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Non-Destructive Detection of Hydride Blister in PHWR Pressure Tube Using an Ultrasonic Velocity Ratio Method

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Abstract

Since Zr-2.5Nb pressure tubes have a high risk for the formation of blisters during their operation in pressurized heavy water reactors, there has been a strong incentive to develop a method for the non-destructive detection of blisters grown on the tube surfaces. However, because there is little mismatch in acoustic impedance between the hydride blisters and zirconium matrix, it is not easy to distinguish the boundary between the blister and zirconium matrix with conventional ultrasonic methods. This study has focused on the development of a special ultrasonic method, so called ultrasonic velocity ratio method for a reliable detection of blisters formed on Zr-2.5Nb pressure tubes. Hydride blisters were grown on the outer surface of the Zr-2.5Nb pressure tube using a cold finger attached to a steady state thermal diffusion equipment. To maximize a difference in the ultrasonic velocity in hydride blisters and the zirconium matrix, the ultrasonic velocity ratio of longitudinal wave to shear wave, V_L/V_S, has been determined based on the flight time of the longitudinal echo and reflected shear echo from the outer surface of the tubes. The feasibility of the ultrasonic velocity ratio method is confirmed by comparing the contour plots reproduced by this method with those of the blisters grown on the Zr-2.5Nb pressure tubes.

Key Words : hydride blister, ultrasonic testing, CANDU pressure tube

1. Introduction

The nuclear fuel channel of Pressurized Heavy Water Reactors comprises horizontal Zr-2.5Nb pressure tube carrying fuel bundles and hot coolant, and a concentric Zircaloy-2 calandria tube. Loose fitted garter springs were provided to prevent a hot pressure tube coming in contact with a cold calandria tube. However, garter springs in some cases shift from their original locations during reactor operation, causing contacts between pressure tubes and calandria tubes. In the case of contact between pressure tube and calandria tube the hydrogen/deuterium migrates to the contact region due to the temperature gradient between colder contact region and hotter pressure tube. When the concentration of hydrogen at the contacts exceeds

Table 1. Acoustic Properties of Zirconium and Zirconium Hydride

Material	Density (10^3kg/m^3)	L-wave Velocity(m/s)	S-wave Velocity(m/s)	Acoustic Impedance L-wave (z=ρc[MRayl])	Acoustic Impedance S-wave (z=ρc[MRayl])	Velocity ratio (V_L/V_S)
α-Zr-2.5Nb	6.48	4780	2380	31.0	15.4	2.01
δ-$ZrH_{1.66}$	5.6	5860	1900	32.8	10.6	3.08

the terminal solid solubility for hydrogen, the zirconium hydride platelets precipitate at the contact areas. Over a length of time, the localized massive hydrides termed as "blister" are produced because of volume expansion.

Hydride blister is one of the key issues to the safety of a PHWR because it accompanies a volume expansion and sometimes cracking on the outer surface of the pressure tubes. The failure of a pressure tube in Pickering Unit 2 reactor in 1983 was attributed to the formation of hydride blisters [1]. Comprehensive research on the mechanism of formation and growth kinetics of a hydride blister, such as hydrogen diffusion with temperature gradient, blister formation threshold, distribution of residual stress is being made [2-8].

Since there is a high possibility of cracking on the surface of a blister and the blister itself is treated as a defect, it should be detected by the non-destructive test methods before it grows to a critical size and cracking. Effort is being made to detect the hydride blister non-destructively [9,10]. However, because the depth of a blister is relatively small (~several tens to hundreds micrometer) and there is little difference in the acoustic impedance between α-zirconium and δ-hydride (Table 1), it is difficult to detect the blisters with conventional ultrasonic methods in principle. We introduced an ultrasonic velocity ratio method (V_L/V_S) to effectively detect the hydride blister in Zr-2.5Nb pressure tubes. The ultrasonic velocity ratio method was applied to the tube specimen with a blister of different size and confirmed a superior capability of detection and reconstruction of the blister to the conventional ultrasonic time-of-flight method.

