JOURNAL OF MATERIALS SCIENCE **35** (2000) 1195–1200

Determination of anisotropic elastic moduli of Zr-2.5Nb CANDU pressure tube materials

Y.-M. CHEONG*, S.-C. KWON, H.-K. JUNG
CANDU Pressure Tube Materials Group, Korea Atomic Energy Research Institute, P.O. Box 105, Yusong, Taejon 305-600, Korea
E-mail: ymcheong@nanum.kaeri.re.kr

In order to determine the elastic stiffness of Zr-2.5Nb CANDU pressure tube materials, the resonance frequencies of a rectangular parallelepiped specimen were measured and compared with the calculated resonance frequencies based on the input data of estimated stiffness of a polycrystalline Zr-2.5Nb specimen, dimensions and density. The estimated elastic stiffness was determined using its orientation distribution function measured by x-ray diffraction and the reported elastic stiffness of a zirconium single crystal. Through comparison of calculated frequencies with that measured by resonance ultrasound spectroscopy, accurate elastic stiffness has been determined by iteration and convergence processes.

1. Introduction

The elastic moduli of materials are important in engineering, such as mechanical design, fracture analysis, life-time estimation of structures. To determine the elastic moduli it requires lots of samples with different compositions, fabrication processes and heat treatments. For anisotropic materials, such as composite materials or textured materials, it is more difficult to determine the elastic moduli, because of its anisotropic properties.

The resonant ultrasound spectroscopy (RUS) is used to determine the elastic stiffness for various shapes of samples, i.e. spherical, cylindrical, or rectangular parallelepiped. Theoretically maximum 21 tensor elements of elastic stiffness for triclinic crystal (the lowest-symmetry crystal) can be determined with one specimen. However, for such a low-symmetry crystal, it is difficult to assimilate properties relating to stress waves and elasticity [1]. Practically RUS can determine 9 tensor elements for orthorhombic symmetry as well as higher-symmetry, such as isotropic, cubic, hexagonal, tetragonal symmetry.

One of the key elements in RUS is to determine the symmetry and the initial estimate of elastic stiffness in advance. The initial estimate should be close to the true value and can be obtained from literatures, experience, other measurements, etc. The test sample should be machined accurately. The calculated resonance frequencies and modes should be matched to the measured values by RUS and the elastic stiffness can be converged by comparison and iteration.

Zr-2.5Nb alloy for the pressure tubes in CANDU (CANadian Deuterium Uranium) reactors have developed a strong texture due to the limited slip system during extrusion process, leading to anisotropic properties. The material properties strongly depend on the orientation distributions of grains, which result in a directional anisotropy of elastic stiffness, thermal expansion coefficients, etc. To characterize the degree of anisotropy, it is necessary to correctly determine the anisotropic elastic moduli depending on the direction of the tube samples. The anisotropy of the Zr-2.5Nb alloy could be treated as orthorhombic symmetry, consist of three principal coordinates such as radial, transverse, and longitudinal direction.

Initial approximated elastic stiffness has been estimated by orientation distribution function (ODF) from the x-ray pole figure data and the elastic stiffness of single crystal zirconium. Based on the initial estimates, anisotropic elastic stiffness of the Zr-2.5Nb alloy has been determined by RUS. Present work is concerned with previous measurements by other methods.

2. ODF and anisotropic elastic stiffness

Polycrystalline Zr-2.5Nb pressure tube materials is a hexagonal closed packed (hcp) structure and is textured along the circumferential direction. The elastic stiffness on the tubular sample coordinate (axial, radial, and circumferential directions) could be expressed as an orthorhombic symmetry and represented as:

$$c_{ij} = \begin{pmatrix} c_{11} & c_{12} & c_{13} & 0 & 0 & 0 \\ c_{12} & c_{22} & c_{23} & 0 & 0 & 0 \\ c_{13} & c_{23} & c_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & c_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & c_{55} & 0 \\ 0 & 0 & 0 & 0 & 0 & c_{66} \end{pmatrix} \quad (1)$$

* Author to whom all correspondence should be addressed.

