CHARACTERISATION OF MICROSTRUCTURE IN 9 % CHROMIUM FERRITIC STEELS USING ULTRASONIC MEASUREMENTS

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ABSTRACT

Ultrasonic measurements have been performed for comprehensive microstructural characterisation in 9 % Cr ferritic steels. The variation in grain size, the effect of solution annealing temperature on microstructure developed, the variation in microstructure of the weld joints, the effect of post weld heat treatment and precipitation behaviour during thermal ageing and creep testing have been successfully evaluated by ultrasonic measurements. The ultrasonic velocity has been correlated with the hardness and the volume fractions of ferrite and martensite. For the first time, it has been shown that ultrasonic velocity measurements can be used for the identification of Ac$_1$ and Ac$_3$ critical temperatures. Ultrasonic attenuation and a new ultrasonic parameter, spectral peak ratio (SPR), have been found useful for the assessment of grain size and identification of various metallurgical features affecting the grain size such as dissolution of carbides and formation of δ-ferrite. The benefits of SPR over conventional attenuation measurements have been demonstrated. The study revealed that a single ultrasonic parameter cannot characterise the solution annealing behaviour completely, and hence ultrasonic velocity, attenuation and spectral analysis of the first backwall echo need to be used in a complementary manner for comprehensive characterisation of the solution annealing behaviour. The ultrasonic velocity measurements performed on different solution-annealed specimens have been used for the imaging of weld metal and heat affected zone, obtaining the weld profile and also for assessment of the post weld heat treatment. The study revealed that the ultrasonic velocity could also be used effectively for investigating the microstructural degradation occurring due to thermal ageing and creep. The present study has provided a better insight into the ultrasonic wave/microstructure interaction and established new methodologies for non-destructive assessment of various microstructures in 9% Chromium ferritic steels useful for practical situations.

1. INTRODUCTION

9Cr-1Mo ferritic steel and its modified versions are currently favoured structural materials for steam generator (SG) applications as they offer useful combination of high temperature mechanical properties and corrosion/oxidation resistance. The advent of new generation power plants have increased the operating steam temperatures and pressures to achieve higher efficiency and better environmental protection. This has led to the development of modified versions of 9 % Cr steels with excellent combination of creep strength and ductility. These steels include plain 9Cr-1Mo steel initially modified by the addition of strong carbide forming elements such as Nb and V (9Cr-1Mo-V-Nb designated as T91 or P91), and further modified by the addition of W (9Cr-0.5Mo-1.8W-V-Nb designated as T92 or P92) 2. The P92 steel is the same as Japanese NF616 steel 3. Modified 9Cr-1Mo steel has been selected for all the steam generator components (shell, tube and tube plate) of Indian prototype fast breeder reactor (PFBR) 4. The selection of modified 9Cr-1Mo steel for PFBR steam generator applications is based on important considerations such as high temperature mechanical properties including creep, low cycle fatigue (LCF) and creep-fatigue interaction, resistance to loss of carbon to liquid sodium and consequent
reduction in strength, resistance to wastage in case of small leaks leading to sodium-water reaction and resistance to stress corrosion cracking in sodium and water media. A major programme is underway at Indira Gandhi Centre for Atomic Research to develop and evaluate the steel indigenously. 9 % Cr steels are recommended for use in the normalised and tempered condition having fully tempered. The components of steam generators operate at high temperatures for long duration and undergo ageing induced microstructural degradation, which in turn influences the mechanical properties. Therefore, it is necessary to characterise the microstructure using non-destructive evaluation (NDE) methods for quality control of fabricated and heat treated components and for evaluating their performance during high temperature service.

At elevated temperatures, the performance of ferritic Cr-Mo steel weldment is considered to be a life limiting factor and a high percentage of failures has been reported to be weld related. In 9 % chromium steels, the microstructure of the weld joint consists of predominantly martensite in the weld metal followed by coarse gain martensite with δ-ferrite, coarse grain martensite, fine grain martensite and inter-critical structure in the heat affected zone (HAZ). In the as-welded condition, weld metal exhibits very high hardness followed by a hardness gradient in the HAZ (rapidly decreasing hardness up to the hardness level of base metal). A proper post weld heat treatment (PWHT) is carried out to temper the weld and HAZ microstructure thus enabling the release of undesirable tensile residual stresses in the weld region and also to improve the toughness and high temperature mechanical properties of the weldment. In order to ensure the desired microstructure and mechanical properties after PWHT, a suitable non-destructive evaluation based technique is essential. Ultrasonic technique offers good promise for the characterisation of microstructures. Further, many a time, it becomes necessary to identify the weld interfaces and the profile for fixing the required beam angle and scan ranges for ultrasonic inspection and for unambiguous defect location and interpretation of the ultrasonic results. Hence, identification of a suitable NDE parameter is necessary for knowing the weld profile before carrying out automated ultrasonic testing for detection and quantitative characterisation of defects in the weldments.