2. Ultrasonic Velocity Ratio Method

When an ultrasonic longitudinal wave is incident on a solid interface, both the longitudinal wave and shear wave are reflected and refracted to the back wall in accordance with mode conversion [11]. As the longitudinal wave velocity in δ-hydride is greater than in α-zirconium shown in Table 1, the reflected wave at the back surface arrives earlier at the blister location. On the other hand, as the shear wave velocity in δ-hydride is less than in α-zirconium, the reflected wave at the back surface arrives later at the blister location. Because the fraction of a blister in the beam path is expected no larger than 10% of the total beam path (thickness of zirconium pressure tube = 4.2 mm), a difference in time-of-flight at the blister location is estimated as a maximum of around 3.0%. In other words, the time difference is too small to distinguish the blister region from the zirconium matrix with conventional longitudinal wave or shear wave.

Based on the characteristics of ultrasonic velocity in δ-hydride and a zirconium matrix, we introduce the parameter of velocity ratio of longitudinal wave to the shear wave (V_L/V_S) that will maximize the difference between the δ-hydride

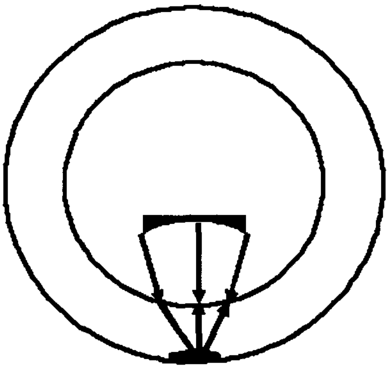

Fig. 1. Schematic Design of Ultrasonic Velocity Ratio Measurement for Blister Detection

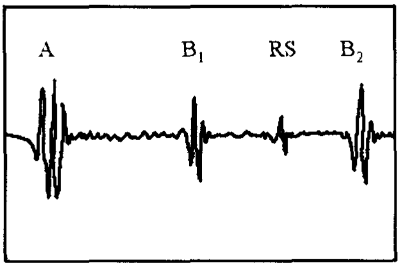

Fig. 2. Echo Pattern of Ultrasonic Velocity Ratio Method in Time Domain

and the α-zirconium matrix. A focused immersion ultrasonic transducer is used for detection of blisters shown in Fig. 1. A typical display of signals obtained from a Zr-2.5Nb pressure tube is shown in Fig. 2. The echo signal "A", "B_1", "B_2" and "RS" denote echo signals from the interface between water and surface, the first reflected echo signal at the back surface with longitudinal mode, double reflected echo signal at the back surface with longitudinal mode, and the first echo signal but converted from longitudinal mode to shear mode at the back surface, respectively.

$$B_1 = \frac{2t}{V_L}, \quad B_2 = \frac{4t}{V_L}, \quad RS = \frac{t}{V_L} + \frac{t}{V_S} \tag{1}$$

, where B_1, B_2 and RS denote the time-of-flight of first back reflected longitudinal wave, second back reflected longitudinal wave, and the reflected shear wave (the converted signal from longitudinal to shear modes at the back surface), and t, V_L and V_S denote thickness of pressure tube, velocity of longitudinal wave and shear wave, respectively. The expression of velocity ratio is derived from the time-of-flight of each echo in Eq. (1).

$$\frac{V_L}{V_S} = 1 + \frac{2(RS - B_1)}{B_2 - B_1} \tag{2}$$

Since both longitudinal and shear wave can be measured with the same normal beam ultrasonic transducer, it is a relatively simple, convenient and reliable method. As shown in Table 1, the velocity ratio is in the range of 2.01 ~ 3.08 depending on the size of blister in the zirconium matrix. When we assume the fraction of blister to the total beam path is 10%, the velocity ratio is calculated as 2.12.

3. Experimental

3.1. Preparation of Blister Specimen

Curved rectangular specimens were cut out of Zr-2.5Nb pressure tube and charged with hydrogen of 200 ppm by an electrolytic method in accordance with our procedures for characterization of Zr-2.5Nb pressure tubes [12]. Vacuum annealing was conducted at the temperature above thermal solid solubility to homogenize the distribution of charged hydrogen within the specimens. As a result, hydrides were found to uniformly distribute over the whole cross section of the specimen. Hydrogen concentration in the specimen was verified by chemical analysis.

