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Assessment of hydrogen levels in Zircaloy-2 by non-destructive testing

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Abstract

A non-destructive assessment of Zircaloy-2 samples charged with hydrogen in the range of 50 to 1150 mg/kg has been made using ultrasonic and eddy current testing. It has been found that the ratio of the longitudinal to the shear wave velocity is a parameter which can be directly correlated with the hydrogen content up to a level of 100 to 200 mg/kg. This parameter together with the values of longitudinal and shear wave velocities can be utilized in a multi-parametric correlation approach for estimation of higher levels of the hydrogen content (up to 1150 mg/kg). The sensitivity at different ranges has been found to be acceptable. Ultrasonic attenuation measurements at higher frequencies and eddy current test parameter are also effective for estimation of hydrogen levels above 250 mg/kg in zirconium alloys. Microstructural characterization including TEM studies have been carried out for studying the influence of the type and the morphology of hydride precipitates on ultrasonic parameters. © 1998 Elsevier Science B.V.

1. Introduction

The coolant channel assembly is an important component in the pressurized heavy water reactors (PHWR). Each channel consists of two concentric zirconium alloy tubes with an annular space between them. The fuel bundles are housed inside the inner tube through which heavy water at high pressure flows to extract the heat generated in the fuel bundles. Zirconium alloys are chosen for these in-core applications because of their low thermal neutron absorption cross section, good corrosion resistance and high melting point. However, they are susceptible to hydride embrittlement. Zircaloy-2 tubes contain hydrogen in the range 10 to 15 mg/kg before they are put into service. During the service life of a reactor, this level goes up due to the pick up of hydrogen which is a corrosion product of Zircaloy-2 with high temperature water. Hydrogen concentration beyond the solubility limit results in the formation of brittle hydrides, which significantly reduce the ductility and the fracture toughness of the tubes [1,2]. The in-service deterioration is further aggravated by irradiation damage. In view of these, the hydrogen concentration in the pressure tubes is to be kept below 50 mg/kg or so. However, a hydrogen assisted cracking can still take place in pressure tubes by a mechanism known as delayed hydrogen cracking (DHC) [3–5], the risk of which can be significantly reduced by lowering the level of hydrogen and the rate of hydrogen pickup. In order to improve the chance of detecting a crack early and to meet the leak before break criterion [6], it is desirable to monitor the hydrogen levels in coolant channels of power reactors by non destructive testing (NDT) techniques to assess their health and to decide as to the optimum use and the replacement of the pressure tubes.

Many of the common NDT parameters appear to be useful to monitor the hydrogen level in zirconium alloys. This study attempts at assessing the influence of the hydrogen content on NDT parameters pertaining to ultrasonic and eddy current techniques. The presence of hydrogen and radiation damage can cause change in density and elastic modulii of the alloy. Therefore, ultrasonic velocity,

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which is influenced by these changes is considered as a possible NDT parameter to monitor the hydrogen level. The attenuation may also be affected, particularly at high frequencies, by the presence of hydrides and hence it is another promising candidate parameter for NDT. Additionally, it is expected that the electrical conductivity of the alloy may change due to hydrogen pick up and radiation damage. This variation in conductivity with hydrogen pick up can be measured using a standard eddy current tester and the hydrogen content can be reliably estimated once a correlation is established between them.

In order to get a better understanding of the behavior of the material, detailed metallographic examination and X-ray diffraction studies have been carried out on unirradiated materials with different hydrogen concentrations. It is assumed that irradiation damage would produce a shift in the absolute value of the NDT parameter but the trend observed on the influence of the hydrogen content on various NDT parameters in the unirradiated material would be valid for the irradiated material as well. The sensitivity of the various NDT techniques has been studied by keeping the limitations of the techniques in view, and simple ways of enhancing the sensitivities have been explored. Finally, this paper brings out the effective NDT technique and NDT parameter suitable for a reliable measurement of the hydrogen content in coolant channels of PHWR.

2. Experimental

2.1. Specimen preparation

Nuclear grade Zircaloy-2 specimens of size 30 × 30 × 3 mm, initially containing 10 to 25 mg/kg hydrogen were hydrogen charged to 50, 70, 100, 250, 550 and 1150 mg/kg by high temperature autoclaving in dilute LiOH solution (0.5 M). Details of the procedure have been given elsewhere [7]. The charging was done slowly in several stages, by analyzing hydrogen content after every step so that the hydrides would be uniformly distributed in the specimen and the hydrogen contents would not vary by more than ±5% among samples (5 numbers in each batch). Characterization of the samples with respect to the phases present and their distribution has been carried out using optical metallography, transmission electron microscopy (TEM) and X-ray diffraction techniques. TEM studies were performed on thin slices of the specimens thinned further by the dual jet thinning technique using an electrolytic solution containing perchloric acid, methanol and *n*-butanol at 20 V and 233 K (−40°C).

2.2. Ultrasonic velocity measurements

As the volume fraction of the hydrides was very small, high precision ultrasonic velocity measurements were carried out using the cross correlation technique. The details of the cross correlation technique are available elsewhere [8]. This technique measures the ultrasonic transit time between two selected back wall echoes precisely with an accuracy of 0.1 ns. A 10 MHz longitudinal wave probe and a 2 MHz shear wave probe were used for the ultrasonic velocity measurements. The measurements were made with ultrasonic waves propagating in the thickness direction. In the case of shear waves polarization parallel to and perpendicular to the rolling direction were used to take into account the effect of the texture present in the Zircaloy-2 specimens. The three velocities measured are designated by the notations V_{31} (shear waves propagating