Macroscopic properties of a sample can be obtained by the integration of the properties, $E_{(g)}$ depending upon the orientation of an individual grain, g and weighting factor, i.e. ODF of grains, $f_{(g)}$ along the total orientation space [2]:

$$\bar{E} = \int E(g) \times f(g)\,\mathrm{d}g \tag{2}$$

In order to represent the elastic stiffness tensor of rank = 4 to 6×6 matrix form, the usual four-to-two contraction scheme is adopted, such as $11 \rightarrow 1$, $22 \rightarrow 2$, $33 \rightarrow 3$, $23 \rightarrow 4$, $13 \rightarrow 5$, and $12 \rightarrow 6$.

The anisotropy of the polycrystalline with textured structure can be calculated as an average of material properties of each grain, if interactions between grains are negligible. The averaged elastic stiffness along the sample coordinate can be estimated by knowing elastic stiffness of single crystal and orientation distribution of grains from the pole figure data by x-ray or neutron diffraction. Based on the pole figure data, the ODF can be calculated by generalized spherical harmonics and series expansion coefficients [3]:

$$\omega(\xi, \phi, \varphi) = \sum_{l=0}^{4} \sum_{m=l}^{4} \sum_{n=-l}^{4} W_{lmn} Z_{lmn}\, e^{-im\phi} e^{-in\varphi}, \tag{3}$$

where Z_{lmn} is the generalized Legendre polynomial defined by Roe [4], W_{lmn} is the series expansion coefficient adopted from by Roe, or notation by Bunge [5]. When a tensor rank with p is averaged, W_{lmn} includes the elements of $l \leq p$, which implies a maximum number tensor element, l, m, and n is 4 for the case of elastic stiffness tensor. W_{lmn} and Z_{lmn} for orthorhombic symmetry have been calculated by Morris [6].

In order to explain actual elastic stiffness, various models or approximations have been suggested. Based on the orientation distribution by a sample coordinate, the elastic stiffness can be estimated by either Voigt's approximation, Reuss approximation, or self-consistent iteration method, etc. Voigt's approximation assumes that the total stress is a sum of the individual stresses on each grain, whereas in the Reuss approximation a sum of the individual strains on each grain is assumed. Both approximations represent two extreme cases and can be regarded as upper and lower bounds [7]:

$$\overline{c_{ijkl}} = \frac{1}{8\pi^2} \int c_{ijkl}(\Omega)\,\mathrm{d}\Omega \qquad \text{(Voigt's approximation)}, \tag{4}$$

$$\overline{S_{ijkl}} = \frac{1}{8\pi^2} \int S_{ijkl}(\Omega)\,\mathrm{d}\Omega \qquad \text{(Reuss approximation)}, \tag{5}$$

where $\overline{c_{ijkl}}$ and c_{ijkl} are the averaged and individual elastic stiffness, $\overline{S_{ijkl}}$ and S_{ijkl} are the averaged and individual elastic compliance, Ω is Euler angle, θ, ϕ, φ and $\int \mathrm{d}\Omega = 8\pi^2$.

3. Elastic stiffness by resonant ultrasound spectroscopy (RUS)

Free vibration or resonance is sensitive to the microscopic and macroscopic properties of the materials. Because RUS can determine accurate elastic stiffness and ultrasonic attenuation, it can be applied to materials characterization, non-destructive testing, etc. [8] Exact analytical solution on the free vibration problem to determine resonance frequencies is not known or available *a priori*. Only approximated solutions are available by numerical analysis, such as the finite element method or minimization of energy. The fundamental theory of resonance was developed by Maynard [9], and theoretical calculations and experiments for the resonance of an elastically isotropic rectangular parallelepiped specimen have been made by Holland [10] and Demarest [11]. Those results have been generalized by Ohno [12] and comprehensive applications to the solid-state physic have been accomplished by Migliori [13].