The hydride blisters were grown on the outer surface of the curved specimen by applying thermal gradients across the thickness using the

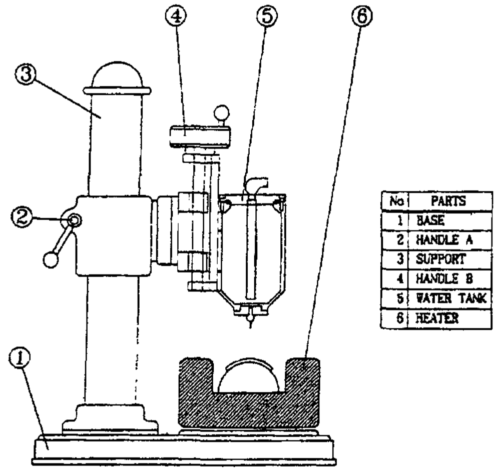

Fig. 3. Experimental Device for Blister Formation and Growth

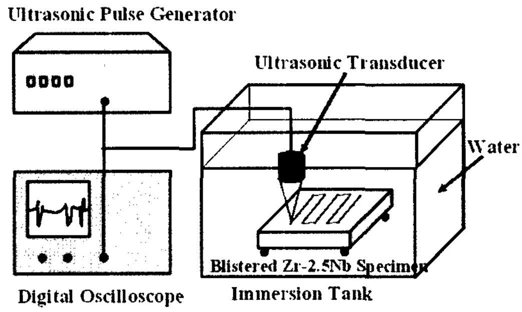

Fig. 4. Block Diagram of Experimental Setup for Blister Detection by Ultrasonic Velocity Ratio Method

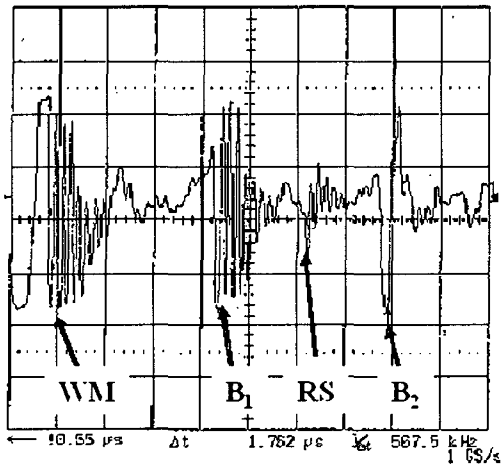

Fig. 5. Typical Echo Pattern in Oscilloscope Screen

device shown in Fig. 3. The inner surface of the specimen sits on the aluminum block kept at 415 ±2℃, while cold spot was made by keeping the outer surface in contact with the aluminum finger cooled by flowing water of 14~16℃. The hydride blisters grew during 7~42 days under temperature gradient. A finite element analysis indicated that the temperature difference between the cold spot and inner surface of pressure tube are around 190 ℃ [3]. This temperature gradient is similar to the thermal condition of contact between pressure tube and calandria tube in the reactor, but higher temperature of the inner surface of the pressure tube and high concentration of hydrogen accelerate the formation and growth of blister in this study.

3.2. Detection of Hydride Blister by Ultrasonic Velocity Ratio Method

Fig. 4 shows a block diagram of experimental setup and a typical waveform displayed on the oscilloscope is shown in Fig. 5. A focused immersion ultrasonic transducer with a frequency of 10 MHz, diameter of 6.35 mm, and spherical focal length of 20.3 mm was scanned along the axial and circumferential direction from the inner surface of pressure tube for transmission and reception of the ultrasound. The longitudinal wave signals reflected at the outer surface of pressure tube(B_1 and B_2), and the signal converted from longitudinal to the shear wave at the outer surface (RS) are seen in Fig. 5. An ultrasonic pulser (JSR DPR -50 plus, Maximum frequency; 50 = MHz) and a digital oscilloscope (Lecroy LC 574C, sampling frequency = 1 GHz) were used to drive the ultrasonic transducer and measure the time-of-flight of reflected echoes. The first minimal points from each multiple oscillating signal are referred for the measurement of time-of-flight with an accuracy of 10^{-9} seconds.

Optimum water distance between the ultrasonic transducer and the inner surface of the pressure tube is required to get the reflected shear mode, which mainly depends on the diameter and focal length of the transducer. Water distance was adjusted to maximize the reflected shear echo signal.

4. Results and Discussion

A typical cross-section view of hydride blister is shown in Fig. 6. Many hydride platelets agglomerate at the cold spot, creating a swollen area and some cracks. A clear boundary was shown between the zirconium matrix and the hydride blister with some cracks open to the outer surface of the tube. The depth of the blisters ranged from 50 to 300 μm depending on the hydrogen concentration and the time when the cold spot was exposed to the thermal gradient. The ratio of blister depth to height of the blisters is estimated as 8 to 1 from the comprehensive study on the mechanism of formation and growth kinetics of hydride blisters [3].

