A Feasibility Study on Determination of Hydrogen Concentration in Zirconium Alloy by Resonant Ultrasound Spectroscopy

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1. Introduction

Resonant ultrasound spectroscopy (RUS) is used to determine the elastic stiffness for various shapes of samples, i.e. spherical, cylindrical, or rectangular parallelepiped. Theoretically a maximum of 21 tensor elements of elastic stiffness for a 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. Practically, RUS can determine 9 tensor elements for orthorhombic symmetry as well as higher-symmetry, such as isotropic, cubic, hexagonal, and 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 the literature, 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.

The 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 the 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 anisotropic elastic constant of the Zr-2.5Nb alloy was determined using initial approximated elastic stiffness which had been estimated by the orientation distribution function (ODF) from x-ray pole figure data and the elastic stiffness of single crystal zirconium.

In this paper, the temperature dependence of mechanical damping and resonance frequencies of the Zr-2.5Nb materials were determined by high temperature RUS.

2. High Temperature RUS

Nuclear grade Zr-2.5Nb CANDU pressure tube materials are used as the specimen. This material exhibits a strong textured structure due to extrusion and cold rolling processes.

The high temperature RUS device was fabricated as shown in Fig. 1. A small furnace and a temperature controller were attached to the basic RUS device, which consists of a synthesizer to generate continuous frequencies and two wideband ultrasonic transducers for sending and receiving signals in a vacuum system. The specimen is inserted between two wave guides with ultrasonic transducers, one is a transmitter and the other is a receiver. Minimal force was applied in order to hold the specimen at the corners to allow free vibration of the specimen, and controlled accurately (< 1 g) by a load cell, spring device, and a positioning device.

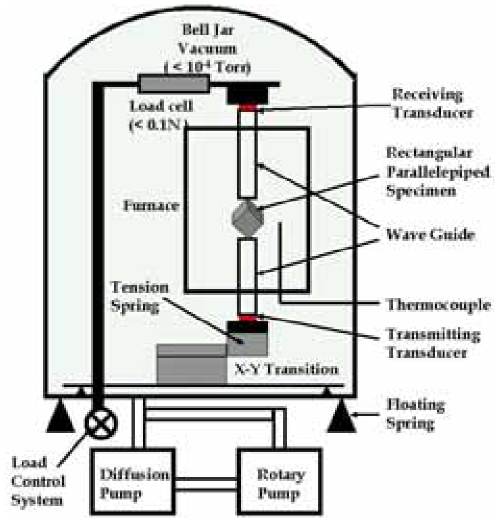


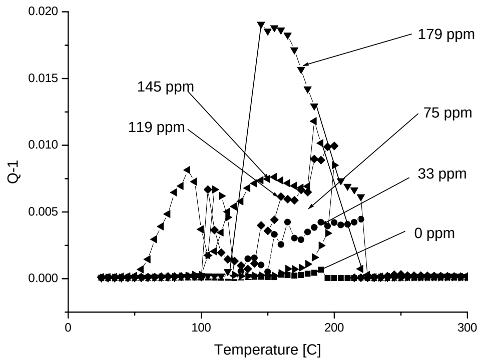
Figure 1 High temperature device for RUS experiment.

Rectangular parallelepiped Zr-2.5Nb samples were machined accurately, with dimensions of 2.5 mm x 3.0 mm x 3.5 mm. Calculated frequencies by an input of dimensions, density, symmetry, and initial estimate of elastic stiffness corresponded to the measured frequencies and vibration modes. Accurate values of elastic stiffness were obtained by comparison and iteration algorithm. The initial 30 resonant frequencies were calculated and compared with the measured values. After iteration and convergence, the final RMS errors were in the range of

0.05 ~ 0.1 %, which can be compared with the RMS error less than 0.2% can be regarded as reliable and accurate.

2.1 Temperature Dependence of Mechanical Damping

Fig. 2 shows higher Q^{-1} in the temperature range of 100~220°C. Those Q^{-1} values increase as the hydrogen concentrations increase. There is no clear explanation of these high Q^{-1} values at this moment, but there are several possibilities of; 1) a transformation of δ-zirconium hydride to γ-zirconium hydrides or vice versa, 2) impurity effect of hydrogen atoms, or so called as 'high temperature Bordoni peak'.



1.

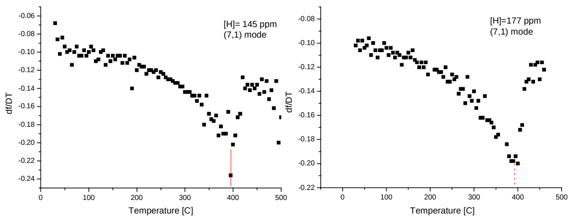
2.

Figure 2. Temperature dependency of mechanical damping of Zr-2.5Nb pressure tube material.

3.

2.2 Temperature Dependence of resonance frequency to temperature

Fig. 3 shows an example of the deviation of resonance frequency to temperature (*dfr/dT*). It can be seen that a minima at a certain temperature and this fact could be corresponded to the terminal solid solubility for dissolution(TSSD) of hydrogen, shown in Fig. 4.



(a) [H] = 145 ppm **(b) [H] = 177 ppm**

Figure 3. Temperature effect of ***dfr/dT*** for determination of hydrogen concentration in zirconium alloy.

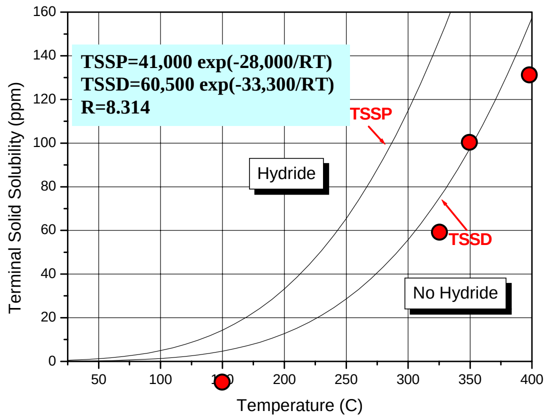


Figure 4. Determination of the hydrogen concentration in zirconium alloy by using the TSSD curve of Zr-2.5Nb alloy.

3. Conclusion

1. The dynamic anisotropic elastic stiffness and mechanical damping Q^{-1} of Zr-2.5Nb pressure tube materials has been determined by resonant ultrasound spectroscopy (RUS). The initial estimate for RUS has been obtained from a consideration of ODF by x-ray pole figures and elastic stiffness of a single crystal zirconium.
2. Higher Q^{-1} in the temperature range of 100~220°C. Those Q^{-1} values increase as the hydrogen concentrations increase.
3. The deviation of resonance frequency to temperature (*dfr/dT*) shows a minima at a certain temperature and this fact could be corresponded to the terminal solid solubility for dissolution (TSSD) of hydrogen.

Acknowledgements

This work was supported by the CANDU pressure tube materials project, as a part of the long-term nuclear research program by the Ministry of Science and Technology.