Comprehensive Characterization of Ageing Behaviour in M250 Maraging Steel using Multi-NDE Techniques

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Abstract: Nondestructive measurements have been carried out in M250 grade maraging steel specimens subjected to solution annealing at 1093 K for 1 h followed by ageing at 755 K for various durations in the range of 0.25 to 100 h. Different NDE techniques such as ultrasonic velocity, magnetic barkhausen emission (MBE), positron annihilation spectroscopy (PAS), eddy current and X-ray diffraction (XRD) have been employed for comprehensive characterization of microstructural features evolved consequent to aging treatment.

1.0 Introduction

M250 grade maraging steel, by virtue of its excellent mechanical properties i.e. ultra-high yield strength combined with good fracture toughness [1], is the preferred structural material for critical applications in advanced technologies. In addition to the above mentioned properties, its high strength to weight ratio, good weldability and easy machinability in the solution annealed condition and dimensional stability during aging make this material as an ideal choice for critical rocket motor casing applications in aerospace industries. The ageing behaviour of the maraging steels has been extensively studied [2-12]. The strength in aged condition is derived from the fine and coherent intermetallic precipitates, whereas low carbon martensitic structure provides the high fracture toughness. Over-aging results in coarsening of the intermetallic precipitates in addition to the reversion of martensite to austenite. These two processes that occur due to overaging affects both tensile and fracture properties of these steels. Hence, characterization of microstructure plays an important role in qualification of fabricated components for service. Particularly, non destructive evaluation (NDE) techniques are most sought as they provide fast and reliable means of characterizing microstructures of actual components.

Various NDE techniques such as ultrasonic, magnetic Barkhausen emission (MBE), positron annihilation spectroscopy, X-ray diffraction and eddy current testing have shown good promise in characterizing the microstructure in various alloy systems. Though considerable work has been done on other alloy systems, so far no systematic study has been carried out for characterization of aging behaviour in maraging steels, which leads to complex microstructure consequent to aging. In view of this, in the present study, an attempt has been made to unfold the effects of various microstructural features of maraging steel that evolve during aging treatment, on different NDE parameters and in-turn to develop nondestructive methodology for comprehensive assessment of ageing behavior of M250 grade maraging steel.

2.0 Experimental

The chemical composition (wt %) of the maraging (M250) steel used in this study is as follows: 17.89 Ni, 8.16 Co, 4.88 Mo, 0.43 Ti, 0.05 Mn, 0.05 Cr, 0.05 Si, 0.05 Cu, 0.096 Al, 0.003 C, Bal. Fe. A plate of maraging steel (M250) was solution annealed at 1093 K for 1 h followed by air cooling. The specimens of approximate dimensions 30x25x7 mm extracted from the solution annealed plates, were encapsulated in quartz tubes under vacuum and aged at 755 K for different durations of 0.25, 1, 3, 10, 30, 40, 70 and 100 h followed by water quenching. The same sets of specimens were used for all the NDE studies using multiple techniques. The Vicker's hardness measurements were carried out on the specimens at 10 kg load. The average value of five hardness measurements has been reported for each specimen. The maximum scatter in the hardness measurements was found to be less than ± 5 VHN.

For ultrasonic measurements, surface grinding of the heat treated specimens was carried out to obtain a constant thickness of 7 mm and plane parallelism to an accuracy of better than ± 2 μm. Ultrasonic velocity was measured at room temperature using 15 MHz longitudinal wave and 5 MHz shear wave transducers. A 100 MHz broad band pulser-receiver (M/s. Accutron, USA) and 500 MHz digitizing oscilloscope (M/s. Lecroy, USA) were used for carrying out the ultrasonic measurements. Cross correlation technique has been used for precise velocity measurements [13]. For the velocity measurements, the ultrasonic signals were digitized at 500 MHz and the gated backwall echoes from the oscilloscope were transferred to a personal computer with the help of GPIB interface and specific software developed in LabVIEW. The accuracy in time of flight measurements was better than 1 ns and the maximum scatter in the ultrasonic velocity was found to be less than ± 2.5 m/s.