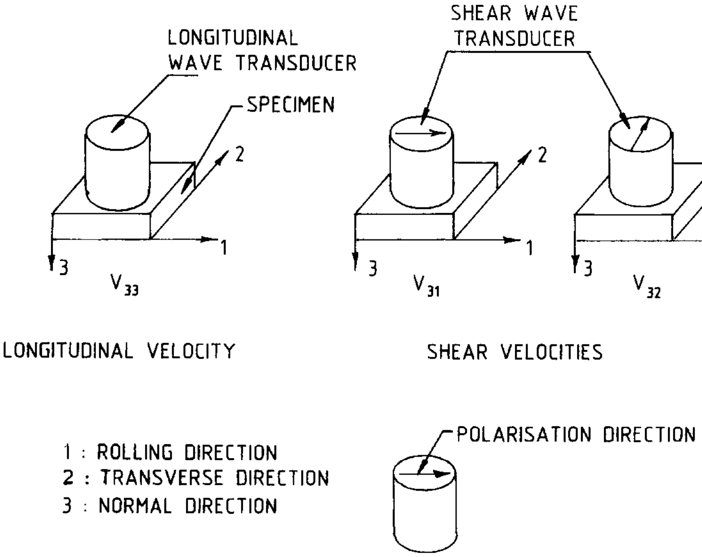

Fig. 1. Arrangement for measuring different velocities.

in the normal direction with polarization parallel to the rolling direction) and V_{32} (shear waves propagating in the normal direction with polarization perpendicular to rolling direction) and V_{33} (longitudinal waves propagating along the thickness direction, i.e., normal direction), Fig. 1 shows the specimen-sensor arrangement for measuring these three velocities. The third and the fourth back wall echoes were used for transit time measurements. Ratios of the measured

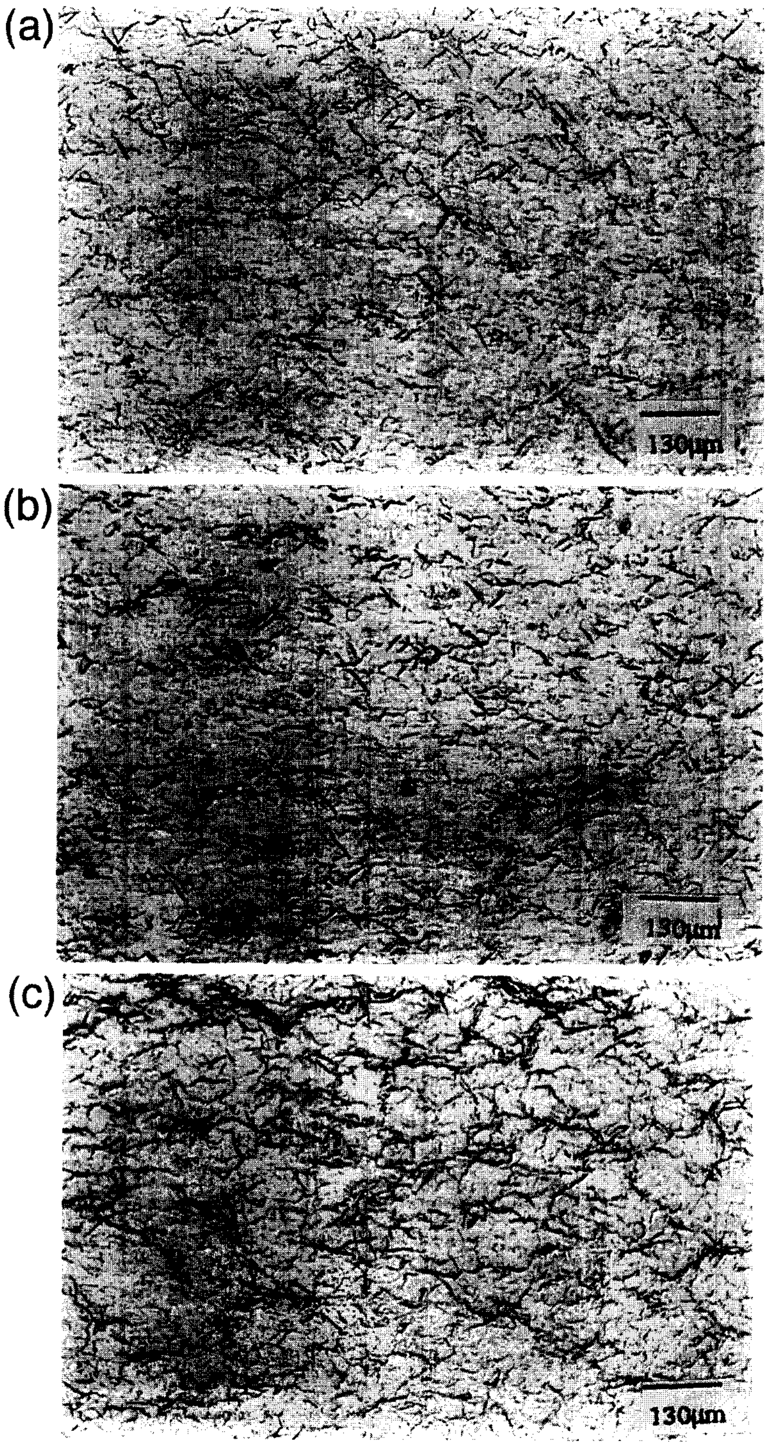

Fig. 2. Photomicrographs of Zircaloy-2 having different hydrogen concentration (a) 85 mg/kg, (b) 235 mg/kg and (c) 600 mg/kg.

velocities were computed (Section 3.3) to evaluate their suitability for the prediction of the hydrogen concentration. While the error due to thickness measurements could not be eliminated while assessing the influence of hydrogen concentration using longitudinal and shear wave velocities independently, the use of the ratio of any two velocities for correlating it with hydrogen concentration would eliminate the necessity for thickness measurement, thus eliminating the error associated with it. In such a situation, one has to measure only the ratio of the transit times of the corresponding waves which would be the same as the ratio of the velocities. Identification of a ratio parameter rather