The eigenvector or eigenfrequency of vibrating solids can be calculated by a theory of the minimization of energy, i.e. a mechanical Lagrangian of elastic solids with some approximations. In classical mechanics, the solution of Lagrangian for free vibration can be expressed as the elastic wave equation,

$$\rho\omega^2 u_i + \sum_{j,k,l} c_{ijkl} \frac{\partial^2 u_k}{\partial x_j \partial x_l} = 0. \tag{6}$$

This equation is solved subject to the vanishing of the i-th component of the surface traction vector,

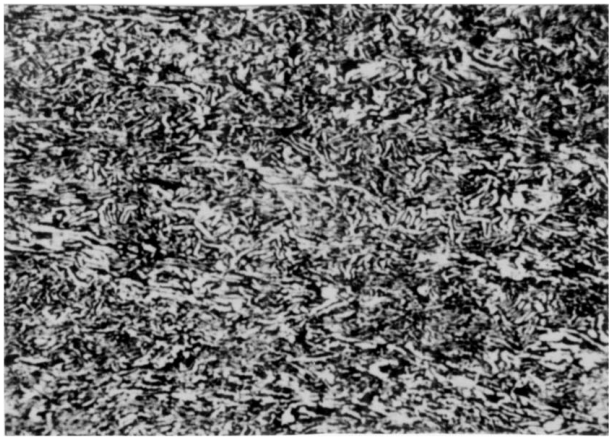
$$\sum_{j,k,l} \vec{n}_j c_{ijkl} = \sum_j \vec{n}_j \sigma_{ij} = 0, \tag{7}$$

where ρ is density, ω is frequency, u_i is the i-th component of the displacement vector, c_{ijkl} is elastic modulus tensor, n_j is unit vector normal to the surface, and σ_{ij} is stress tensor. While the direct (forward) problem (calculation of resonance frequencies based on sample description) is challenging in its own right, the inverse problem (calculation of elastic constants from measured frequencies) is considerably more difficult. A combination of u_i satisfying those conditions is displacement corresponding to the normal mode free vibration frequency. Based on this fact, an algorithm for the calculation of frequencies corresponding to the minimization of energy using Rayleigh-Ritz method have been developed by Demarest [11]. The resonance frequencies can be calculated for the sample with known density, dimension, orientation, and elastic stiffness. Actually, because the solution of the inverse problem is not simple and no exact solution is available, nonlinear optimization procedures are the best option. A computer code has been developed based on a fast and efficient solution of the direct problem, which is then used in an iterative Levenburg-Marquardt scheme to solve the inverse problem with the figure of merit [14].

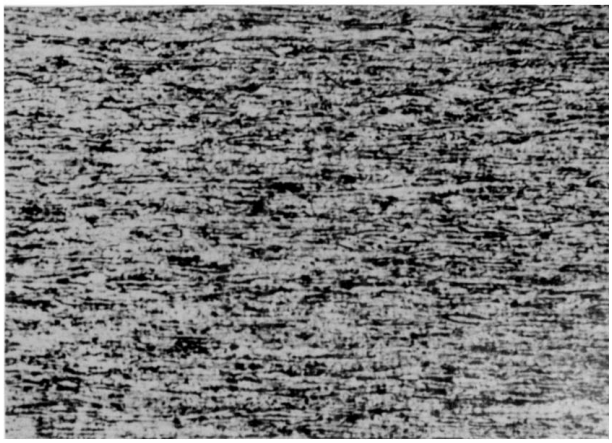
4. Experimental

4.1. Specimen

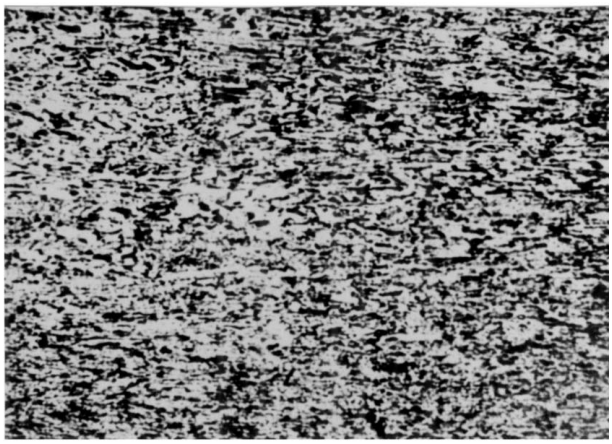
Nuclear grade Zr-2.5Nb CANDU pressure tube materials are used as specimen. As shown in Fig. 1, the grains are elongated along the circumferential direction. Fig. 2