The velocity of ultrasonic wave is influenced by the elastic moduli and the density of the material, which in turn are mainly governed by the amount of various phases present and the damage to the material. The ultrasonic attenuation, which is the sum of absorption and scattering, is mainly dependent upon the damping capacity and the scattering from the grain boundary, wedge cracks (if present) etc. in the material. Both ultrasonic velocity and attenuation have been applied for the characterisation of microstructures and for the evaluation of material properties to ensure the absence of unacceptable discontinuities and presence of desired microstructure with acceptable properties during fabrication and heat treatments. Ultrasonic in-service inspection is carried out to detect any unacceptable degradation in microstructure and formation and extension of defects in a component during service. Ultrasonic parameters, such as velocity and attenuation, have been correlated with the microstructural features evolved during heat treatments in ferritic steels, superalloys, aluminum alloys and many other materials. This technique has also been used successfully for the evaluation of yield strength, fracture toughness, grain size and volume fraction of second phases. Dubois et al. have used on-line laser based ultrasonic attenuation measurement for monitoring phase transformation in steels. They observed that the ultrasonic attenuation increases initially with increase in temperature followed by a decrease in the intercritical region (between Ac₃ to Ac₁). Above Ac₃, the attenuation increases sharply with temperature due to increase in grain size. Such a variation in the ultrasonic attenuation with increasing solution-annealing temperature indicated that a single ultrasonic parameter might not be sufficient to characterise the several microstructural features in ferritic steels. In addition to attenuation, other parameters are needed to determine the amount of different phases in intercritical region (between Ac₁ to Ac₃) and to identify the various critical temperatures more unambiguously. It was realised that a multi-parameter approach can be more appropriate in such conditions.

In Nickel base superalloy Inconel 625, ultrasonic velocity measurements have been correlated with precipitation of carbides and various intermetallic phases such as γ”, Niₓ(Cr,Mo) and δ. Linear correlations have been established between ultrasonic
velocity and yield stress for precipitation of various phases. It has been observed that the dissolution of Ni$_2$(Cr, Mo) phase and precipitation of δ-phase have more effect on ultrasonic velocity as compared to that on yield stress, whereas dissolution and precipitation of γ” has more influence on yield stress than on ultrasonic velocity. In order to explain the influence of coherent precipitates on ultrasonic velocity, Fouquet et al. have pointed out that in presence of precipitates, the material can be considered as a composite of three components - matrix, matrix-precipitate interface and precipitate. Since ultrasonic velocity is dependent upon the Young’s modulus and density of the material, it is dependent upon the Young’s modulus and density of these three components. In incoherent matrix-precipitate interface, the loss of coherency leads to decrease in the Young’s modulus of the interface and therefore reduces the effective ultrasonic velocity of the material. The formation of coherent precipitates leads to an increase in the ultrasonic velocity and their coarsening or dissolution results in a decrease in the velocity.

Ultrasonic velocity and attenuation measurements in austenitic stainless steel weldment indicated that the attenuation measurement is useful for studying the variation in grain size across the weldment. It has also been pointed out that in austenitic stainless steel weldments, ultrasonic velocity is mainly affected by the grain orientation and not by the microstructure, i.e. the amount of ferrite in the weld metal.

Ultrasonic spectroscopy is a further development of the pulse-echo technique, which uses broadband ultrasound and analyses the spectra of the echo pulses. In an analogy with light, the specimen is insonified with white sound. The defects or microstructure alter the ‘colour’ of the wave travelled through the material and the change in the colour of the sound gives the information about the material. Ultrasonic spectroscopy has been used for the characterisation of defects and microstructural features. It has been reported that, in stainless steel, as the grain size increases, the peak frequency and the full width at half maximum of the autopower spectrum of the first back wall echo decrease. The shift in the peak frequency has also been used for evaluation of structural variations induced by tensile deformation in SUS304 stainless steel. The spectral peak frequency in SUS304 steel was found to increase with increase in the tensile elongation, which was attributed to the formation and growth of martensite structures due to tensile deformation resulting in smaller crystalline grains, thus reducing the attenuation due to ultrasonic scattering. Gericke has illustrated the use of ultrasonic spectroscopy for grain size determination. The frequency characteristic of the transducer was found to show two well defined humps centered at frequencies of about 3.5 and 6 MHz, respectively. The corresponding frequency responses of steels with varying grain sizes showed that, as the mean grain size increased, the ratio of the heights of the two frequency humps changed, the higher frequency being preferentially attenuated by coarser grained material.

The present paper deals with the application of various ultrasonic parameters for comprehensive characterisation of microstructures in 9 % Cr ferritic steels. An attempt has been made to correlate the various ultrasonic parameters with microstructure in terms of the variations in grain size and microstructural constituents with solution annealing temperature ranging from α-ferrite (1073 K) to γ + δ phase regions (1673 K), imaging of various regions of weld joint such as weld metal and HAZ and assessment of post weld heat treatment in modified 9Cr-1Mo steel. The paper also presents the variation in ultrasonic velocity with the degradation occurring due to thermal ageing and creep in 9Cr-1Mo ferritic steel.