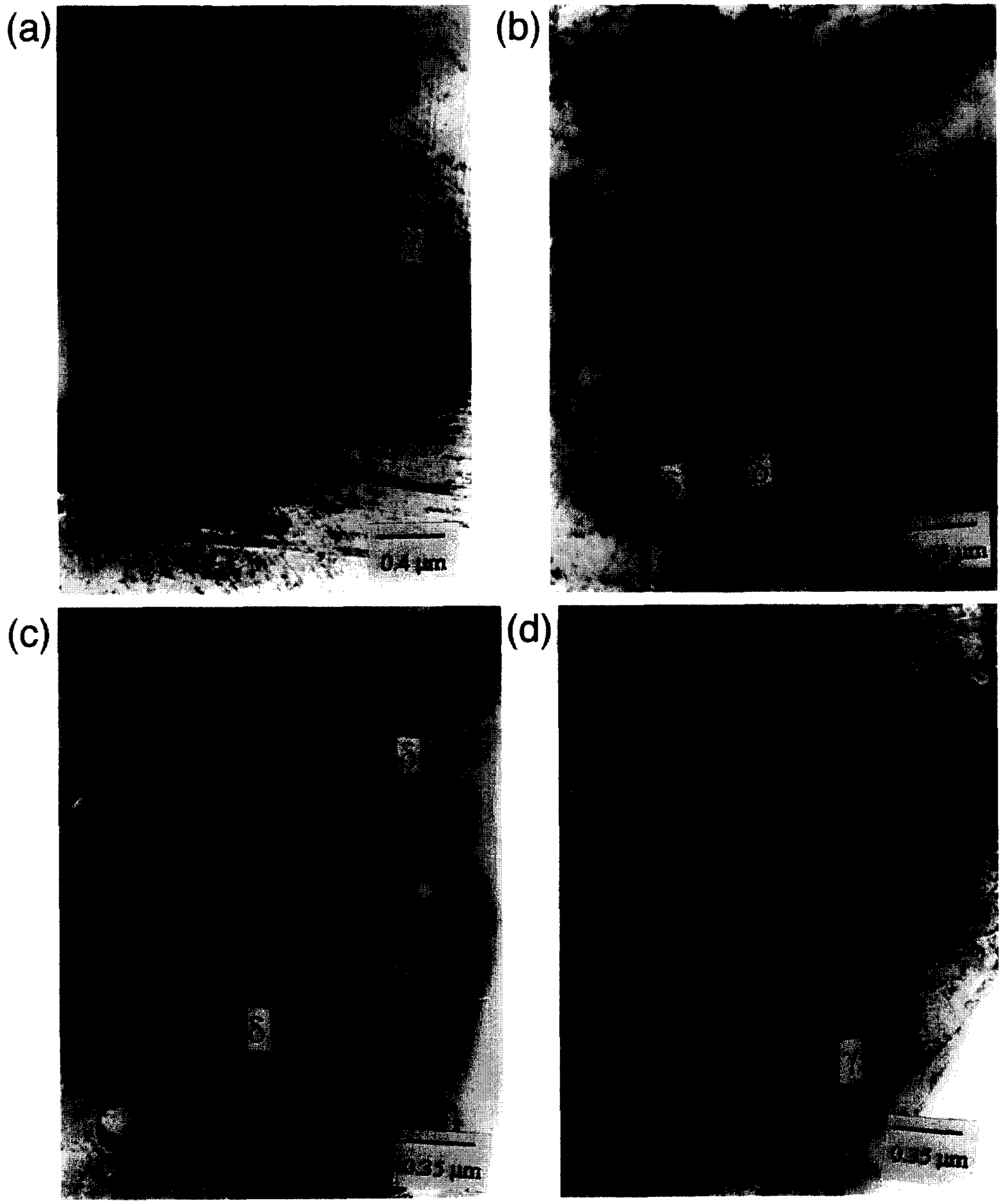

Fig. 3. Hydride morphology in Zircaloy-2 samples containing different hydrogen levels as observed in TEM (a) and (b) 85 mg/kg; (c) and (d) 235 mg/kg; (e) 600 mg/kg. SAD patterns of hydrides in Zircaloy-2 specimens containing different hydrogen levels (f) 85 mg/kg; (g) and (h) 600 mg/kg.

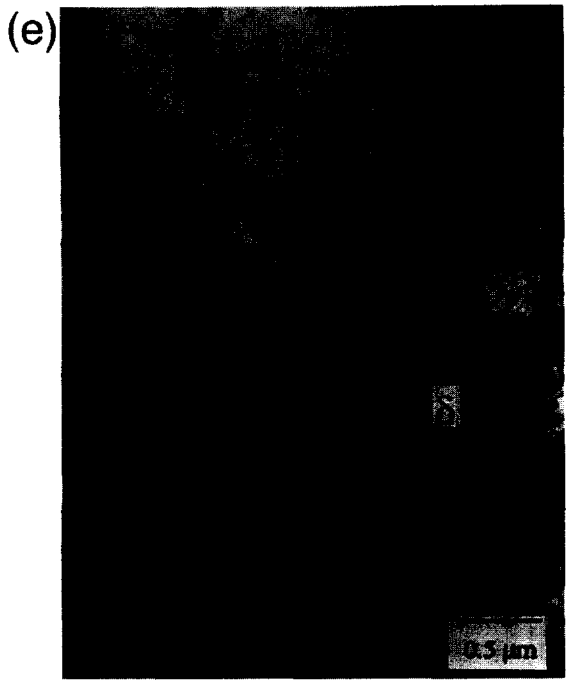

Fig. 3. (cont.)

than of an absolute velocity is advantageous not only because it improved accuracy for the evaluation of hydrogen concentration, but also it enables one to bypass the practical difficulties in making the thickness measurements.