a) TN



b) LN



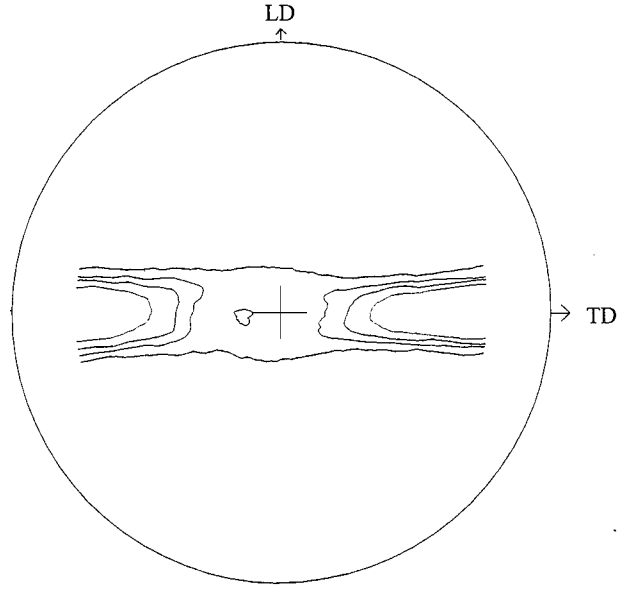
c) RN

Figure 1 Optical microstructures of CANDU pressure tube at a) the L section normal to a tangential direction, b) the T section normal to a longitudinal direction, and c) the R section normal to a radial direction.

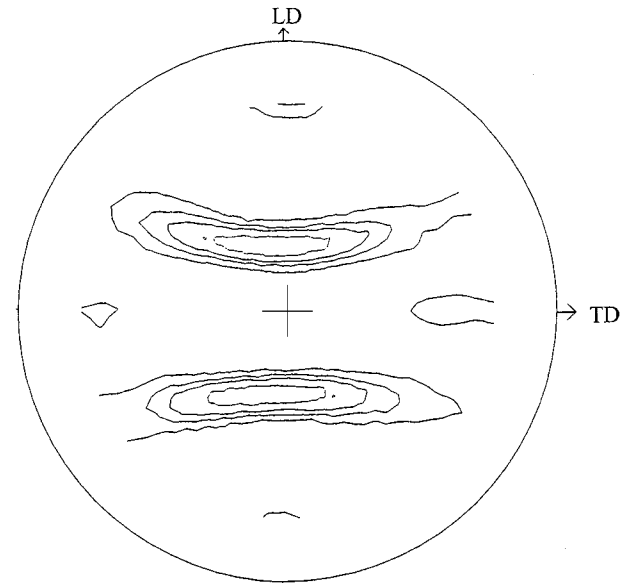
shows the pole figures of (00.2), (10.0), and (10.1) for the Zr-2.5Nb alloy. This material exhibits strong textured structure due to extrusion processes.