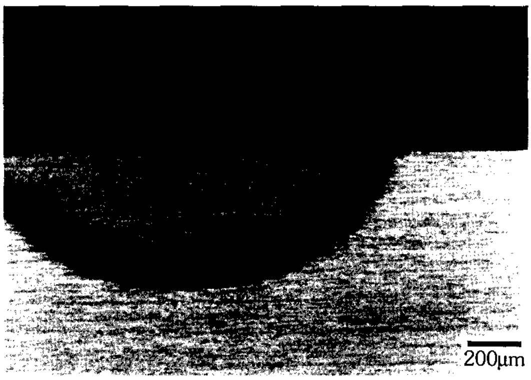

Fig. 6. Typical Cross-sectional View of Hydride Blister

The reflected beam intensity was estimated at the blister interface for the case of longitudinal wave, shear wave, and mode conversion from longitudinal to shear wave, respectively. The fraction of ultrasonic energy reflected from the interface depends on the ratio of acoustic impedances (Z_2/Z_1) and the angle of incidence. When the angel of incidence is 0° (normal incidence), the reflection coefficient, R, defined as the ratio of reflected beam intensity, I_r, to the incident beam intensity, I_i, is given by [13]:

$$R = \frac{I_r}{I_i} = (\frac{Z_2 - Z_1}{Z_2 + Z_1})^2 = (\frac{r-1}{r+1})^2 \qquad (3)$$

where Z_1 is the acoustic impedance of medium 1 (zirconium matrix), Z_2 is the acoustic impedance of medium 2 (hydride blister), and r equals Z_2/Z_1 and is the impedance ratio or mismatch factor. From Eq. (3), the fraction of acoustic energy reflected at the hydride blister interface are calculated as 0.08% for the case of longitudinal wave and 3.4% for the case of shear wave. It means that most of ultrasonic energy transmitted through the interface and little echo signal is expected from the interface. In other words, even though interface between blister and zirconium matrix is clearly defined with an optical microscope as in Fig. 6, it is hard to see the reflected signal at the interface by ultrasonic means because most of ultrasonic energy is transmitted through the interface.

The depths of blister normally observed in reactors range from few tens to hundreds micrometer while the critical depth of blister for the fracture of pressure tubes is reported to be around ~450 μm [2]. When a blister of 500 μm deep is formed on the pressure tube of 4.2 mm thick, the ratio of time-of-flight in the region with blister to those in the region without blister is estimated as 2.3 % for the case of longitudinal wave (L-L mode), 3.0% for the case of shear

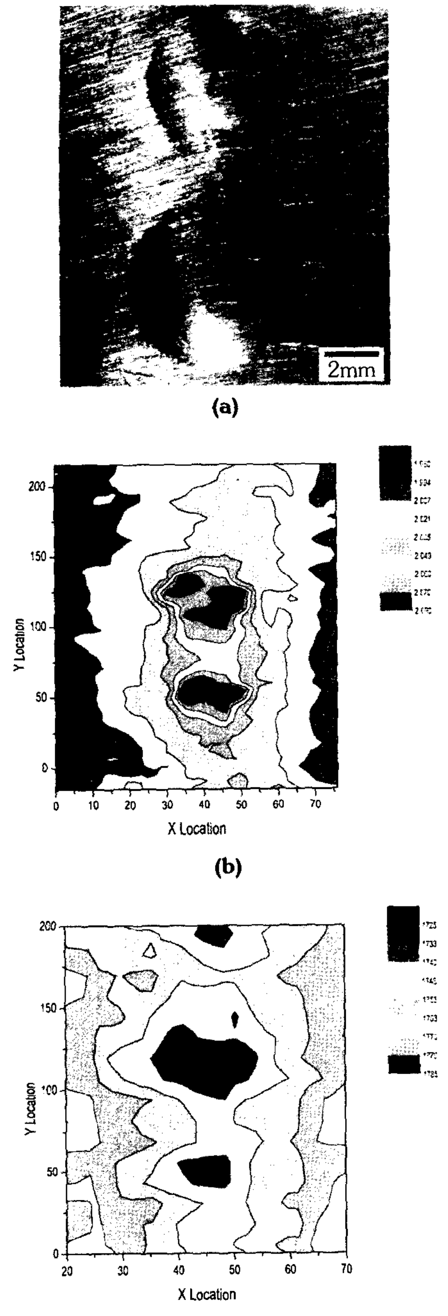

Fig. 7. (a) Optical Image of Outer Shape of Blisters Grown for 42 days with Hydrogen Concentration of 200 ppm, (b) Contour Plot with Velocity Ratio Method, (c) Contour Plot of Longitudinal Wave Time-of-flight Method

