-phase alloys, when the primary phase makes up most of the material.

The paper begins with the expansion of the CODF in terms of generalized spherical harmonics and a discussion of the physical significance of the expansion coefficients W_{lmn} , in terms of the crystallite's c-axis orientations. Various methods used to obtain the W_{lmn} ultrasonically are summarized and discussed using a general theory. Results are presented for a sample of pure zirconium and six samples of Zr-2.5%Nb alloys containing 5 to 10 % beta (cubic) phase. The ultrasonic texture coefficients are compared to those obtained by neutron diffraction. A discussion of measurement errors, second phase effects, and averaging methods follows.

THE CRYSTALLITE ORIENTATION DISTRIBUTION FUNCTION

Texture is described by a crystallite orientation distribution function (CODF) that describes the probability of finding a single crystallite in a specific orientation. The crystallite orientation is described by the three Euler angles, ψ , θ , and ϕ . These angles relate the crystallite's internal coordinates to an external system of coordinates. This paper considers the particular case of hexagonal crystallites in an aggregate of orthorhombic symmetry. In this case, ψ and θ represent the "longitude" and the "latitude" of the crystallite's c axis with respect to the aggregate's axes, and ϕ represents the rotation of the crystallite around its own c axis.

The CODF, denoted here by $w(\psi, \theta, \phi)$, may be expanded in terms of generalized spherical harmonics,

$$w(\psi, \theta, \phi) = \sum_{l=0}^{\infty} \sum_{m=-l}^l \sum_{n=-l}^l W_{lmn} Z_{lmn}(\cos \theta) e^{-im\psi} e^{-in\phi} \quad (1)$$

where $Z_{lmn}(\cos \theta)$ is a generalization of the associated Legendre function, and W_{lmn} are the coefficients of the expansion.² The normalization condition for the CODF is

$$\int_0^{2\pi} \int_0^{2\pi} \int_{-1}^1 w(\psi, \theta, \phi) d(\cos \theta) d\psi d\phi = 1 \quad (2)$$

It has been shown that the effective (second order) elastic constants of a polycrystalline aggregate depend only on expansion terms for which $l \leq 4$.³ This very important result implies that fine details of the CODF will not be measurable with linear ultrasonics. Conversely, all linear acoustic properties and all linear mechanical properties of a polycrystalline aggregate only depend on a small number of texture coefficients.

Because of symmetry properties of orthorhombic aggregates of hexagonal crystallites,² the W_{lmn} are real, $W_{lm0} = W_{l\bar{m}0}$ and only the following texture coefficients with $l \leq 4$ are non zero: W_{000} , W_{200} , W_{220} , W_{400} , W_{420} , and W_{440} .⁴ Moreover, Z_{lmn} always satisfies the relation $Z_{lmn} = (-1)^{m+n} Z_{l\bar{m}\bar{n}}$. Using these relations, we can write

$$w(\psi, \theta, \phi) \approx \sum_{l=0,2,4} \sum_{\substack{m=-l \\ m \text{ even}}}^l W_{lm0} Z_{lm0}(\cos \theta) \cos m\psi \quad (3)$$

To better understand the meaning of the non-zero W_{lm0} , we plot $w_{lm0}(\theta, \psi) = Z_{lm0}(\cos \theta) \cos m\psi$ in spherical coordinates in Figure 1. In Figure 1, the vertical axis is the macroscopic z axis and the circle in the xy plane is labeled in degrees from the x axis. The new basis function, $w_{lm0}(\theta, \psi)$, is related to the probability of finding the c axis of a hexagonal crystallite pointing in the (θ, ψ) direction if all other texture coefficients are zero. Note that $w_{lm0}(\theta, \psi)$ can be positive or negative.

The w_{000} function is independent of θ and ψ , is equal to $\sqrt{2}/2$, and is trivially shown as a sphere in Figure 1. It represents an isotropic distribution. It is also the only term that integrates to a non zero value in the normalization integral (2). Therefore, W_{000} is always equal to $\sqrt{2}/8\pi^2$. The w_{200} function is axisymmetric and has two positive lobes in the $\pm z$ directions and a negative ring in the xy plane. Negative values of $w_{lm0}(\theta, \psi)$ are represented as positive radii in the $(\pi - \theta, \psi + \pi)$ direction. However, the $w_{lm0}(\theta, \psi)$ considered have point symmetry and Figure 1 can be thought of as $|w_{lm0}(\theta, \psi)|$. Although negative probabilities are unphysical, the individual w_{lm0} are never observed individually,