Magnetic Barkhausen emission measurements were performed using encircling pick up coil (5000 turns) and magnetizing the sample at a frequency of 66 mHz. A U-shaped electromagnet assembly was used to magnetize the maraging steel specimen (30mmx25mmx7mm) fixed between two conical pole pieces of an electromagnet. The maximum field was set to 1500 Oes for complete magnetic saturation of the specimen. This corresponds to a magnetization field strength (H) of 80 0000 A m^{-1}. The MBE signal was amplified using a low noise preamplifier and a post amplifier (80 dB). The detailed experimental setup for MBE measurements has been reported elsewhere [14].

XRD measurements were carried out using MAC Science MXP18 X-ray diffractometer with Cr K_α radiation and the samples were analyzed in the complete angular range of 60-130°. The volume percent of austenite formed by reversion of martensite during ageing, was determined by direct comparison of the integrated intensities of the (111) and (200) planes of the γ phase with the intensities of the (110) and (200) planes of α phase.

Positron Doppler broadening measurements were carried out using a high purity germanium detector having an energy resolution of 1.4 keV at annihilation gamma ray energy of 511 keV. A defect sensitive line shape S-parameter viz. the ratio of central peak counts to total counts around 511 keV γ-ray is deduced from these measurements. The S-parameter signifies the positron annihilation events with low momentum electrons of the medium. Various defects such as vacancies, vacancy clusters, dislocations and precipitate–matrix interfaces act as trapping sites for positrons, leading to an increase in the S-parameter value [15].

The eddy current (EC) testing was carried out using MIZ-20A ZETEC system with optimized testing parameters such as frequency (10 kHz) and phase (5°). Specially designed probe consisting of transmit-receive (T/R) coil was used, which was relatively insensitive to permeability variations. The EC response is measured in terms of signal amplitude i.e. magnitude of real and imaginary components.

3.0 Results and Discussion

Figure 1 shows the variations in hardness, ultrasonic longitudinal wave velocity and positron annihilation parameter (S parameter) with aging duration. The variation in the ultrasonic velocity with aging time exhibits similar trends as that of the hardness. The velocity initially increases with aging time, peaked at intermediate ageing duration followed by continuous decrease at longer durations of ageing. In contrast, the S parameter decreased drastically from the solution annealed conditioned up to 1 h of aging and then increases with further aging.

Figure 2 show the variations in hardness, MBE rms voltage, volume % of austenite and EC amplitude value with aging duration. The variation in the EC amplitude with aging time exhibits similar trends as that of the volume of austenite determined from XRD. They were initially constant and increased drastically at longer aging time. In contrast, the MBE rms value remained constant initially and drastically dropped at longer aging durations.

