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Assessment of microstructure stability of cold worked Ti-modified austenitic stainless steel during aging using ultrasonic velocity measurements and correlation with mechanical properties

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Abstract

As ultrasonic velocity is sensitive to the changes in texture, it is a more reliable technique than mechanical property measurements for assessment of microstructural stability (recrystallization behaviour) of cold worked alloy where recrystallization is coupled with precipitation. Hence, ultrasonic velocity measurements have been employed for studying the influence of Ti/C ratio on the microstructural stability of cold worked Ti-modified austenitic stainless steel during isochronal aging. In this alloy precipitation of TiC is known to retard recovery and recrystallization. The variation in ultrasonic velocity with aging temperature exhibited a three stage behaviour at all three frequencies employed (2, 10 and 20 MHz) and correlated well with the microstructural changes. Based on the microstructural investigations, the three stages have been identified to be recovery, progress of recrystallization and completion of recrystallization. There was one to one correspondence between the variation in the hardness, strength values and the variation in the ultrasonic velocity values as a function of aging temperature in assessing the microstructural changes, except when the interaction between the TiC precipitation and recrystallization is stronger.

1. Introduction

In cold worked materials, simple recrystallization (nucleation of new dislocation-free grains) is often accompanied by various microstructural changes like recovery, decomposition of solid solution, precipitation of second phases, etc. Use of parameters such as hardness, strength, electrical conductivity, internal friction etc. for assessment of recrystallization is less reliable if recrystallization is coupled with precipitation [1]. Hence, actual temperature or time for the onset and completion of recrystallization cannot be determined accurately by these techniques based on mechanical and physical property variations. However, the actual onset and completion of recrystallization can be determined in this type of material if a technique is sensitive only to the changes in texture caused by recrystallization. Though techniques based on X-rays are sensitive to changes in texture, their measurements are restricted to surface only.

Ultrasonic velocity measurements have been successfully used to characterize microstructural changes during hardening and tempering in steels, age hardening in aluminium alloys and to estimate grain size in austenitic stainless steels etc. [2–5]. Jayakumar [6] has used ultrasonic velocity measurements for estimating the degree of cold work (CW) in an austenitic stainless steel. Since recrystallized microstructure exhibits a different

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grain texture compared to unrecrystallized microstructure in alloys and thus possesses different elastic constants, the measurement of ultrasonic velocity has the potential for determining the degree of recrystallization. Also, the technique enables volume measurements averaged throughout the thickness of the specimen. Generazio [7] used successfully ultrasonic attenuation measurements for determining the onset, degree and completion of recrystallization in super alloys. Stiffler et al. [8] have employed ultrasonic velocity measurement technique using horizontally polarized shear waves for determining the degree of recrystallization in an aluminium sheet. Vasudevan et al. reported first the use of ultrasonic thickness meter (velocity measurements) for characterizing annealing behaviour in a cold worked austenitic stainless steel [9]. Vasudevan et al. [10–13] also reported the superiority of ultrasonic velocity measurements over mechanical property measurements to precisely measure the time and temperature for onset of recrystallization in this type of alloys.

It is important to study the influence of Ti/C ratio on the microstructural stability of 20% cold worked 15Cr–15Ni–2.2Mo–Ti modified austenitic stainless steel during aging, because this material in the 20% cold worked condition is the reference material for the clad and wrapper components of Prototype Fast Breeder Reactor (PFBR) in India. The amount of secondary TiC precipitation mainly depends on the Ti/C ratio of the alloy [14]. Exposure at elevated temperatures during service would degrade the cold worked microstructure due to recovery and recrystallization. Degradation in the cold worked microstructure in turn would decrease the resistance of the material against irradiation damage. In the cold worked condition, precipitation of TiC is reported to interact strongly with recovery and recrystallization processes [15–17] and hence use of techniques such as hardness measurements to study the microstructural stability can only give approximate results. Metallography and hardness testing techniques could be used to obtain information about the degradation of microstructure of these core components during post-irradiation examination. However, these techniques will be cumbersome when applied to testing of highly radioactive materials. On the other hand, techniques based on ultrasonics are proven for quick and quantitative microstructural characterization and having scope for non-contact measurements. Hence, this technique has potential to be used during post-irradiation examination to study the microstructural stability of cold worked alloy exposed to elevated temperatures and irradiation environments.