4.2. Calculation of the initial estimate of anisotropic elastic stiffness

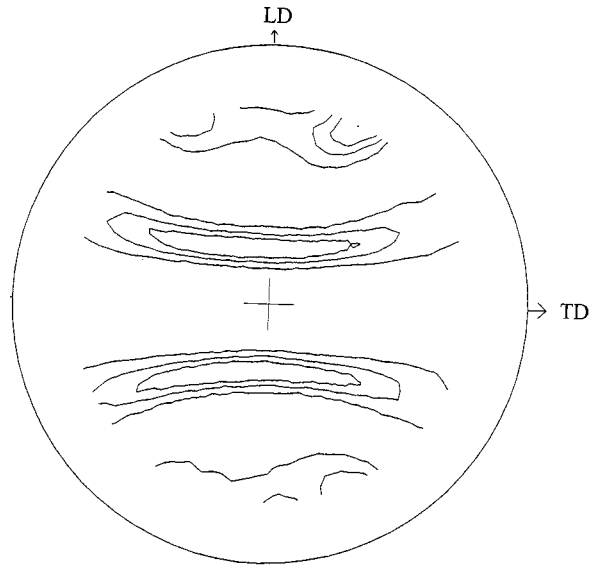
The ODF of the Zr-2.5Nb alloy has been calculated using a computer program, "popLA", by the Los Alamos National Laboratory [15]. The raw data file of x-ray



a) Pole figure : 00.2



b) Pole figure : 10.0



c) Pole figure : 10.1

Figure 2 Pole figures for Zr-2.5Nb pressure tube materials.

TABLE I Elastic stiffness of single crystal Zirconium (hcp)

$c_{11}=c_{22}$	c_{33}	c_{44}	$c_{66}=\frac{1}{2}(c_{11}-c_{12})$	c_{13}	c_{12}
1.434	1.648	0.320	0.353	0.653	0.728

Notation of hcp single crystal: 1(=2) = a-axis, 3 = c-axis.
unit: 10^{11} N/m^2.

TABLE II Anisotropic elastic stiffness of Zr-2.5Nb pressure tube materials by orientation distribution function (ODF)

	c_{11}	c_{22}	c_{33}	c_{23}	c_{13}	c_{12}	c_{44}	c_{55}	c_{66}
Voigt's approx.	1.449	1.490	1.446	0.687	0.699	0.713	0.340	0.343	0.375
Reuss approx.	1.437	1.473	1.440	0.691	0.702	0.720	0.338	0.341	0.365
Self-consistent	1.443	1.482	1.443	0.689	0.700	0.717	0.339	0.342	0.370

Notation of sample orientation: 1 = RD (Radial Direction), 2 = TD (Transverse Direction), 3 = LD (Longitudinal Direction).
unit: 10^{11} N/m^2.

pole figure was converted to the ASCII format as required by the program. The angles of orientation distribution are represented by one of the notations by Euler, Roe-Matties, or Bunge [15].

With the elastic stiffness of single crystal zirconium [16], shown in Table I, and the weight factor of individual grain obtained from the ODF, averaged elastic stiffness of the polycrystalline Zr-2.5Nb alloy have been obtained using Voigt's approximation, Reuss approximation, or self-consistent method by iteration. The subscripts in Table I are referred to the crystallite coordinate, i.e. 1(=2) = a-axis, 3 = c-axis in the hcp single crystal orientation.

Initial estimates of the elastic stiffness of the polycrystalline Zr-2.5Nb alloy are shown in Table II. The subscripts are referred to the tubular sample coordinate, i.e. 1 = radial, 2 = transverse, and 3 = longitudinal direction. The anisotropic nature along axial, radial, and circumferential directions can make it as the orthorhombic symmetry, which requires 9 independent elastic stiffness.

4.3. Determination of anisotropic elastic stiffness by RUS

Rectangular parallelepiped Zr-2.5Nb samples were machined accurately, with dimensions in a range of 2.5–5.5 mm. The sample is inserted between two ultrasonic transducers, one is transmitter and the other is a receiver, and minimal force was applied in order to hold the specimen at the corners as allow free vibration of the specimen (see Fig. 3). Because Voigt's approximation and Reuss approximation represent two extreme cases, the elastic stiffness by self-consistent method by iteration was adopted for the initial estimate of elastic stiffness for the RUS. Calculated frequencies by an input of dimensions, density, symmetry, and initial estimate of elastic stiffness were corresponded to the measured frequencies and vibration modes. Accurate values of elastic stiffness have been obtained by comparison and iteration algorithm.