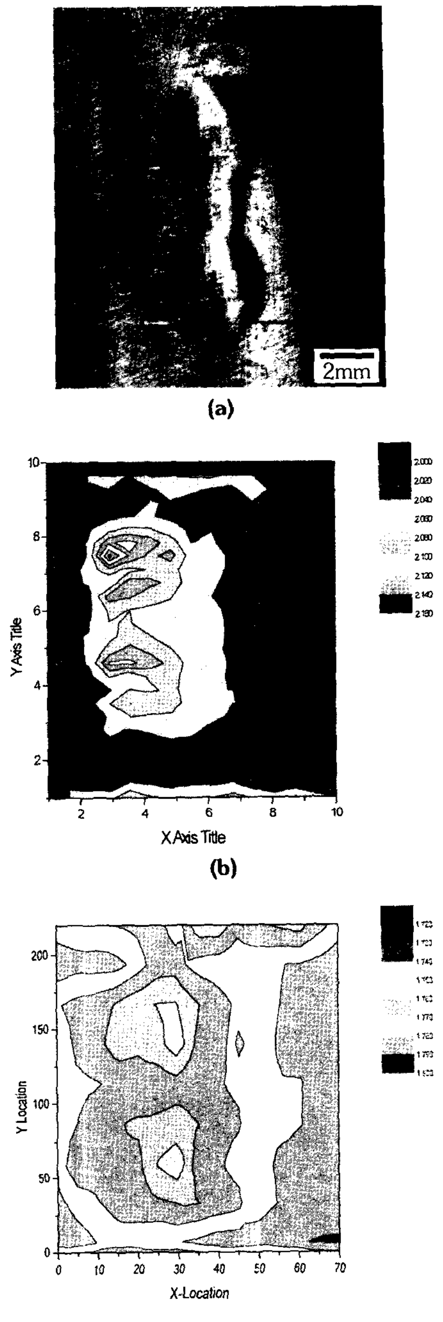

Fig. 8. (a) Optical Image of Outer Shape of Blisters Grown for 30 Days with Hydrogen Concentration of 200 ppm, (b) Contour Plot with Velocity Ratio Method, (c) Contour Plot of Longitudinal Wave Time-of-flight Method

wave(S-S mode), and 16.9% for the case of longitudinal-shear mode conversion (L-S mode). This fact infers the velocity ratio method is more effective in detecting the blisters than other conventional time-of-flight methods.

Time differences between the first and the second back wall echoes, B_2-B_1 and the reflected shear and first back wall echoes, can be measured by a digital oscilloscope with accuracy of 10^{-9} seconds. Using the values of velocity ratio calculated from Eq. (2), we can plot a contour of velocity ratio which represents the blister formed on the surface of the Zr-2.5Nb pressure tube as shown in Fig. 7. Fig. 7(a) shows an optical image of outer surface of blister specimen grown for 42 days with the hydrogen concentration of 200 ppm. Fig. 7(b) and (c) show contour plots made by the ultrasonic velocity ratio method and conventional longitudinal time-of-flight method, respectively. Both methods reproduce the actual size and shape of hydride blisters. Similar results are obtained from the specimen grown for 30 days with the same hydrogen concentration of 200 ppm, shown in Fig. 8. This indicates that both the velocity ratio method and the conventional time-of-flight method can be applicable to the detection of large blisters.