2. EXPERIMENTAL

2.1 Test Specimens, Microstructural examinations and Hardness Testing

The chemical compositions of both the plain and the modified 9Cr-1Mo steels investigated in the present study are given in Table I. For investigating the solution annealing behaviour of modified 9Cr-1Mo steel, various specimens of 12 x 12 x 60 mm size were soaked for a duration of 5 min. at different temperatures ranging from α-ferrite (1073 K) to γ + δ ferrite phase region (1623 K) followed by oil quenching. These treatments were intended to obtain different microstructures and grain sizes in the specimens simulating the different regions in the heat affected zone of the weldment. Also, the range of
temperatures of these treatments included the solution annealing (austenitising) treatment generally given to the ferritic steels. Surface grinding of these samples was carried out to obtain the specimens of 10.5 ± 0.2 mm thickness with plane parallelism to an accuracy of better than ± 5 μm. Metallographic examination was carried out to reveal the microstructures in different specimens prepared by usual metallographic technique and by etching with Villella’s reagent. After completing all the ultrasonic measurements, in order to reveal the prior austenite grains, these samples were given further heat treatment at 1033 K for 1 h and aged for 2000 h at 923 K. The linear intercept method as per ASTM standard E112-88 was used to find out the average grain size. The hardness of these specimens was also measured using Vicker’s hardness tester at a test load of 10 kg.

For imaging of weldment and assessment of post weld heat treatment in modified 9Cr-1Mo steel, weld joint specimens of 60 mm length and 12 mm width were machined from 12 mm thick weld pad in the as-welded condition. Some of these specimens were given a PWHT of 1033 K for 1 h. Surface grinding of these specimens was carried out to make the surfaces plane parallel with an accuracy better than ± 5 μm. Metallographic examinations were carried out to reveal the various regions of the weld joints. The Vicker’s hardness measurements were carried out in the weld metal and HAZ at distances of regular intervals from the weld interface.

In order to investigate the precipitation behaviour and microstructural degradation in 9Cr-1Mo steel, the as-received quenched and tempered (Q + T: 1223 K/5 h water quenched followed by 1023 K/8 h air cooled) specimen blanks of 12 mm diameter and 60 mm length were machined in the thickness direction from a 300 mm thick forging. These blanks were thermally aged (TA) at 793 and 873 K for durations ranging from 10 to 5000 h. For long ageing times, the samples were taken from the unstressed shoulder regions of the creep specimens tested at 793 and 873 K. For investigating the influence of creep deformation, specimens were obtained from the gauge length region of creep specimens tested at 873 K for rupture lives ranging from 11 to 15175 h. The details of creep tests are described elsewhere. The Vicker’s microhardness measurements were carried out at 0.490 N load on all the specimens. For ultrasonic measurements, surface grinding of these specimens was carried out to obtain a constant thickness of 5 mm and plane parallelism to an accuracy of ± 2 mm. Transmission electron microscopic (TEM) studies were performed on the carbon replicas using analytical transmission electron microscope (ATEM), Philips CM 200. The energy dispersive analytical X-ray (EDAX) spectra and selected area diffraction (SAD) patterns were taken for the identification of various precipitates.

2.2 Ultrasonic Measurements

The experimental setup used for the ultrasonic measurements is shown in Fig.1. 100 MHz broad band pulser-receiver (M/s. Accutron, USA) and 500 MHz digitizing oscilloscope (Tektronix TDS524) were used for carrying out the ultrasonic measurements. For these measurements, the rf signals were digitized
at 500 MHz and the gated backwall echoes (1024 ns duration) from the oscilloscope were transferred to the personal computer with the help of GPIB interfacing and LabVIEW software. Ultrasonic velocities were measured using 5 MHz shear and 15 MHz longitudinal beam transducers. Cross correlation technique has been used for precise velocity measurements. The accuracy in time of flight measurement was better than ± 1 ns. The scatter in the measurement of longitudinal and shear wave velocities is ± 3 m/s and ± 2 m/s, respectively. Ultrasonic attenuation measurements were made using 15 MHz longitudinal wave transducer with fixed perspex delay line of 10 mm. Spectral analysis has been carried out on the rf signal corresponding to the first backwall echo obtained by 20 MHz longitudinal beam transducer. Two distinct peaks have been found in the autopower spectrum of the first backwall echoes for all the specimens, when 20 MHz transducer is used. The ratio of these two peaks has been used as an ultrasonic parameter. Specific softwares were developed for automatic on-line measurement of precise time of flight and attenuation of ultrasonic waves and also for the peak ratio measurements from the autopower spectrum.