Figure 1. Three dimensional representation of the basis functions $w_{lm0}(\theta, \psi)$ for $l \leq 4$ and even values of m . The vertical axis is the z axis and a circle labeled in degrees from the x axis is drawn in the xy plane. The radii of the circles are: 1.0 for w_{000} , w_{420} , and w_{440} , 1.2 for w_{220} , and 2.0 for w_{200} and w_{400} . The sign of each lobe alternates, as described in the text.

but as part of the expansion (1). In particular, they must be added to the constant w_{000} term. Consequently, positive or negative values of W_{200} indicate that the c axes of the hexagonal crystallites are preferentially aligned along the z axis or in the xy plane, respectively. The w_{220} has two positive lobes along the x axis and two negative lobes along the y axis. Consequently, positive or negative values of W_{220} indicate that the c axes of the hexagonal crystallites are preferentially aligned along the x or y axes, respectively. The w_{400} function is axisymmetric and has two positive lobes along the z axis, one positive ring in the xy plane, and two negative "cones" around the z axis. The effect of w_{400} on texture can be quite complex, but one important effect is to sharpen the w_{200} distribution and to increase the probability of finding the crystallite's c axis aligned along the macroscopic z axis. Alternately, negative values of W_{400} may occur with positive values of W_{200} and a non zero value of W_{220} . This results in a twin peak about the z axis. This texture may be observed in Zr rolled plates when the z axis is defined as the direction transverse to the rolling direction. The function w_{420} is considerably more complicated and consists of 12 lobes of alternating sign, with negative lobes along the x axes. The function w_{440} consists of 8 lobes of alternating signs, all located in the xy plane, and with positive lobes along the x and y axes.

From the above representation of the w_{lm0} functions, it is clear that the expansion of the CODF in terms of the generalized spherical harmonics treats the z axis differently from the x and y axes. Although the basis functions form a complete set, the limited number of coefficients that can be obtained by acoustic measurements makes the choice of macroscopic axes important because a better choice can reduce the error sum-of-squares, i.e., the integral of the squares of the difference between the CODF and its truncated expansion. Conventional pole figures of the various basis functions and of the CODF can be obtained using a contour plot of a projection of the above representation. Given that only the probability distribution of the crystallite c axes can be obtained, the (0002) pole figure is the only relevant one. Pole figures of w_{400} , w_{420} , and w_{440} on a xy plane projection have been published elsewhere.⁵

ULTRASONIC MEASUREMENTS OF TEXTURE

Recently, we have shown how to relate the angular dependence of the acoustic velocity of various acoustic modes to the five texture coefficients W_{200} , W_{220} , W_{400} , W_{420} , and W_{440} .¹ The effective elastic constants of the aggregate, $\overline{C}_{ij}$, are expressed as⁶

$$\overline{C}_{ij} = C_{ij}^0 + \Delta C_{ij}, \quad (4)$$

i.e., as a sum of an isotropic value,

$$C_{ij}^0 = C_{ij}^0(c_{sc}), \quad (5)$$

and a texture dependent term,

$$\Delta C_{ij} = \Delta C_{ij}(c_{sc}, W_{lmn}), \quad (6)$$

subject to the weak anisotropy condition

$$\Delta C_{ij} \ll C_{ij}^0. \quad (7)$$

In (4) and (5), c_{sc} are the single crystal elastic constants. These expressions can be found using either Reuss, Voigt, or Hill averages.⁷

Experimentally, the angular dependence of the acoustic velocity in one to three principal planes of the orthorhombic aggregate is fitted to the approximate equation

$$v(\gamma) \approx v_0 + P + Q \cos 2\gamma + R \cos 4\gamma, \quad (8)$$

where v is the acoustic velocity of the aggregate, γ is the propagation angle in the principal plane, v_0 is the isotropic velocity, and P , Q , and R are the fitted parameters. In the weak anisotropy condition (7), equation (8) is a good approximation to the solution of the Christoffel's equation,⁸ and the fitted parameters can be expressed in terms of the effective elastic constants. Therefore, we can write

$$\begin{aligned} P &= P(\bar{C}) = P(c_{sc}, W_{lmn}) \\ Q &= Q(\bar{C}) = Q(c_{sc}, W_{lmn}) \\ R &= R(\bar{C}) = R(c_{sc}, W_{lmn}) \end{aligned} \quad (9)$$

These expressions have been obtained for bulk longitudinal and shear waves, Rayleigh surface waves, and Lamb symmetric plate waves.¹