In the present investigation, precise ultrasonic velocity measurements have been carried out in the frequency range 2–20 MHz to characterize the microstructural stability of cold worked titanium modified austenitic stainless steel with different Ti/C ratios during aging. Ultrasonic velocity values were correlated with the microstructures obtained using optical metallography. Velocity measurements have also been compared with that of hardness and strength measurements.

2. Experimental

Ti-modified austenitic stainless steel whose composition is similar to the alloy specified in ASTM A771 (UNS S38660) which is also known as alloy D-9 has been chosen as the prime candidate material for the fuel clad and hexagonal wrapper components of the PFBR. Chemical compositions of the two alloys with different Ti/C ratios used in the present investigation are given in Table 1. The alloys used in the investigation were produced from virgin raw materials by vacuum induction melting followed by vacuum arc remelting. Then the cast ingots were hot forged and hot rolled to 30 mm diameter rods. The rods were then reduced to 11 mm diameter by rotary swaging. The rods were solution annealed at 1343 K for half an hour. The average grain size after annealing was found to be in the range of 20–30 μm. The rod was deformed in tension in an Instron 1195 universal testing machine to impart 20% CW. Specimens cut from the cold worked rod were isochronally aged in the temperature range 873–1173 K for times of 1, 100, 1000 h.

Ultrasonic measurements have been performed using 2–20 MHz broad band longitudinal wave probes. Ultrasonic velocity is determined by measuring the time taken for the ultrasonic waves to travel the thickness of the material with parallel faces. In the present work, a PC based system using LabVIEW software was used to determine the ultrasonic velocity values. This software takes the specimen thickness as input. Totally, five measurements were made on each specimen and the average of the ultrasonic velocity was obtained for each specimen. The overall variation obtained in the velocity

Table 1
Chemical composition (wt%) of the Ti-modified austenitic stainless steels with different Ti/C ratios used in this study

C	0.0523	0.0522
Ni	15.04	15.09
Cr	15.1	15.05
Mo	2.26	2.248
Ti	0.21	0.315
Mn	1.51	1.51
Si	0.5	0.51
N	0.006	0.0066
S	0.0033	0.0025
P	0.011	0.011
B	0.001	0.0011
Fe	Balance	Balance

measurement by this method at a specified location was ±3 m/s. Optical metallography has been used to reveal the microstructural features of the 20% CW and aged specimens. Vickers hardness was measured using a load of 10 kg on all the specimens. A minimum of five hardness measurements were made and the average value was taken. Room temperature tensile testing was carried out to determine the strength properties. X-ray diffraction were carried out to study the change in the texture. Onset and completion of recrystallization were confirmed by estimating the volume fraction of material recrystallized using quantitative metallography.

Table 2
Ultrasonic velocities (m/s) in different material conditions at 2, 10, 20 MHz longitudinal wave frequencies for the alloy with Ti/C = 6

Material condition	Ultrasonic velocities (m/s)		
	2 MHz	10 MHz	20 MHz
20% cold worked	5883	5921	5911
Recrystallized	5723	5772	5758