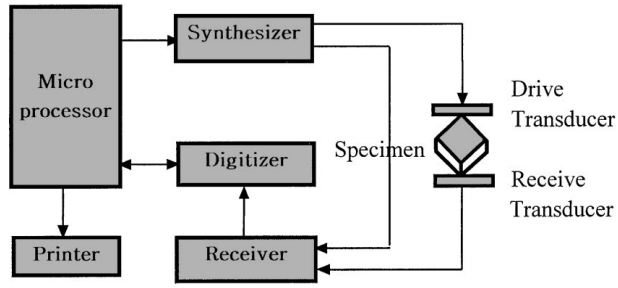


Figure 3 Block diagram of resonant ultrasound spectroscopy (RUS).

Fig. 4 shows a typical resonance ultrasound spectrum of the Zr-2.5Nb alloy. For the case of a specimen dimensions of 3.582 mm×3.486 mm×5.318 mm, initial 30 resonance frequencies are in the range of 180–600 kHz. Generally if the RMS error is less than 0.2%, the results could be regarded as reliable and accurate.

5. Results and discussion

Table III shows anisotropic elastic stiffness of the Zr-2.5Nb alloy by RUS. Table IV shows the Young's moduli, which are the inverse of the elastic compliance, $E_{ii}=1/S_{ii}$.

Based on the pole figures for (0002) in Fig. 2, f-coefficients or Kearn's factors, which represent the degree of orientations along the c-axis, are calculated as $f_{\mathrm{T}}=0.6$, $f_{\mathrm{R}}=0.33$, $f_{\mathrm{L}}=0.07$, where the subscripts denote T = transverse, R = radial, and L = longitudinal direction [17, 18]. This fact implies that approximately 60% of (0002) pole are aligned along the transverse direction (or circumferential direction), 33% along the radial direction, and 7% along the axial direction. Because the elastic stiffness along the c-axis in the single crystal zirconium is greater than along the a-axis, i.e. $c_{33} > c_{11}(=c_{22})$ (subscripts follows single crystal coordinate; 1(=2) = a-axis, 3 = c-axis in hcp crystalline structure), the elastic stiffness along a direction of higher f-coefficient should be greater than the other directions. Either, the elastic stiffness estimated by ODF

TABLE III Anisotropic elastic stiffness of Zr-2.5Nb pressure tube materials by resonant ultrasound spectroscopy (RUS)

c_{11}	c_{22}	c_{33}	c_{23}	c_{13}	c_{12}	c_{44}	c_{55}	c_{66}
1.4708	1.5269	1.4533	0.7191	0.7538	0.7446	0.3381	0.3425	0.3696

Notation of sample orientation: 1 = RD (Radial Direction), 2 = TD (Transverse Direction), 3 = LD (Longitudinal Direction).
unit: 10^{11} N/m^2.

TABLE IV Anisotropic Elastic Moduli of Zr-2.5Nb pressure tube materials converted from the elastic compliance

Young's moduli			Shear moduli			Bulk modulus
E_{11}	E_{22}	E_{33}	G_{44}	G_{55}	G_{66}	K
96.18	104.32	96.79	33.81	34.59	36.96	98.70

Notation of sample orientation: 1 = RD (Radial Direction), 2 = TD (Transverse Direction), 3 = LD (Longitudinal Direction).
unit: 10^{11} N/m^2.