If enough acoustic modes are observed in one or several principal planes so that at least five different values of P , Q , and R are known, the W_{lmn} can be found by inverting (9).¹ (Note that if not all W_{lmn} are desired, fewer values of P , Q , or R may be required.) If more than five different values of P , Q , and R are known, equations (9) become overdetermined and a least squares fitting procedure can be used.⁹ However, it has been pointed out that the W_{lmn} evaluated using the P values (absolute velocity measurements) are much more sensitive to errors in the single crystal elastic constants and density than those evaluated using only Q and R values (relative velocity measurements).¹ On the other hand, the R values are usually smaller and more difficult to measure than the Q values. Therefore, the choice of procedure to use for the inversion of (9) is very dependent on the amount and quality of the acoustic data.

THE ACOUSTIC MICROSCOPE

The measurements presented here were made with a 225 MHz line-focus acoustic microscope.¹⁰ This method of measurement has the advantages that only small samples and relatively little machining are needed, and that the acoustic velocity of surface waves may be measured at any angle on the sample surface. However, the acoustic microscope requires careful polishing of the sample surfaces and scans of the elastic properties are made on a relatively small volume near the surface. Therefore, the technique is limited to samples with small grain sizes and it is sensitive to surface texture only. Moreover, the microscope measures leaky Rayleigh waves (LRW) and leaky surface-skimming compressional waves (LSSCW) instead of the pure Rayleigh waves and bulk longitudinal waves described in the above theory. The LRW are thought to behave essentially as pure Rayleigh waves.¹¹ We assume that the LSSCW behave as bulk longitudinal waves, but recent work indicates that this assumption may not always be valid.¹²

RESULTS

Pure Zr sample

To test the validity of the inversion of (9) and of our experimental technique, we have obtained the texture coefficient of a 99.8 % pure Zr rod (Goodfellow Corporation, Malvern,

PA, part #ZR007910) using both acoustic measurements and neutron diffraction. The neutron diffraction measurements were made at the NRU reactor at AECL Research, Chalk River Laboratories. The acoustic measurements were obtained using the LRW and the LSSCW propagating in the $x = 0$ and $y = 0$ planes (Acoustic 1) and in the $x = 0$ and $z = 0$ planes (Acoustic 2). One hundred velocities were measured in intervals of 2° . The single-crystal elastic constants used are from the literature¹³ and the density was measured. The sample grain size was of the order of $20 \mu\text{m}$ but this large grain size did not seem to affect the quality of the data. The results are shown in Table 1. The numbers in parentheses are the error estimates on the least significant digits. For the neutron diffraction measurements, the errors were deduced from the consistency of five different pole figures. For the acoustic measurements, the errors are obtained from the statistical error of the fitted values of P, Q, and R. To this statistical error, one must add an unknown systematic error resulting from the choice of averaging method. In Table 1, Hill's averaging was used. The choice of averaging method will be discussed further below.

Table 1. Comparison of pure-zirconium texture coefficients obtained using neutron diffraction and acoustic measurements. The acoustic measurements were made using LRW and LSSCW in the $x = 0$ and $y = 0$ planes (Acoustic 1) and in the $x = 0$ and $z = 0$ planes (Acoustic 2). Hill's averaging is used.

	Neutron	Acoustic 1	Acoustic 2
W_{200}	-0.0156 (4)	-0.0147 (4)	-0.0141 (7)
W_{220}	0.0002 (1)	-0.0010 (5)	0.0017 (3)
W_{400}	0.0097 (2)	0.0136 (10)	0.0111 (7)
W_{420}	-0.0001 (1)	0.0006 (5)	-0.0011 (2)
W_{440}	0.0000 (1)	0.0007 (9)	0.0018 (5)

Zr-2.5%Nb alloys

Metals are usually alloyed to tailor various physical properties. Therefore, acoustic texture measurement techniques are of limited use unless they can be applied to alloys. Some complications arising in alloys are the presence of additional phases and possible changes in the single crystal elastic constants caused by the solute. We do not attempt to model each phase separately and obtain texture coefficients for each phase but restrict ourselves to alloys where the primary phase constitutes most of the material, so that the second phase can be ignored in first approximation.