2.3. Ultrasonic attenuation measurements

Ultrasonic attenuation measurements in Zircaloy-2 specimens with different hydrogen concentrations were made at the frequencies of 23, 50 and 75 MHz. These high frequency attenuation measurements were carried out using a high frequency ultrasonic system (Model 10150PR pulser-receiver and Model 10150G stepless gate) supplied by M/s. Accu-Tron, USA. The measurements were made by the contact technique. It should be pointed out that the use of the contact technique had introduced a relatively high scatter in the data obtained. Additionally, the high frequency system did not have a rectified output module. Therefore, attenuation measurements were made by using the peaks of the rf signals of successive back wall echoes. The peaks were measured from the display of the rf signal on an oscilloscope. Since there is not much variation in the pattern of the signals (number of cycles, shape of rising and falling portions, etc.) of successive back wall echoes except the reduction in the overall amplitude, the use of the peaks in the rf signals of successive back wall echoes was thought to be adequate and reasonable for attenuation measurements. In spite of these limitations, meaningful correlations between the microstructural changes and the attenuation could be obtained. The high frequency longitudinal wave transducers used were of contact type with delay line and were supplied by M/s. Accu-Tron, USA.

2.4. Conductivity measurements

The conductivity measurements were made using model AP/04 ALCOPROBE supplied by M/s. Inspection Instruments (NDT), Berkshire. The conductivity values were given in terms of the percentage of the international annealed copper standard (IACS). Standard specimens having electrical conductivity values of 1% and 2.85% IACS were used to adjust 0% and 100% respectively, on the display of the equipment. A linear relationship was assumed in the selected range. The testing frequency employed in this probe was 100 kHz. Lift off corrections were incorporated during the testing. As the frequency used was low (100 kHz), it was thought that by using a regular eddy current system and employing a higher frequency, the sensitivity may be improved for the detection of hydrogen. Accordingly, ECT was carried out using a Model MIZ-20A eddy current equipment supplied by M/s. Zetec, USA. Considering the specimen geometry and the variable to be monitored, an absolute surface probe was employed. ECT measurements were carried out at various test frequencies ranging from 100 kHz to 1 MHz. Though lower frequencies are associated with higher depths of penetration, lower sensitivities restrict the use of the lower frequencies. On the contrary, unwanted signal due to lift-off increases with increasing frequency. In view of the opposing effects of the frequency, a test frequency of 400 kHz was found to be optimum. At this frequency, the depth of penetration would be approximately 1 mm. A good phase discrimination between the signals associated with lift-off and conductivity variations could also be observed and this could be utilized to avoid any interference from the lift-off.

3. Results

3.1. Characterization of hydrogen charged Zircaloy-2 specimens

Optical microscopic studies revealed that the hydrides are very uniformly distributed through the length and thickness of the specimens (Fig. 2(a)–(c)). X-ray diffraction studies could not reveal any significant hydride peaks as the volume fraction of the hydrides was smaller than the detection sensitivity of the XRD technique. Fig. 3(a)–(e) show the TEM micrographs of the hydrided alloys at different hydrogen levels. The following features were noticed from the TEM study:

(i) Two types of hydrides were noticed: twinned hydrides (type I) and plate shaped hydrides (type II indicated by γ and δ in Fig. 3(a)–(e). Those of the type I were

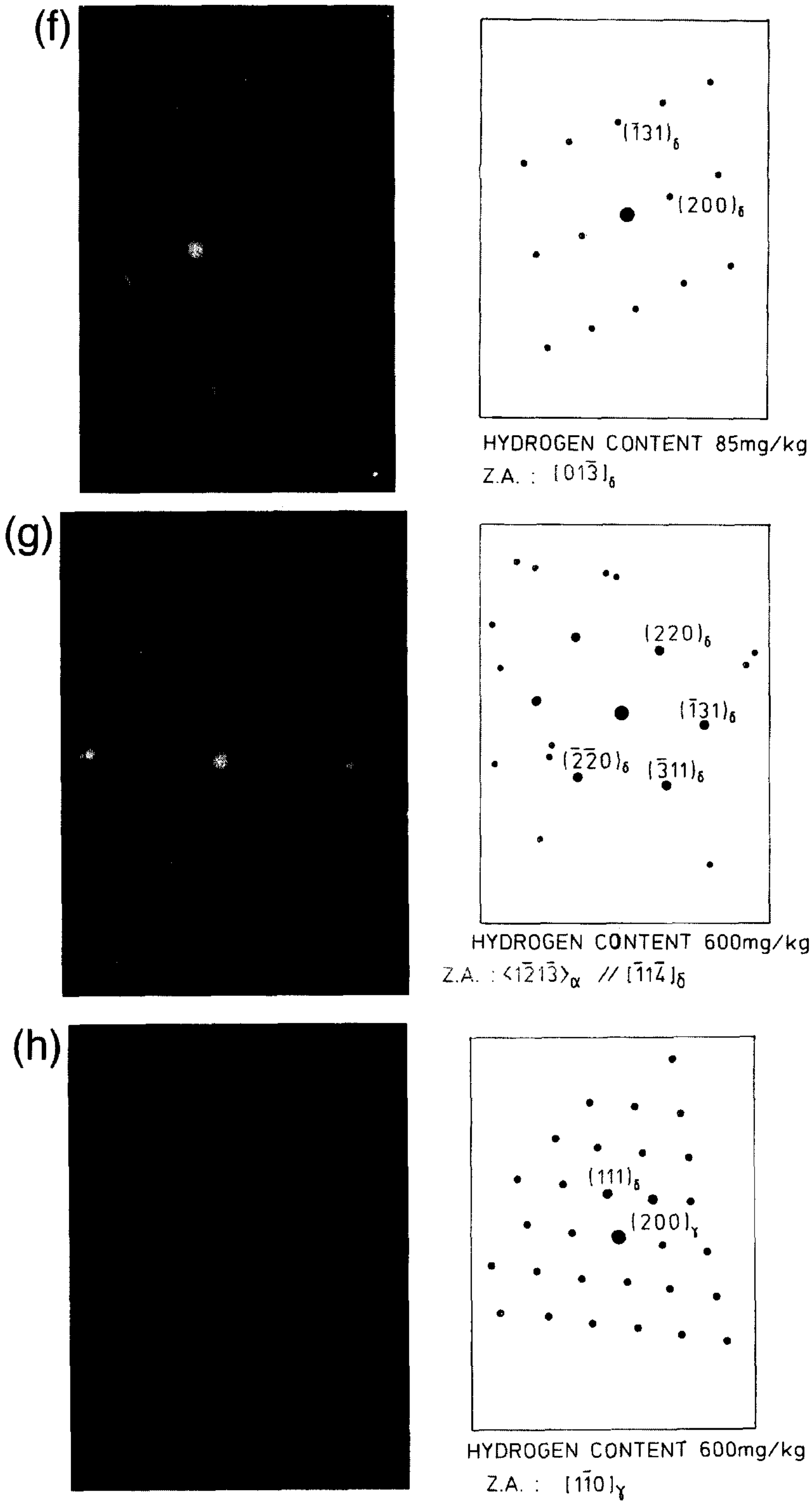
(f)
(1̄31)δ
(200)δ
HYDROGEN CONTENT 85mg/kg
Z.A. : [013̄]δ
(g)
(220)δ
(1̄31)δ
(2̄2̄0)δ
(3̄11)δ
HYDROGEN CONTENT 600mg/kg
Z.A. : <12̄13̄>α // [1̄14̄]δ
(h)
(111)δ
(200)γ
HYDROGEN CONTENT 600mg/kg
Z.A. : [11̄0]γ