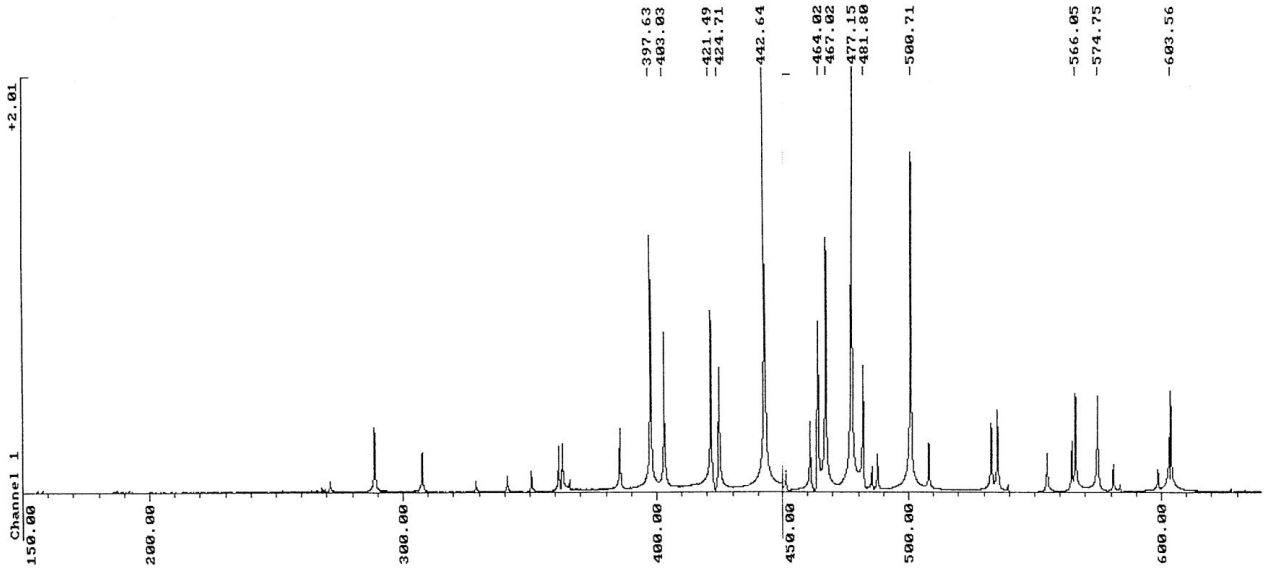


Figure 4 Typical resonance spectrum of rectangular parallelepiped Zr-2.5Nb pressure tube materials.

or RUS results in $c_{22} > c_{11} > c_{33}$ (subscripts follows sample coordinate; 1 = radial direction, 2 = transverse direction, 3 = longitudinal direction), which indicates the highest elastic stiffness is along the transverse direction and the lowest along the longitudinal direction, which is in accord with the expectation.

There are little differences between the elastic stiffness by ODF and by RUS, shown in Tables II and III. The elastic stiffness estimated by ODF is based on the orientation distribution of an individual crystallite. All models for macroscopic (specimen) properties from microscopic (crystallite) properties, such as Voight's approximation, or Reuss approximation share common assumptions: i) absence of voids, non-homogeneity, ii) cohesion of crystallites occur through very thin grain boundary regions that are deformed relative to the crystal interiors, iii) randomly orientated grains, and iv) grains large enough so that interfaces remain non-important [7]. In addition, microscopic variations, such as size and shape of crystallites, the effect of alloying elements, the existence of β-phase, dislocation density and distribution, are not reflected in the estimation by ODF. Therefore the elastic stiffness determined by RUS with initial estimates by ODF could be closer to the true value.

One of the important factors in determining elastic stiffness by RUS is the initial input of the elastic stiffness be close enough to the true values in order to converge during iteration processes. In an early study, we attempted the averaging method by f-coefficients from x-ray pole figure data. Using those values as the initial input for RUS calculations, relatively high RMS errors between calculated and measured frequencies indicated the RUS measurement was not reliable. It seems that f-coefficients, which implies simple fractions of crystallites along the 3 major axes, could not be a proper approximation to obtain 9 independent elastic stiffness for the samples. However, using the initial estimate by ODF, RMS error less than 0.2% indicates the method is reliable and accurate.