To verify the method's applicability to alloys, we measured six samples of Zr alloyed with 2.5% Nb. The samples are made of 90 to 95% hexagonal α phase and 5 to 10% cubic β phase. The α phase contains up to 1% of Nb in solution and the β phase contains from 30 to 50% Nb. The β phase is known to have a much weaker texture than the α phase.¹⁴ The grain sizes are of the order of $1 \mu\text{m}$. The samples were annealed to remove possible residual stress effects. The acoustic texture was obtained using the same α -Zr single crystal elastic constants as for pure Zr, and the measured density of the alloy. The acoustic texture coefficients of these six samples and of the pure Zr sample are plotted in Figure 2 as a function of the α phase coefficients obtained by neutron diffraction. For the acoustic measurements, Hill's averaging and the two acoustic waves LSSCW and LRW propagating in the $x = 0$ and $y = 0$ planes are used.

Figure 2. Texture coefficients as measured with ultrasound versus those obtained by neutron diffraction for one pure Zr and six Zr-2.5%Nb samples. The linear least squares fits shown include all data points.

In all cases (Fig. 2) the relation between the acoustic and neutron diffraction measurements is linear, the slope is nearly equal to one, and the y intercept is nearly zero. There occurred measurement difficulties for one sample: the measured acoustic velocities contained abnormally large errors, equation (8) was poorly satisfied, and some of the fitted values of P, Q, and R were unreliable. The measurements were repeated a second time without improvements and we do not know why this sample behaved differently. Consequently, the two points with neutron values of W_{420} near -0.002 and the two points with neutron values of W_{440} near 0.0015 are likely to be outliers. The linear relationship between neutron and acoustic measurements appears best for the W_{200} and W_{220} and, to a lesser extent, the W_{400} coefficients. Perhaps this should not be surprising because their

corresponding basis functions are the simplest ones and have the largest amplitude (see Figure 1). Another reason for the better linearity of W_{200} and W_{220} might be that, because of symmetry, these coefficients must be equal to zero for orthorhombic aggregates of cubic materials² and consequently, the β phase is not expected to contribute.

These results show that the acoustic behavior of the samples is dominated by the texture of the primary α phase. This is not surprising because the effective elastic constants, in the Reuss or Voigt approximations, are volume averages of the individual phases and because the primary α phase represents 90 to 95 % of the material. On the other hand, the elastic constants and density of the β (cubic) crystallites can be substantially different from those of the α (hexagonal) crystallites¹⁵ but because it is weakly textured, the β phase should affect mainly the isotropic component of the sample's effective elastic constants. Only relative velocity measurements were used in the texture measurement; therefore, the β phase is not expected to affect the results significantly. The results also show that the 1% Nb dissolved in the α phase does not affect the elastic properties of the crystallites sufficiently to prevent texture measurements.

Voigt, Reuss and Hill averaging

The influence of averaging method on the texture coefficients obtained using ultrasonic methods is best illustrated by Figure 3. The figure is similar to Figure 2 for the Hill average of W_{200} but, for each acoustic measurement, the Voigt and Reuss values are indicated. In all three cases, the linear relationship is recovered but the three slopes differ somewhat: the slope is equal to 0.97 for Voigt averaging, 1.16 for Hill averaging (Figure 2), and 1.43 for Reuss averaging. The three lines meet in a point at a W_{200} acoustic value near zero and a neutron diffraction value near -0.0025. Therefore, the choice of averaging method introduces a systematic relative error as large as $\pm 23\%$ in the texture coefficient. The systematic absolute error is unexplained but may be due to measurement uncertainties or to the various approximations made: single phase material and LRW and LSSCW instead of pure Rayleigh and bulk longitudinal waves.

Figure 3. W_{200} as measured with ultrasound for Voigt (circles) and Reuss (squares) averaging methods versus W_{200} obtained by neutron diffraction for one pure Zr and six Zr-2.5%Nb samples.

CONCLUSION

We have used an acoustic microscope to measure the LRW and LSSCW velocities as a function of angle in two principal planes of one pure Zr and six Zr-2.5% Nb samples. The texture measurements of the pure Zr sample agree reasonably well with the neutron diffraction measurement. Discrepancies are related to the statistical error of the fitted P, Q, and R parameters, to the choice of averaging method, and to the use of two types of waves, LRW and LSSCW, that are only an approximation to the pure Rayleigh and bulk longitudinal acoustic modes. A linear relationship was found between the texture coefficients obtained by ultrasonics and those obtained by neutron diffraction for one pure Zr and six Zr-2.5%Nb samples. The presence of a 5 to 10% of a secondary phase and of up to 1% Nb in solution in the primary phase does not invalidate the measurement. This is an important result for the future use of acoustic methods to evaluate the texture of alloys. Our results also show systematic relative errors caused by the choice of averaging method.

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