Ultrasonic velocity measurements have been carried out across the weld-line in two perpendicular sections (Fig. 2) using 15 MHz longitudinal wave transducer of 4 mm diameter. To study the feasibility of the detection of various regions in the weldment, ultrasonic velocity measurements have been carried out across the weld line at three different depths, such as near crown, middle and near root as shown in Fig. 2a (Scan I, Scan II and Scan III respectively), as ultrasonic waves always see the similar microstructure throughout the specimen in the wave propagation direction. But during practical conditions, this surface would not be accessible for the measurements to be carried out. The only accessible surface would be the top surface and hence to evaluate the weld profile under practical conditions, ultrasonic velocity measurements have also been carried out from the top surface (Fig. 2b). The plot of time of flight (TOF) against scanning distance across the weld is plotted on-line using a PC based TOF measurement system, US-TOF, supplied by M/s. Fraunhofer Institute for NDT (IZFP), Germany. At the scan speed used in the present study, three data points were recorded for each millimetre length of the specimen. The scatter in the ultrasonic velocity measurements is less than ± 2.5 m/s.

3. RESULTS AND DISCUSSION

3.1 Solution Annealing Behaviour

Typical photomicrographs for the specimens with various microstructures and grain sizes obtained by employing solution annealing temperatures ranging from α-ferrite region to γ + δ region are shown in Fig. 3. It can be seen that in the specimens heat treated at temperatures below lower critical temperature (Ac1 ~ 1100 K), the microstructure is composed of only ferrite with laths and carbides (tempered martensite) (Fig. 3a). With increase in
solution annealing temperature above $A_{C_1}$, partial austenitisation during soaking resulted in the microstructure with ferrite and freshly formed martensite (Fig. 3b). Further increase in temperature in the ferrite + austenite region (between $A_{C_1}$ and $A_{C_3}$) provided higher amount of austenite during soaking and higher volume fraction of martensite after quenching. The amount of martensite increased till the upper critical temperature ($A_{C_3} \sim 1180$ K) and only lath martensite with fine prior austenitic grains was observed at 1223 K (Fig. 3c). With increase in temperature above $A_{C_3}$ a marginal increase in grain size was observed up to $\sim 1373$ K (Fig. 3d). Beyond this, a rapid increase in the prior austenitic grain size was observed (Fig. 3e). The marginal increase in the grain size can be attributed to the presence of fine vanadium and niobium carbides ($V_4C_3$ and NbC), which restrict the grain growth.

Fig. 3: Microstructures of the specimens solution annealed at (a) 1073 K (b) 1148 K (c) 1273 K (d) 1373 K (e) 1473 K and (f) 1623 K for 5 Minutes, followed by oil quenching.
process during austenitisation in the temperature range 1223-1373 K. The dissolution of these carbides at temperatures higher than 1373 K results in a rapid growth in the prior austenite grain size. At about 1490 K (just above $A_c_3$), $\delta$-ferrite starts forming at the prior austenite grain boundaries, which restricts the grain growth resulting in relatively finer prior austenitic grain size and duplex martensite + $\delta$-ferrite microstructure (Fig. 3f).

The variations in ultrasonic longitudinal and shear wave velocities with soaking temperature are shown in Figs. 4(a) and (b), respectively. The variation in hardness with soaking temperature is also shown in these figures. In the specimens heat treated at temperatures below $A_c_1$ i.e., 1100 K, both the ultrasonic longitudinal ($V_l = 5995$ m/s) and shear ($V_s = 3310$ m/s) wave velocities were the highest. With increase in temperature above $A_c_1$ (1100 K), a rapid decrease in both the velocities till $A_c_3$ temperature i.e., = 1180 K was observed. Like sharp changes in the ultrasonic velocities, the hardness value increased rapidly with increasing temperature between $A_c_1$ and $A_c_3$ temperature. The sharp decrease in the ultrasonic velocities and a rapid increase in the hardness in the intercritical region are attributed to the increase in the amount of martensite with increase in the soaking temperature. Above $A_c_3$ temperature i.e., 1180 K, the velocities remained almost constant ($V_l = 5950$ m/s and $V_s = 3270$ m/s) up to about 1323 K. Beyond this, the ultrasonic velocities were found to decrease marginally. The decrease in the ultrasonic velocities above 1323 K can be attributed to the increase in the grain size. The difference in the longitudinal wave velocity between ferrite and martensite is 0.75 % (Fig. 4a), whereas this difference for shear wave velocity is observed as 1.21 % (Fig. 4b). These observations suggest that the shear wave velocity could be more reliable than longitudinal wave velocity for the same accuracy in the time of flight measurement.

The variations in the ultrasonic attenuation and grain size with soaking temperature are shown in Fig. 5. It can be seen that both the ultrasonic attenuation and grain size exhibited similar trend in their variations with soaking temperature. The specimens heat treated below $A_c_1$ temperature exhibited nearly a constant attenuation of $= 0.25$ dB/mm. With increasing temperature above $A_c_1$, a decrease in the attenuation...
was observed and this decrease in attenuation is ascribed to increase in the amount of martensite (having low attenuation) with increasing temperature. Ultrasonic attenuation was found to be minimum (= 0.07 dB/mm) in the specimens quenched from the temperatures just above the $A_C_1$, i.e., in which the prior austenite grain size was also found to be minimum. Attenuation values exhibited a marginal increase in the specimens heat treated above 1323 K followed by a rapid increase at temperatures above 1373 K. Both marginal and rapid increase in attenuation are attributed to the respective increase in the grain size. At temperatures above ~ 1473 K, the ultrasonic attenuation again decreased with increasing temperature due to decrease in grain size as a consequence of δ-ferrite formation.