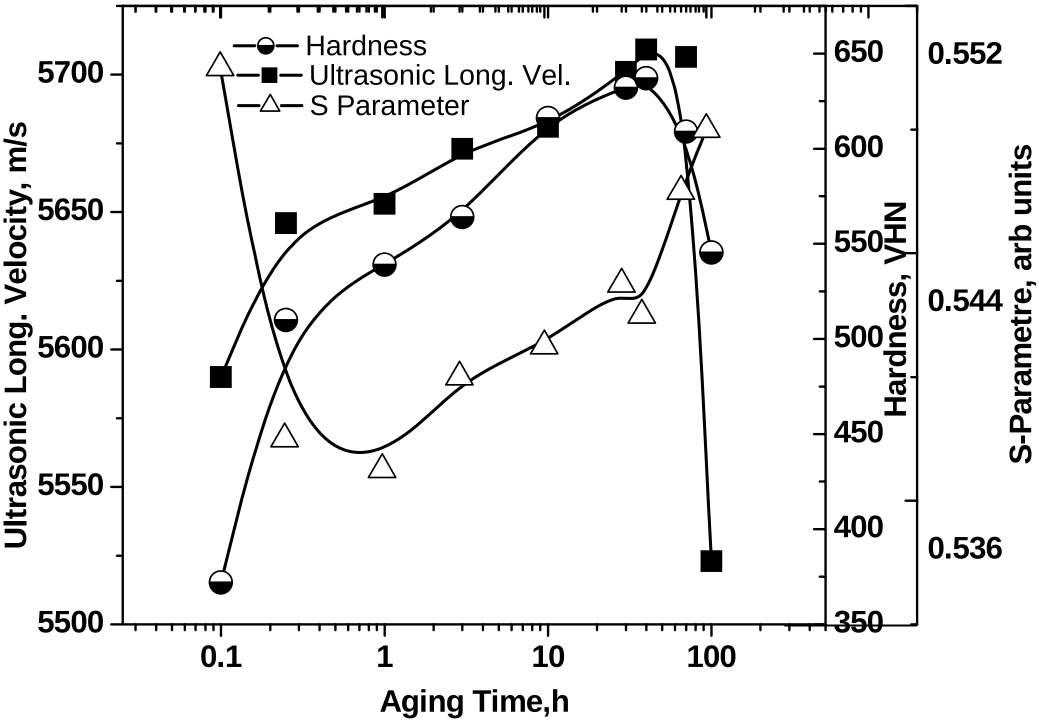

Fig. 1. Variation in hardness, ultrasonic wave velocity and Positron Annihilation (S Parameter) with aging at 755 K.

The initial increase in the hardness up to 3 h is attributed to the precipitation of Ni_3Ti intermetallic precipitates from the martensite matrix. The detailed transmission electron microscopy (TEM) results substantiating this have been reported elsewhere [16]. The continuous increase in the hardness at intermediate durations (10-40 h) is attributed to additional precipitation of fine Fe_2Mo precipitates from the solid solution. The decrease in the hardness upon ageing for longer durations is attributed to the formation of soft reverted austenite phase. The variation in ultrasonic velocity with aging exhibited similar behaviour as that of hardness. The initial increase in the ultrasonic velocity is attributed to the precipitation of mainly Ni_3Ti intermetallic phase, which increases the hardness up to 616 VHN. The subsequent steep rise in velocity reaching a maximum is attributed to the precipitation of Fe_2Mo along with Ni_3Ti. The variation in ultrasonic longitudinal wave velocity and hardness with ageing time could reveal various stages of precipitation and reversion of austenite; however the initiation of the reversion of austenite at 30 h of ageing could not be identified by ultrasonic velocity measurements. Though TEM studies revealed the initiation of reversion of austenite occurring below 30 h itself [16], decrease in hardness and ultrasonic longitudinal wave velocity is observed only after 40 h of ageing. This is attributed to the fact that the precipitation of Fe_2Mo, which tends to increase the hardness and velocities, continues to take place in parallel with the austenite reversion. Hence, decrease in the hardness and ultrasonic longitudinal wave velocity due to formation of austenite can be felt only when this decrease is more than the increase in these parameters due to continued Fe_2Mo precipitation. Drastic drop in velocity at longer aging duration is attributed to the formation of reverted austenite. Austenite has been reported to be a soft phase having lowest modulus among all the phases present in ferritic steels [17].