Fig. 3. (cont.)

characterized by regions containing a stack of fine twins. Sometimes, more than one set of twins were present in the same region. Selected area diffraction (SAD) patterns (Fig. 3(f)–(h)) revealed the presence of different variants of tetragonal hydride crystals which were twin related with each other.

(ii) The second variety (Type II) was a well developed plate shaped hydride, the volume fraction of which increased with the hydrogen content of the sample.

(iii) The type I and the type II hydrides were identified to be the metastable γ and the equilibrium δ hydride phases respectively based on the SAD patterns taken from them (Fig. 3(f)–(h)).

(iv) In low hydrogen specimens, hydride precipitation was associated with a high degree of lattice strain as evidenced by the high dislocation density in the vicinity of the precipitate (Fig. 3(b)–(c)). At higher hydrogen concentrations, the dislocation density in the α-Zr matrix was seen to be reduced.

(v) Analysis of SAD patterns and estimation of lattice parameters revealed that the samples containing less than about 100 mg/kg H exhibit predominantly γ hydride precipitates. Samples with higher hydrogen contents show a mixture of γ and δ hydride precipitates.

The present observation agrees with the earlier investigations [9] in that low hydrogen levels and short duration of charging favor the γ-hydride precipitates and high hydrogen levels and longer duration of charging favor the δ hydrides.

3.2. Conductivity measurements

Conductivity measurements using ALCOPROBE did not show any detectable change in the conductivity with the increase in the hydrogen concentration up to 100 mg/kg. However, from 200 mg/kg onwards, a decrease in the conductivity from 2.6% IACS to 2.45% IACS was observed with increase in the hydrogen concentration up to 530 mg/kg of hydrogen. Fig. 4 shows the variation in the eddy current parameter with the hydrogen concentration. It can be seen from Fig. 4 that there is no detectable change in the eddy current parameter among the specimens with 15, 70 and 100 mg/kg of hydrogen. However, there is an increase in the eddy current parameter in the specimens with 240 and 530 mg/kg of hydrogen. The ECT parameter is found to increase from 0 value to 4 arb. units in the specimen with 240 mg/kg of hydrogen, and to 21 arb. units in the specimen with 530 mg/kg of hydrogen. Under the experimental conditions used, the ECT parameter is found to saturate in the case of specimen with 1150 mg/kg of hydrogen. These results indicate that unambiguous assessment of hydrogen concentration in Zircaloy-2 using ALCOPROBE and eddy current test system is possible from 200 mg/kg of hydrogen onwards. Even though it is expected that the ECT system should have higher sensitivity for detection of hydrogen because of the higher frequency (400 kHz) employed as compared to that (100 kHz) used in the ALCOPROBE, such a trend is not observed.

3.3. Ultrasonic velocity measurements

Fig. 5 shows the variation in the ultrasonic longitudinal and shear wave velocities with the hydrogen concentration. It can be seen from the Fig. 5 that the changes in the velocities with increase in the hydrogen concentration are

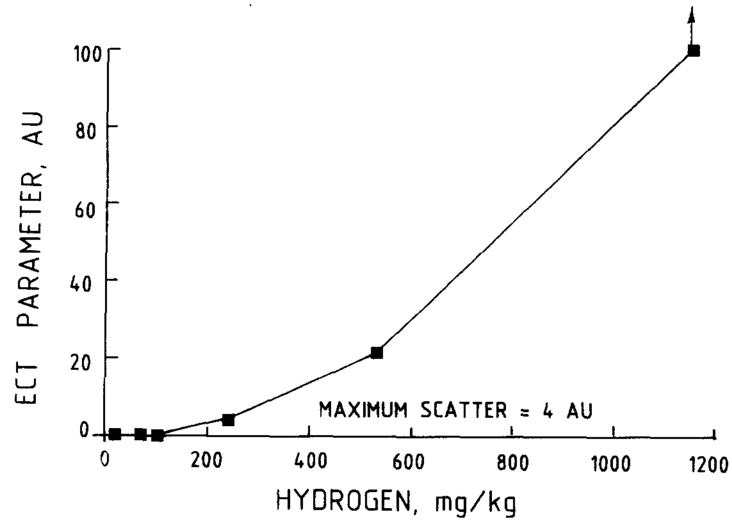

Fig. 4. Variation in eddy current parameter with hydrogen concentration in Zircaloy-2.