It is not easy to get elastic modulus along radial or transverse direction in the tubular shaped sample by conventional methods. It has been reported that the approximated elastic modulus for α-zirconium is 98.6 GPa [19]. Ashida [20] reported that a relation of Young's modulus to temperature as $E = -0.0656T + 115.1$ by measurement of resonance frequencies in the bending and torsion test, which comes out the Young's modulus of 95.4 GPa at room temperature. Northwood *et al*. [21] reported that Young's moduli of Zr-2.5Nb alloy are $E_{\mathrm{L}} = 97.0$ GPa, $E_{\mathrm{T}} = 95.2$ GPa, $G = 35.9$ GPa. All these reported values are restricted to a certain direction or assumption of isotropic properties. However, as shown in Table IV, RUS can determine all the elements of elastic stiffness, including Young's moduli along the radial, transverse, and radial directions as well as shear moduli and bulk modulus at once. Because of the differences in materials, especially in anisotropic properties, small differences of the elastic moduli are quite natural.

6. Summary

1. Anisotropic elastic stiffness of Zr-2.5Nb pressure tube materials has been determined by RUS. The initial estimate for RUS has been obtained from consideration of ODF by x-ray pole figures and elastic stiffness of single crystal zirconium.

2. RMS error less than 0.2% in RUS indicates that the measurement is reliable and accurate.

3. There is a little difference between the elastic stiffness by ODF and RUS. It implies that the values by RUS are closer to the true values.

Acknowledgements

This work was supported by the CANDU pressure tube materials project, as a part of long-term nuclear research program by the Ministry of Science and Technology. The authors would like to express their appreciation to

Dr. Yong-Che Kim for sincere discussions on the theory of ODF and analysis of x-ray pole figures.

References

1. L. D. LANDAU and E. M. LIFSHITZ, in "Theory of Elasticity," 2nd ed. (Pergamon Press, London, 1970) p. 37.
2. Y. C. KIM *et al*., *J. Korean Inst. Met. & Mater* **29** (1991) 866.
3. M. L. DUNN, H. LEDBETTER, P. R. HEYLIGER and C. S. CHOI, *J. Mech. Phys Solids* **44** (1996) 1509.
4. R. J. ROE, *J. Appl. Phys.* **36** (1965) 2024.
5. H. J. BUNGE, in "Texture Analysis on Materials Science," translated by P. R. Morris (Butterworth & Co., 1982) p. 97.
6. P. R. MORRIS, *J. Appl. Phys.* **40** (1969) 447.
7. H. LEDBETTER, *ASTM STP* **1045** (1990) 135.
8. A. MIGLIOLI *et al. Phys. Rev.* **41** (1990) 2098.
9. J. D. MAYNARD, *J. Acoust. Soc. Am.* **91** (1992) 1754.
10. R. HOLLAND, *ibid.* **43** (1968) 988.
11. H. H. DEMAREST, *ibid.* **49** (1971) 768.
12. I. OHNO, *J. Phys. Earth* **24** (1976) 355.
13. A. MIGLIORI *et al., Physica B* **183** (1993) 1.
14. A. MIGLIORI and J. SARRAO, in "Resonant Ultrasound Spectroscopy," (John Wiley & Sons, Inc., 1997) p. 53.
15. S. I. WRIGHT and U. F. KOCKS in "popLA, Preferred Orientation Package-Los Alamos," (Los Alamos National Laboratory, 1995).
16. E. S. FISHER and C. J. RENKEN, *Phys. Rev.* **135**(2A), (1964) A482.
17. Y. S. KIM *et al.*, Development of zirconium alloys for pressure tubes, Final Report, KAERI/RR-1766-96, Korea Atomic Energy Research Institute, Korea, 1997.
18. K. S. CHOO, S. C. KWON and Y. S. KIM, *J. Korean Nucl. Soc.* **30**(4) (1998) 318.
19. P. K. DE *et al., J. Nucl. Mater.* **252** (1998) 43.
20. Y. ASHIDA *et al., J. Appl. Phys.* **80** (1996) 3254.
21. D. O. NORTHWOOD, I. M. LONDON and L. E. BAHEN, *J. Nucl. Mater.* **55** (1975) 299.

Received 28 January
and accepted 11 August 1999