Unlike hardness and ultrasonic velocity, S parameter drastically decreases upon ageing up to 1 h duration and increases continuously beyond this up to 100 h of ageing (Figure 1). The initial decrease in S parameter up to 1 h of ageing is attributed to annihilation of defects and reduction in defect density associated with precipitation. The main defects observed in these steels are vacancies and dislocations. Reduction in the defect density with aging occurs especially during the initial aging time, when defect density is the highest. Hence, the decrease in the S-parameter in the initial regime (up to 1h) of aging is attributed to two simultaneous mechanism i.e. the reduction in defect density due to martensitic recovery and predominantly the precipitation of intermetallics preferentially on defects, which results in decrease in trapping sites. On the other hand, coarsening of precipitates acts as trapping sites due to the strain field associated with them, leading to the increase in the S parameter. Since the two processes affect the S parameter in opposite ways, net effect is manifested as continuous decrease up to 1 h showing the reduction in defect structure as the dominant feature beyond this the precipitation is found to be the dominant factor. Though at longer ageing durations (30-100 h), reversal of austenitic also take place, the S parameter seems to have subtle affect as it results in only structural change with less effect on defect concentration. The continuous increase in S parameter up to 100 h is the result of continuous precipitation of intermetallics, which acts as trapping sites for positrons.

Figure 2 shows the variation in EC amplitude with aging time for M250 maraging steel. EC amplitude remained almost constant up to 10 h of aging. Aging for 30 h resulted in drastic increase in EC response compared to 10 h. The EC amplitude continued to increase with further aging for longer durations reaching a value of 2.0 V at 100 h. Because of the specially designed probe, the effect of permeability is nullified and the change in EC response can be explained in terms of change in conductivity only. Though the EC amplitude remained constant with aging time up to 10 h, the hardness increases continuously in this regime due to precipitation of intermetallics. From this it can be inferred that the change in resistivity is negligible with precipitation of Ni_3Ti initially and Fe_2Mo at later stage. Upon ageing for 30 h, the EC amplitude increased drastically due to the presence of austenite in the microstructure. From the TEM investigations [16], it was also evident that austenite is observed earliest in the specimen aged for 30 h. Habiby et al. [18] have also reported similar observations in similar steel upon ageing. Even a very low volume fraction of austenite (less than 1% in specimen aged for 30 h) could be easily revealed because of the substantial difference in the resistivity values for martensite and austenite. The resistivity values for pure iron and stainless steel differ widely i.e. $2463\text{x}10^{-7}$ ohm-m and $7.496\text{x}10^{-7}$ ohm-m respectively. The eddy current parameter could be correlated with the amount of reverted austenite and hence has potential for in-situ detection and assessment for austenitic reversal on actual components.

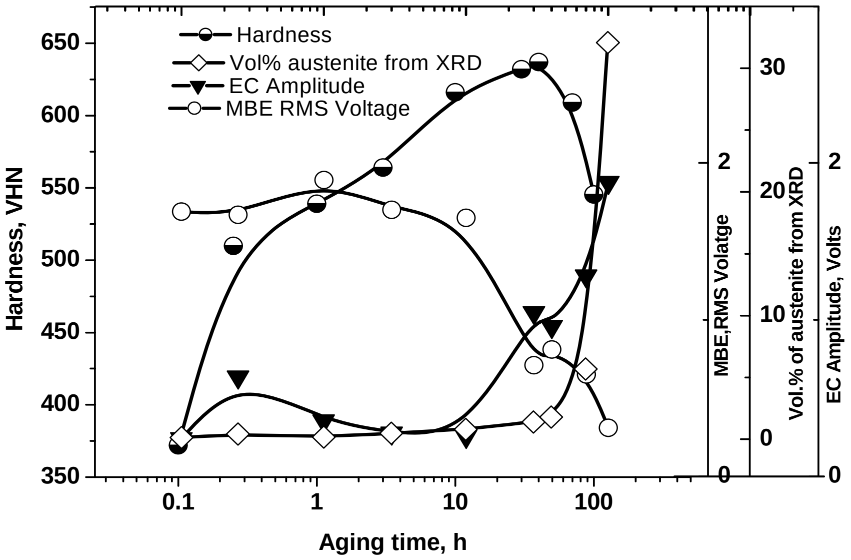

Fig. 2. Variation in hardness, magnetic barkhausen emission RMS,
Eddy current RMS value and Volume % of austenite
determined by XRD with aging at 755 K.