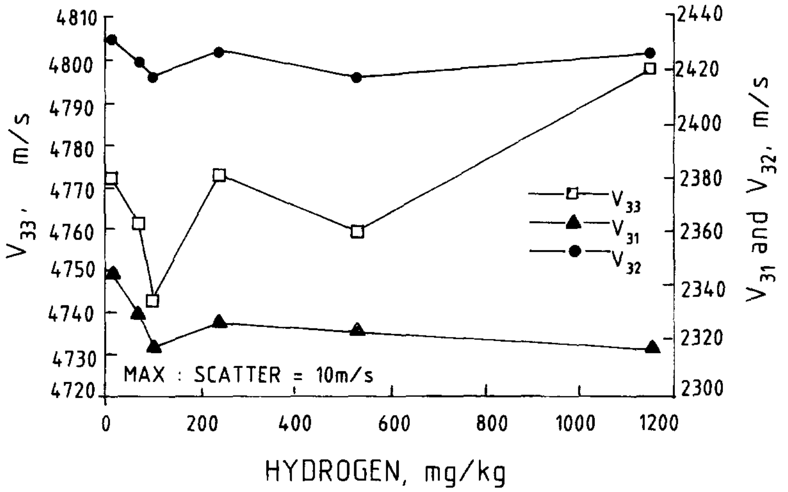

Fig. 5. Variation in longitudinal and shear velocities with hydrogen concentration in Zircaloy-2.

complex. There is a decrease in the velocity for longitudinal and shear waves with increase in the hydrogen concentration up to 100 mg/kg, followed by an increase in the velocities up to hydrogen concentration of 250 mg/kg and then followed by a slight decrease in the velocities in samples with 500 mg/kg hydrogen and again considerable increase in the velocities in the sample with 1150 mg/kg hydrogen. The fluctuations observed in the velocities may not permit estimation of the hydrogen concentration in the total range studied. However, both the permitted and the normally encountered hydrogen concentration is Zircaloy-2 coolant tubes is below 100 mg/kg [10]. In this range, the velocity is found to decrease with increase in the hydrogen concentration and hence the estimation of the hydrogen concentration in Zircaloy-2 up to 100 mg/kg is possible by adopting ultrasonic velocity measurements. The decrease in the velocity in the specimens with 100 mg/kg of hydrogen as compared to that in the reference specimen with 15 mg/kg of hydrogen is about 0.6% for V_{33} and 1.2% and 0.5% for V_{31} and V_{32} respectively. These changes

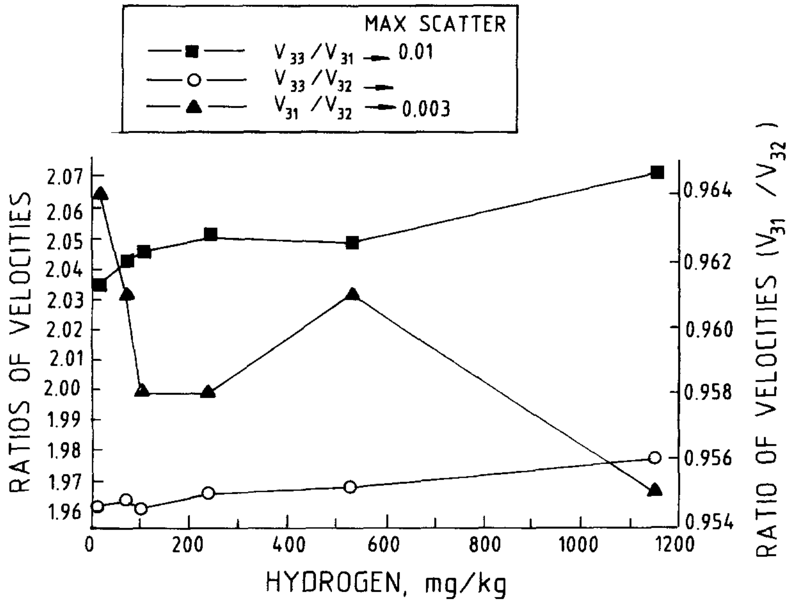

Fig. 6. Variation in ratios of velocities with hydrogen concentration in Zircaloy-2.

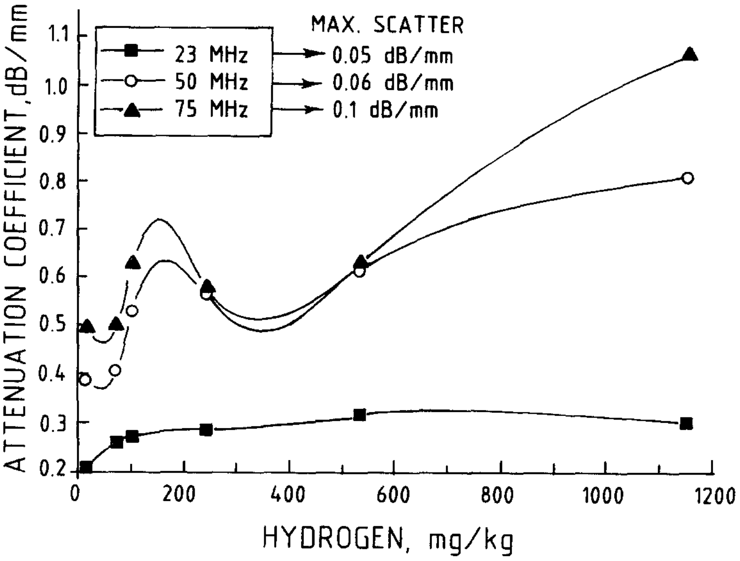

Fig. 7. Variation in ultrasonic attenuation coefficient with hydrogen concentration in Zircaloy-2.

are more than the ±0.1 accuracy obtainable in the velocity measurements by using the cross correlation technique adopted in this study. The use of V_{31} (shear waves polarized parallel to the rolling direction) would be ideal for the estimation of the hydrogen concentration with high sensitivity as this velocity shows maximum change for a given concentration of hydrogen.