The variation in the attenuation with the soaking temperature and grain size can be explained with the help of the attenuation theory. Ultrasonic waves in a polycrystalline material are attenuated by structural boundaries and grains. The total attenuation coefficient $\alpha$ may be expressed as

$$\alpha = \alpha_s + \alpha_a,$$

where $\alpha_a$ is related to the losses due to thermoelasticity, magnetic hysteresis and dislocation damping, i.e., ultrasonic absorption, and $\alpha_s$ takes into account the losses due to the scattering of the ultrasonic waves. The attenuation is dominated by Rayleigh scattering, when the wavelength of the ultrasonic waves ($\lambda$) is greater than the grain size $d$ (the condition that is valid in this study i.e., $\lambda = 400 \mu m$ and the values of $d$ are in the range 20-130 $\mu m$). The ultrasonic attenuation coefficient due to the Rayleigh scattering ($\alpha_s$) in polycrystalline materials can be expressed as

$$\alpha_s = S \cdot d^3 \cdot f^4,$$

where $S$ is a scattering factor that depends on the elastic properties of the material (including sound velocity), $d$ is the average grain size in the specimen and $f$ is the frequency.

It can be seen in Fig. 5 that, for the same prior austenite grain size, the attenuation is found to depend on microstructure. For example, the line $d_A d_B d_C$ represents constant grain size value of ~ 50 $\mu$m obtained at three soaking temperatures of $T_A = 1096 K$, $T_B = 1382 K$ and $T_C = 1625 K$. The corresponding microstructures for these conditions are (I) ferrite, (II) martensite and (III) martensite with δ-ferrite, respectively. The attenuation values corresponding to these soaking temperatures are $\alpha_A$ (0.22 dB/mm), $\alpha_B$ (0.1 dB/mm) and $\alpha_C$ (0.337 dB/mm), respectively. Papadakis reported that the scattering is proportional to prior austenite grain volume and is much weaker in martensite than in pearlite and its associated phases i.e., ferrite and cementite. Our results are in agreement with this, i.e., the martensite structure exhibits the lowest attenuation. Even in the presence of martensite, the specimen with martensite and δ-ferrite exhibits higher attenuation than that in the specimen with ferrite and carbide. This is attributed to the presence of coarser laths in the ferrite (Fig. 3a), which makes the ferrite more isotropic than δ-ferrite. Besides this, the presence of two phases makes the sample with martensite and δ-ferrite most anisotropic and hence it exhibits the highest attenuation for the same grain size.

Figures 6a and 6b show typically the changes in the first backwall echoes and the autopower spectra with the change in the grain size in the specimens with martensitic structure. Two distinct peaks centered around 7.0 MHz and 17.5 MHz have been found in the autopower spectrum of the first backwall echoes in all the specimens, when 20 MHz transducer is used. The relative heights of the two peaks change with the change in the grain size. The spectral peak ratio (SPR) is found to increase with increase in grain size. Figure 7 shows the variation in SPR with soaking temperature exhibiting a similar behaviour as shown by ultrasonic attenuation in the variation of attenuation with soaking temperature (Fig. 5). The SPR decreased with increase in soaking temperature in the intercritical region (1100 to 1180 K) because of the decrease in the grain size. Beyond this, SPR increased with increase in the soaking temperature due to increase in grain size. The SPR increased rapidly above 1373 K due to the sharp increase in grain size. Like attenuation, the SPR decreased with increase in soaking temperature above 1473 K due to decrease in grain size.

The change in SPR with grain size can be explained on the basis of ultrasonic attenuation theory (eq. 2) and the spectral analysis of the ultrasonic waves.
When the ultrasonic wave is attenuated in the material, it can be assumed that the oscillatory amplitude decays exponentially as the propagation length of the ultrasonic wave increases. If ultrasonic wave is considered as the spectral distribution, which has an oscillatory pulse of one or two cycles with an exponential decay envelope, the amplitude of the ultrasonic wave $g(t)$ is expressed as

$$g(t) = A \exp(-\alpha t) \sin(2\pi f_0 t - \phi) \quad (\phi/f_0 < t < (2 + \phi)/f_0)$$

(3)

where $\alpha$ is the attenuation coefficient, $t$ is time, $f_0$ is the carrier frequency, $\phi$ is phase and $A$ is the maximum amplitude of the pulse wave. The spectrum power distribution $|G(f)|$ is derived by Fourier transform

$$|G(f)|^2 = \frac{1 + \exp\left(-\frac{2\alpha f_0}{f_0}\right)}{\alpha_f^2 + 4\pi^2 (f - f_0)^2}$$

(4)

From the ultrasonic scattering model (eq. 2), the attenuation increases with increase in grain size and frequency. As the grain size increases, $\alpha$ and also the frequency dependency of $\alpha$ increase. Therefore, as the prior austenitic grain size increases, more attenuation takes place (Fig. 6) and the ratio of the two peak heights in the autopower spectrum changes (Fig. 8b) due to the increased attenuation of higher frequency components.