Figure 2 also shows the variation in MBE peak height and volume % of reverted austenite (measured by XRD) with ageing duration for the isothermally aged specimens at 753 K. The MBE peak height remains almost constant up to 10 h of aging. Beyond this, substantial drop in MBE rms voltage was observed on aging up to 100 h.

MBE rms voltage remains almost constant up to 10 h regime showing that the increase in MBE rms voltage due to dislocation annihilation is compensated by decrease in MBE rms due to precipitation of intermetallic phases. Hence, a net manifestation of constant voltage is obtained. Beyond 10 h of ageing, the substantial decrease in MBE rms is attributed to the formation of reverted austenite due to dissolution of Ni rich precipitates. An interesting observation is that the onset of austenitic reversion is picked up sensitively by MBE whereas it requires appreciable amount to be detected by XRD (~2% at 40 h). Otherwise early detection of this austenite reversion requires support of TEM. The increase in austenite from 30 h to 100 h is evident from XRD. From Fig. 2, it is also evident that upon 40 h, 70 h and 100 h of ageing, the volume of austenite increases continuously as 2, 6 and 30 % respectively. The substantial decrease in MBE with reversal of austenite is attributed to the fact that the paramagnetic austenite impedes domain wall movement in addition to reducing the total domains taking part in magnetization. This clearly indicates that the MBE technique is very sensitive to characterize the initiation of austenitic reversion.

Ultrasonic velocity showed good promise in characterizing the intermetallic precipitation process and was able to pick up the austenite information only when high amount of austenite is precipitated. Its drawback is with respect to obtaining any information about the defect structure and early detection of initiation of austenite reversion. Hence, this technique is found to be very sensitive to monitor the intermetallic precipitation behaviour. Positron annihilation studies were found to be very sensitive to defect structure and precipitation to some extent. The austenite reversion has almost no effect on positron annihilation parameter. Hence this technique can be used for high sensitive characterization of defects such as vacancies or dislocations. Magnetic Barkhausen emission study showed good promise in identifying the non-magnetic austenite phase compared to intermetallic precipitation and defect structure. Hence, MBE technique can be used selectively for very sensitive determination of early initiation of reversion of austenitic phase. Eddy current technique is also found to have good promise in early detection of initiation and characterization of the austenitic reversion ahead of ultrasonic and almost on par with MBE. As this technique is more amenable to shop floor, it can be used for determination of volume % of austenite in actual components with ease. XRD technique was found to be good in characterizing austenite but was unable to determine the austenitic initiation early due to low sensitiveness of the technique for austenite volume fraction (less than 2 %). Hence XRD technique can not be used for recognition of austenite initiation and moreover its portability to shop floor and to component site is difficult. However, XRD technique can be used as a benchmark for establishing correlations for quantitative estimation of volume fraction of reverted austenite using MBE and ECT techniques.

4.0 Conclusion

Various NDE techniques have been used in the present study for comprehensive characterization of microstructural features generated by ageing the solution annealed M250 maraging steel at 755 K for different durations in range of 0.25-100 h. Each technique provided complimentary information with regard to complex microstructural features that evolve during the aging treatment of maraging steel. Ultrasonic velocity was found to be more sensitive to the precipitation of intermetallics, whereas magnetic Barkhausen emission could clearly identify the onset of austenitic reversion. Positron annihilation spectroscopy could clearly identify the reduction in defect structure during initial aging periods, in addition to the characterization of continued precipitation of intermetallics at longer aging periods. XRD studies were used for quantitative determination of amount of reverted austenite. Eddy current parameters could be correlated with the amount of reverted austenite and has potential for in-situ assessment for austenitic reversion on actual components. The present study has clearly brought out the complementary nature of various NDE techniques for comprehensive characterization of ageing behaviour in maraging steels

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