The accuracy of the velocity measurements by the cross correlation technique directly depends on the thickness of the specimen. While the error due to thickness measurements cannot be eliminated from the longitudinal and shear velocities measured independently, the ratio of these two velocities would be free from the error associated with the thickness measurements. The ratios of the velocities, V_{33}/V_{31}, V_{33}/V_{32} and V_{31}/V_{32} are also computed for different specimens. The variation in these ratios with hydrogen concentration is shown in Fig. 6. It can be seen from the Fig. 6 that, in general, there is a monotonic increase in the velocity ratios V_{33}/V_{31} and V_{33}/V_{32} with increase in the hydrogen concentration in the total range studied, i.e. up to 1150 mg/kg. On the other hand, the ratio V_{31}/V_{32} is found to decrease with the increase in the hydrogen concentration. The V_{33}/V_{31} shows a change (increase) of 0.5% at the hydrogen concentration of 100 mg/kg and 1.8% for the hydrogen concentration of 1150 mg/kg. These values are higher as compared to the corresponding values for V_{33}/V_{32} and V_{31}/V_{32}, for example, the values for V_{33}/V_{32} are 0.25% and 0.9% respectively. This suggests that the use of V_{33}/V_{31} would give better accuracy in the estimation of the hydrogen concentration. This parameter together with the independent longitudinal and shear wave velocities can be utilized in a multi-parametric correlation approach for estimation of the hydrogen concentration up to 1150 mg/kg with acceptable sensitivity in different ranges of hydrogen concentration i.e. giving weighage for V_{31} or V_{33} below 100 mg/kg and for V_{33}/V_{31} above 100 mg/kg. This is because of the fact that the change in the V_{33}/V_{31} up to 100 mg/kg is small (less than 0.5%) as compared to the change in the V_{33} (0.6%) and, in particular, V_{31} (1.2%).

3.4. Ultrasonic attenuation measurements

The variation in the attenuation coefficient with the hydrogen concentration in different specimens at frequencies 23, 50 and 75 MHz are shown in Fig. 7. It can be seen from the Fig. 7 that the attenuation coefficient increases, in general, with the increase in the hydrogen concentration at all the frequencies. Even though, the measurements at 23 MHz show a tendency for an increase in the attenuation coefficient with the increase in the hydrogen concentration, it is too small to be used for reliable assessment of the hydrogen concentration when compared to the scatter band. The change in the attenuation coefficient within 100 mg/kg of hydrogen is very small even at the frequencies 50 and 75 MHz. When compared this increase with the scatter band, this data is also not reliable for the assessment of the hydrogen concentration. However, beyond 100 mg/kg of hydrogen, there is a definite increase in the attenuation coefficient at the frequencies 50 and 75 MHz. Therefore, these results suggest that, at higher frequencies, attenuation measurements can be adopted to assess the hydrogen concentration beyond 100 mg/kg in Zircaloy-2.

4. Discussion

For the lower hydrogen concentrations used in this study, the conductivity values of the Zircaloy-2 with different hydrogen concentrations is not available in the literature for comparison. It was reported that $ZrH_{1.96}$ and $ZrH_{1.54}$ have higher conductivities compared to pure zirconium [11]. However, an increase in resistivity (decrease in the conductivity) with increase in the hydrogen concentration for H/Zr ratios between 1.5 to 1.6 was also reported by Bickel [12]. The measurements made by the National Bureau of Standards (NBS) showed a resistivity value of 57 μohm cm at 311 K for zirconium with 41.7 at.% hydrogen [13]. The resistivity of pure zirconium was reported as 45.6 μohm cm at 295.7 K by Roberts et al. [14] and a value of 42.6 μohm cm at 300 K by Berlincourt [15]. These data indicate that there should be a decrease in the conductivity when the hydrogen is added in low concentrations to zirconium. Therefore, the reduction in the conductivity observed in this study is in agreement with the expected behavior.

The fluctuations observed in the ultrasonic velocity with increase in the hydrogen concentration up to 1150 mg/kg (Fig. 5) are attributed to the changing morphology of the zirconium hydrides and the accompanying lattice strain variation. Table 1 gives the physical parameters of α-Zr, γ-ZrH and δ-$ZrH_{1.66}$ [16,17].

The density and modulus for both the types of hydrides are less than that of α-Zr. However, the ratio of the modulus to density (the square root of which represents velocity) may vary depending on the distribution of hydride plates in the matrix which will alter locally the density and the modulus values. It is for this reason that the modulus depends on the morphology and the nature of the hydrides formed. TEM studies clearly showed that hydrides exist in distinct morphologies-twinned γ hydrides and δ plates. At low concentrations, the twinned γ hydrides are predominant, whereas at higher concentrations, the δ plate morphology is predominant. Again, for a given hydrogen concentration, the effect due to γ hydrides is more than the effect due to large δ plate hydrides as the former influences a larger volume fraction of the matrix due to the coherency strain (Fig. 3(a)–(b)). Hence, the velocity ratio have a more pronounced effect at lower hydrogen levels than at higher hydrogen levels. With the increase in hydrogen level a transition from the twinned γ hydride regions to well developed δ hydride plates occurs resulting in a relaxation of the accumulated strain in the matrix by plastic flow. This transition may cause a change in the velocity of the ultrasonic waves differentially with respect to longitudinal and shear waves. The ratio of the velocities in the longitudinal and the shear waves is therefore affected most strongly by the morphological transition of the hydrides.