Figures 8a and 8b show the effect of the coupling condition on the first backwall echo and its autopower spectrum, respectively, for the specimen solution annealed at 1573 K. It can be seen that when the coupling condition was not good, the amplitude of the first backwall echo and its autopower spectrum decreases. In the autopower spectrum, the amplitude of both the peaks decreased maintaining the peak ratio almost constant and therefore the peak ratio remains independent of variations in the couplant condition. Similar behaviour has been found in all the other specimens used in this study. The independence of ultrasonic spectral parameter on couplant condition is in agreement with our earlier work on type 316 austenitic stainless steel. These
observations clearly indicate that if these spectral parameters are used for grain size measurement then the error involved in the measurements due to variation in the couplant condition can be minimised for on-line and/or when a large number of measurements are to be made by moving the specimen/transducer. This has an important practical significance for gain size measurement by ultrasonic measurements. In addition to this, since only first back wall echo is required for the grain size measurements, this approach can be used for grain size measurements even in thicker and highly attenuating materials, where obtaining multiple back wall echoes with good signal to noise ratio is very difficult.

3.2 Weld Imaging and Evaluation of PWHT

The microstructures of the ferritic steel weldment consist of weld metal and HAZ microstructures apart from that of base metal. The microstructures in the HAZ are similar to that of the base metal specimens solution annealed at different temperatures ranging from $\alpha$-ferrite (1073 K) to $\gamma + \delta$-ferrite phase region (1673 K), which have been characterised and discussed in the previous section. In this section, an attempt has been made to characterise the different regimes of the weldment using the information obtained on solution annealing material. The step-wise investigation involved feasibility study, imaging of weldment and assessment of PWHT. The practical significance of this work is discussed.

3.2.1 Feasibility Study

Figure 9 shows the variation in ultrasonic velocity with scanning distance across the weld-line at three different depths, such as near crown, center and near root (indicated in Fig. 2a and Fig. 9 as Scan I, Scan II and Scan III) of the weldment in as-welded condition. Ultrasonic velocity is found to be constant ($\sim 6005$ m/s) in the parent base metal and it decreases as the HAZ is approached. Ultrasonic velocity decreases rapidly up to the weld interface followed by a gradual decrease to a minimum

![Fig. 9](image_url)

Fig. 9: Variation in ultrasonic velocity with scanning distance across the weld-line at three different depths. (WM – Weld metal, HAZ – Heat affected zone).
(~ 5925 m/s) at the centre of the weld and an increase in the other side of weld in a symmetric manner. The difference in the width of the weld at different depths can be seen clearly in the velocity plot. The decrease in ultrasonic velocity in the weld metal is attributed to the presence of martensitic structure with lower elastic modulus compared to that of the ferrite. In addition to this, the presence of residual stress would also have some contribution on the change in the ultrasonic velocity. However, the expected influence of residual stress on ultrasonic velocity is much less (~ 20 m/s) as compared to the large change in the velocity (80 m/s) observed in the present investigation. Further, the present study has been carried out on the specimens cut from the weld-pad, and hence majority of the residual stress would have got released. Even after the PWHT (discussed in next paragraph), considerable difference in the velocity exists between the weld and the parent metal (~ 30 m/s), which further indicates that in 9Cr-1Mo ferritic steel weldments, the variation in ultrasonic velocity around the weld is predominantly governed by microstructure rather than by residual stress. Ultrasonic velocity has been found to be minimum (~ 5925 m/s) at the weld centre near the crown (Scan I in Fig. 2a). The minimum velocity at the weld centre increases towards the root of the weld (~ 5941 m/s in Scan III in Fig. 2a). In the weld metal, the higher ultrasonic velocity at the root compared to that in the crown is attributed to marginal tempering of the martensitic structure in the root region due to thermal cycle during the subsequent weld passes.

The variations in ultrasonic velocity and hardness with scanning distance across the weld-line at the centre of the weld (Scan II in Fig. 2a) in the as-welded and after PWHT conditions are shown in Fig. 10. After PWHT, ultrasonic velocity increases from 5930 m/s to 5970 m/s in the weld metal, showing the possibility for the assessment of PWHT and its adequacy using ultrasonic velocity measurements. Even though ultrasonic velocity increases after PWHT, it does not reach the velocity of that of the parent base metal. This can be ascribed to the fact that the heat treatment for 1 h at 1033 K given on the weldment is not sufficient for complete tempering of the microstructure. This is further supported by the microhardness measurements made in the as-welded and tempered specimens (Fig. 10). In the as-welded specimen, the maximum hardness was found at the centre of the weld (550 VHN). The hardness of the parent metal is 225 VHN. Even after PWHT, hardness at the centre of the weld is 300 VHN, i.e. 75 VHN more than the parent metal (Fig. 10). In the parent metal, a slight decrease in ultrasonic velocity is observed after PWHT. It can be seen in Fig. 4a that the minimum ultrasonic velocity in the HAZ could be 5950 m/s only and hence this value can be used for identifying the boundary between the HAZ and the weld metal. In the weld metal, ultrasonic velocity is further lower due to martensitic structure with high dislocation density. This is also supported by the hardness measurements, which shows that the maximum hardness in the specimens simulating the weld microstructure is 450 VHN only (Fig. 4) as compared to 500 VHN at the centre of weld metal in the as-welded specimen (Fig. 10).