However, the smaller the blister becomes, the more difficult their detection, leading to a demonstration of the advantage of the velocity ratio method over the conventional time-of-flight method. Fig. 9(a) shows an optical image of a smaller blister grown for 7 days under the same condition as shown in Figs. 7 and 8. The velocity ratio method clearly reproduced the contour of the blister as shown in Fig. 9(b), while the conventional time-of-flight method could not as shown in Fig. 9(c). Therefore, it is concluded that the velocity ratio method is a very effective tool to detect and reconstruct smaller blisters. Even though it is yet to be investigated further how

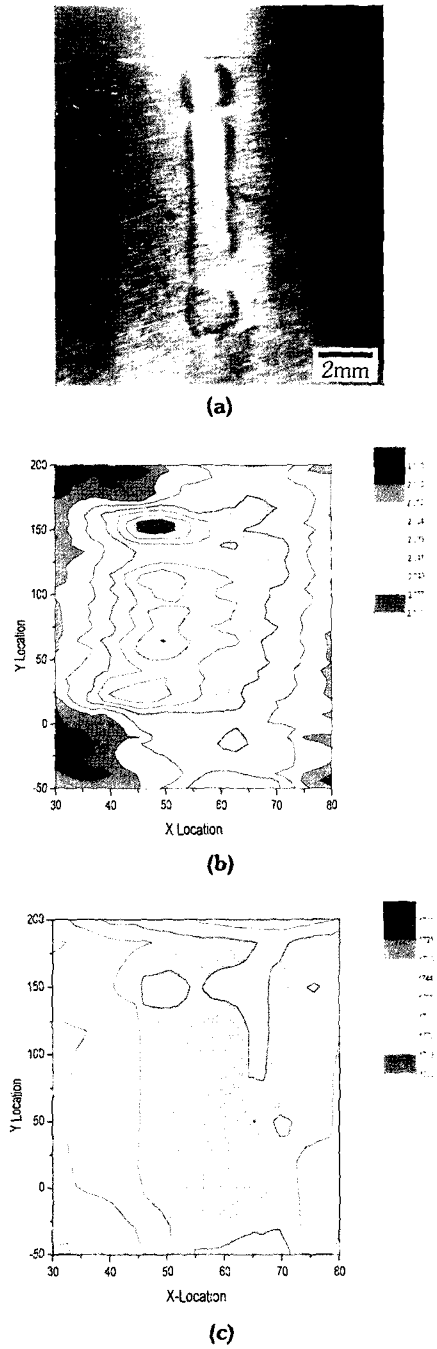

Fig. 9. (a) Optical Image of Outer Shape of Blisters Grown for 7 Days with Hydrogen Concentration of 200 ppm, (b) Contour Plot with Velocity Ratio Method, (c) Contour Plot of Longitudinal Wave Time-of-flight Method

small blisters can be detected by this method, it seems to contribute to a progress in non-destructive detection of blisters grown on the Zr-2.5Nb pressure tubes.

5. Implementation to In-service Inspection of Pressure Tube

We successfully detected the hydride blister on the outer surface of pressure tubes by the velocity ratio method with a similar ultrasonic transducer used in ultrasonic module of the fuel channel inspection systems (10 MHz, focused immersion type).

In the fuel channel inspection system used for the in-service inspection of pressure tube, such as AFCIS (AECL Fuel Channel Inspection System), the digital ultrasonic data of normal beam channel could be obtained from ultrasonic data acquisition system. If the measurement of time-of-flight of each signal including the RS signal is done successfully, we can get the values of velocity ratio at the positions by measuring the time differences, B_2-B_1 and RS-B_1. Contour plot of velocity ratio could be possible by an accurate helical scan of inspection head and signal processing algorithm.

6. Conclusions

An ultrasonic velocity ratio method has been developed for non-destructive detection of hydride blisters in the pressure tube. Hydride blister specimens were fabricated from the Zr-2.5Nb pressure tube specimens with a steady state thermal diffusion device. Time-of-flights of longitudinal wave and reflected shear wave from the outer surface of the pressure tube were measured and converted to the values of a velocity ratio of longitudinal wave to shear wave (V_L/V_S). From the contour plots by scanning inside the pressure tube, the ultrasonic velocity ratio method shows superior capability of detection and reconstruction of the shape of smaller blisters to the conventional time-of-flight method.

This method could be implemented during in-service inspection if we could acquire reflected shear echo signal from the normal beam ultrasonic transducer in the fuel channel inspection systems. By measuring the time-of-flight of the first and the second back wall echo signal and reflected shear echo signal between the first and second back wall echo, the values of velocity ratio could be calculated and plotted in a x-y plane.

Acknowledgements

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