Table 1
Physical properties of α-Zr, γ-ZrH and δ-$ZrH_{1.66}$

Physical Property	α-Zr	γ-ZrH	δ-$ZrH_{1.66}$
Crystal structure	hcp	bct	fcc
Density (g/cm^3)	6.5	5.84	5.65
Elastic modulus (Gpa)	98.6	84.5	83.1

It should also be pointed out that the rates of reduction in the modulus and density with hydrogen concentration are quite different (figs. 7.30 and 7.20 of Beck and Mueller in Ref. [17]), and therefore the square root of the ratio of the modulus to density, which represents the velocity, may increase or decrease depending on the individual rates of reduction in the density and modulus at the specified concentrations. In the absence of independent availability of exact values for modulus and density at the concentrations used in this study, it was be difficult to estimate the velocity variation with the hydrogen concentration. It should also be noted that fluctuations in the difference in the transit time between the SH and SV waves with the hydrogen concentration within the range of 0 to 100 mg/kg in a coolant tube of Zr–Nb alloy was reported by Alers and Moles [10]. They attributed this fluctuation to (i) non-existence of any relationship between the hydrogen concentration and the velocities or (ii) the relationship is obscured by unknown factors. Moles and Kramer [18] also reported that, even though 14% increase in volume takes place due to precipitation of hydrides, it was not reflected in the velocity values measured conventionally. However, imaging of hydride platelets with reasonable contrast by employing defocussed scanning acoustic microscopy indicated that there is a small difference in the velocity between the matrix and the platelets [18]. The present results suggest that unless precise velocity measurements are made, it would be very difficult to detect hydrogen at low concentrations in Zr alloys.

Moles and Kramer [18] reported that the measurements made on Zr–Nb alloy with hydrides using scanning laser acoustic microscopy (SLAM) indicated only a small change in the attenuation (about 3 dB) in the case of shear waves. However, their measurements using longitudinal waves did not show measurable change in the attenuation in presence of hydrides. Similar to the observations made in the present study, it is reported that, in the case of alloys with low concentrations of hydrogen, hydrides are found to form always as needles along the (1120) in the early stages and then as plates on the [1010] in the later stages of precipitation [19]. Precipitation occurs at grain boundaries and as well as intragranularly and is accompanied by the generation of significant dislocation loops at the interface and in the α-Zr matrix [19]. The large volume change of about 18% on the formation of the γ hydrides in Zircaloy-2 results in the loss of coherency with the generation of the dislocation loops having 1/3⟨1120⟩ Burgers vectors [20,21]. It is clear from the above that, with the increase in the amount of precipitation, there is a large volume change accompanied by the formation of dislocation loops at the interface between the matrix and the hydride precipitate and within the matrix. It is also reported that the α-phase has a coarse grain structure in a single-phase region [17].

The observed increase in the attenuation with the increase in the hydrogen concentration is attributed to these changes in the microstructure which would increase the absorption (based on the Graneto and Lucke model of dislocation damping) and scattering components [22–26]. The small changes in the attenuation in the specimens with lower hydrogen concentration is associated with the formation of needle shaped hydrides and the elastic strain accommodation and the relatively small increase in the dislocation density as compared with the large platelets where the strain is accommodated by plastic relaxation. It should be pointed out that the changeover in the morphology of the hydrides from needle type to plate type coincides with the by the weak maximum in the high frequency attenuation observed at lower hydrogen concentration (Fig. 7).

It was reported from internal friction studies that the addition of hydrogen to zirconium results in three peaks at temperatures of about 228 K, 278 K and 503 K [27,28]. Increase in the hydrogen concentration was found to increase the height of the peaks. This happens even beyond the solubility of the hydrogen, indicating that not only the hydrogen in solution in the matrix, but also the hydrides which form beyond the solubility limit contributes for the internal friction. The low temperature peaks were attributed to the stress induced movement of the single hydrogen atoms. The peak at 278 K was attributed to the movement of atom pairs of hydrogen from (0,0,3/8) and (0,0,7/8) locations to (1/3,2/3,1/8) and (1/3,2/3,7/8) locations in the tetrahedral sites of the α-Zr lattice sites. The peak at 503 K was attributed to the atom exchange between lattice sites, which is influenced by the coherency stresses between the metal and the hydride precipitates. The observed increase in the attenuation coefficient with the increase in the hydrogen concentration might also result from such relaxation processes.

The possible influence of a change in the morphology of zirconium hydride due to precipitation under stress has not been addressed in this study. Stress induced reorientation of hydrides in zirconium alloys has been the subject of a large number of investigations [29–31]. However, under normal operating conditions the stresses in the pressure tube do not exceed the threshold value of stress needed for reorientation and hence the hydrides would be randomly oriented as in the present study. It is likely that the NDT parameters would be influenced by the orientation distribution of hydride precipitates and this aspect is currently being investigated.

5. Conclusion

NDT parameters like ultrasonic velocity and attenuation can be used to monitor hydrogen in zirconium alloys. The ratio of the longitudinal wave velocity to shear wave velocity with polarization direction parallel to the rolling direction would give best possible parameter to monitor hydrogen up to about 100 to 200 mg/kg. Ultrasonic velocity measurements using a 10 MHz longitudinal wave probe and a 2 MHz shear wave probe have shown that this parameter together with the independent longitudinal and shear wave velocities can be utilized in a multi-parametric correlation approach for estimation of hydrogen concentration up to 1150 mg/kg with acceptable sensitivity in different ranges of hydrogen. Ultrasonic attenuation at higher frequencies and eddy current parameters are also effective in monitoring hydrogen beyond 250 mg/kg in zirconium alloys.

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