### 3.2.2 Ultrasonic Velocity Based Imaging of Weld Profile

Ultrasonic velocity measurements carried out across the weld line at three different depths (near crown, centre and near root as shown in Fig. 2a) of the weldment in as-welded condition have been used to image the weld structure. Figure 11 shows the ultrasonic velocity based image of the weldment in as-welded condition. Ultrasonic velocity in the parent metal has been taken as greater than 6000 m/s. In the intercritical region, it has been taken as less than 6000 m/s but greater than 5950 m/s, as discussed earlier in the case of simulated weld specimens. The weld metal is defined as having velocities less than 5925 m/s.
The ultrasonic velocity image of the weldment is found to almost replicate the actual photograph of the macro-etched weldment, as shown in Fig. 12. The slight difference in the weld profile (near crown and root) is because the velocity scan nearest to the crown and root could be done only 2 mm away from the edges, as the diameter of the transducer used was 4 mm and hence the weld profile details very close to the crown and root could not be revealed.

3.2.3 Study with Practical Significance

The variation in ultrasonic velocity with scanning distance across the weld-line in as-welded and PWHT conditions are shown in Fig. 13. The measurements were performed from the top surface of the weldment as depicted in Fig. 2b (this surface would be accessible for measurements on any weldment of a component). As discussed earlier, with increased fraction of martensitic phase in the direction of wave propagation, ultrasonic velocity should decrease. This can be seen in Fig. 13 that the ultrasonic velocity decreases as the measurement location is slowly moved from parent metal region to centre of the weld i.e., with increased fraction of weld metal (martensitic structure) in the direction of wave propagation.

Comparison of ultrasonic velocity plot in Fig. 13 with the photomacrograph of the weldment (Fig. 12) reveals that the ultrasonic velocity profile almost replicates the weld profile and hence such measurements can be employed effectively to depict the weld profile in the ferritic steel weldments. Further, it can be seen clearly in Fig. 13 that the ultrasonic velocity increases (5910 m/s to 5955 m/s) in the weld region after PWHT due to the tempering of the martensitic structure. This suggests that the PWHT can be monitored on actual weldment using ultrasonic velocity measurements. This study has the practical significance even during the repair welding and subsequent PWHT, as the weld profile can be determined and also the adequacy of the PWHT with proper microstructure can be assessed non-destructively. This study has been carried out on the specimens of 12 mm thickness with plane parallelism of ± 5 μm. For practical applications of the determination of weld profile, the weld bead should be flushed properly and the thickness variations should be as less as possible. Whereas, for the assessment of PWHT (where the difference in velocity in as-welded and PWHT conditions is measured), the variation in thickness can be taken care of by carrying out the measurements at the same location before and after PWHT.

3.3 Microstructural Degradation during Thermal Ageing and Creep

The variation in ultrasonic velocity in 9Cr-1Mo steel with ageing time at 793 and 873 K and creep exposure at 873 K are shown in Fig. 14. Based on the variation in ultrasonic velocity with exposure time, the whole ageing period has been divided in four different regimes; regime-I, regime-II, regime-III and regime-IV for better clarity in the discussion. In Fig. 14, the various regimes are shown for thermal ageing at 873 K. Regime-I consists of increase in ultrasonic velocity with ageing time at shorter durations (up to 10 h at 873 K and 100 h at 793 K). At intermediate durations, ultrasonic velocity decreased with ageing time (up to 1000 h at 873 K and 2200 h at 793 K) in regime-II followed by increase in ultrasonic velocity (up to 5000 h at 873 K and 10850 h at 793 K) in regime-III. At long durations, continuous decrease in ultrasonic velocity with ageing time (beyond 5000 h at 873 K) was observed in regime-IV. The variation in hardness with ageing time also
exhibited four different regimes similar to that of ultrasonic velocity (Fig. 15).

TEM micrographs for specimens thermally aged for different durations are shown in Fig. 16. The selected area diffraction (SAD) patterns for the indicated precipitates are shown in insets. Q+T as-received specimen exhibited presence of precipitates at prior austenite grain and lath boundaries and in the intralath matrix regions. Figure 16a shows globular precipitates in the interlath and acicular precipitates in the intralath regions. The analysis of SAD pattern and EDX spectrum revealed that the intralath precipitates (shown by a needle in Fig. 16a) are mainly of Cr$_2$X type with hexagonal close packed structure. This chromium rich precipitate has been identified as Cr$_2$N. The globular precipitates at the grain and lath boundaries are mainly of M$_{23}$C$_6$ [(Cr Fe)$_{23}$C$_6$] type with face centred cubic structure.

The initial increase in the ultrasonic velocity in regime-I can be ascribed to the further formation of fine Cr$_2$N precipitates in the intralath regions at short durations of ageing compared to that in Q+T condition (Fig. 16 a and b). This increased precipitation at short duration of ageing results from the ageing of specimens at lower temperatures of 793 and 873 K than the 1023 K used for tempering treatment. The formation of fine precipitates in specimen aged at 873 K saturates early compared to
that in specimen aged at 793 K resulting in lower duration of regime-I at 873 K. The gradual decrease in ultrasonic velocity in regime-II is attributed to the coarsening and consequent decrease in number density of Cr$_2$N precipitates. The reversal in trend in the velocity in regime-III is attributed to the occurrence of secondary precipitation in the steel (Fig. 16c), which also counter acts the effect of coarsening of Cr$_2$N precipitates. The secondary precipitates (as indicated by needle in Fig. 16c) are identified as Fe$_2$Mo (Laves phase) with hexagonal structure. With further ageing in this regime, both ultrasonic velocity and hardness increase due to the increased amount of Fe$_2$Mo precipitation (Fig. 16d). The sharp reduction in the ultrasonic velocity and hardness observed in regime-IV at 873 K can be ascribed to excessive ageing at long durations resulting in coarsening of precipitates (Fig. 16e). The effect of ageing temperature on precipitation kinetics is exhibited by shifts in the regimes of ultrasonic velocity to lower ageing time with increasing temperature. Further, the absence of regime-IV at lower ageing temperature of 793 K could have resulted from the fact that this regime of rapid decrease in the velocity at long durations may occur at higher ageing time at 793 K than that used in this investigation. The possibility of the use of ultrasonic velocity to characterize the thermal ageing behaviour in 9Cr-1Mo ferritic steel was also attempted in our centre a few years ago.

The variation in ultrasonic velocity with creep exposure at 873 K also exhibited a behaviour similar to that observed for thermally aged specimens (Fig. 14) and suggested that the creep damage in 9Cr-1Mo ferritic steel is mainly due to the microstructural degradation, and not due to cavitation damage which is expected to decrease the velocity continuously with increasing amount of damage. This is in agreement with the detailed investigation performed on the creep behaviour of 9Cr-1Mo steel. Analysis of creep data in terms of creep rate-rupture life relationships indicated high creep damage tolerance factor and metallographic investigation exhibited absence of typical creep damage in the form of wedge cracks and r-type cavities in the steel. The steel retained its high creep ductility and the failure mode remained transgranular characterised by dimples resulting from microvoid coalescence. All these observations suggested that 9Cr-1Mo steel is not prone to the typical creep damage and the creep failure results from microstructural degradation. The creep enhanced precipitation can be seen by the shift in the regimes to lower time as well as higher ultrasonic velocity in creep exposed condition compared to that in thermal ageing. The study demonstrated that the ultrasonic velocity is sensitive to microstructure of the steel and the use of this technique can be extended to assess the microstructural degradation and its influence on the state of health of a component during service, by judicious selection of time periods between any two consecutive inspections. Further, the variation in the thickness of the actual component can be taken care of by carrying out the ultrasonic velocity measurements at the same place during service or by using the ratio of ultrasonic longitudinal and shear wave velocities, if feasible. This ratio is same as the transit time ratio for these two wave modes and hence the thickness is not required to be known at the measurement location.

4. CONCLUSIONS

The study revealed that ultrasonic parameters can be successfully used for comprehensive characterisation of the solution annealing behaviour of ferritic steels. The ultrasonic velocity can be used to determine $\text{Ac}_1$ and $\text{Ac}_3$ temperatures, hardness and the volume fraction of martensite and ferrite in the specimens heat treated between $\text{Ac}_1$ and $\text{Ac}_3$ temperatures. Ultrasonic attenuation and a new ultrasonic parameter, spectral peak ratio (SPR), are found to be sensitive to grain size and these can be used to determine grain size, temperature to the onset of dissolution of NbC and V$_4$C$_2$ and the formation of $\delta$-ferrite i.e., the $\text{Ac}_4$ temperature. The independence of spectral peak ratio on couplant condition has an important practical significance, that the error involved for the on-line grain size measurement by ultrasonic measurements can be reduced by exploiting this parameter. Ultrasonic velocity can also be used for the imaging of weld profile in ferritic steels and for the evaluation of the adequacy of PWHT. These results suggest that the ultrasonic parameters can be used effectively as a quality control tool for the initial and post fabrication heat treatment of the components. Further, the ultrasonic velocity has been found to be very sensitive to the microstructural degradation in 9Cr-1Mo ferritic steel occurring due to thermal ageing and creep exposures and therefore, it has potential for life assessment applications of these steels.
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REFERENCES