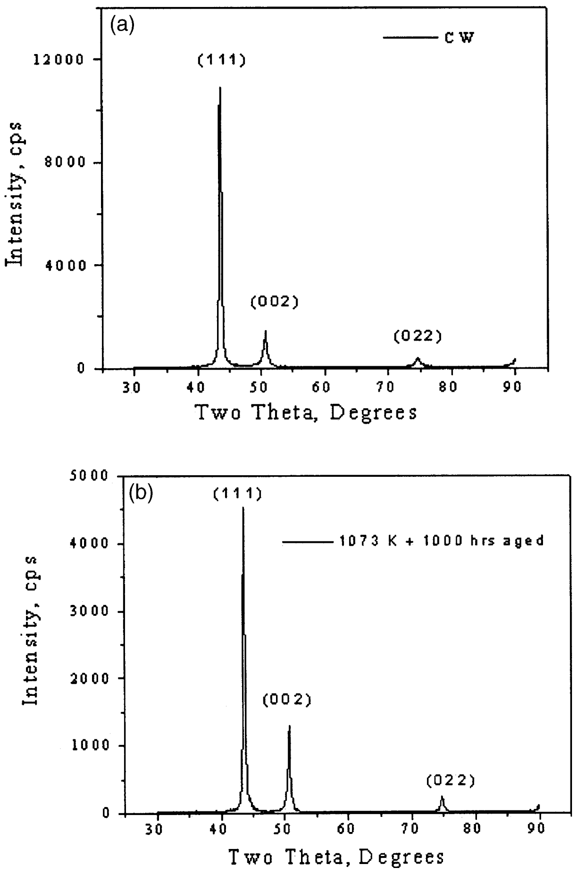

Fig. 1. X-ray diffraction data for the (a) 20% cold worked and (b) 20% CW + 1073 K, 1000 h aged.

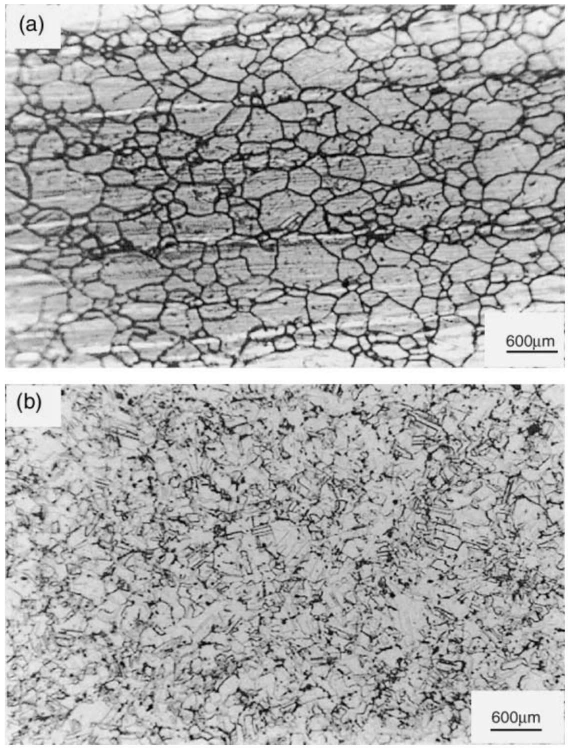

Fig. 2. Optical microstructure for the samples: (a) 20% cold worked and (b) 20% CW + 1073 K, 1000 h aged.

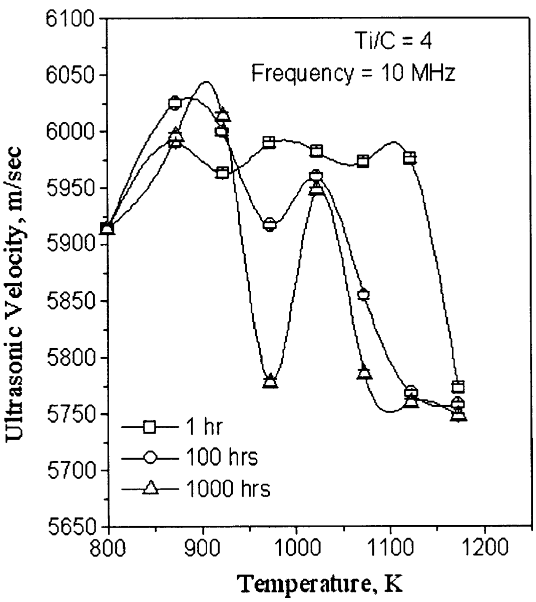

Fig. 3. The variation of ultrasonic velocity with aging temperature for alloy D-9 with Ti/C ratio 4.

3. Results and discussion

The velocity, hardness and yield strength values measured for the cold worked condition are marked on the ordinate of the respective figures showing the variation of velocity, hardness and strength with aging temperature. The ultrasonic velocity values measured on the 20% cold worked and recrystallized samples using longitudinal waves at 2, 10, 20 MHz frequencies are given in Table 2. There is a difference of 2.5% (about 150–160 m/s) between the velocity values measured on the samples in the different material conditions i.e. cold worked and recrystallized condition. In order to find out the reason for the large difference in the velocity values, X-ray diffraction pattern for cold worked and completely recrystallized samples were obtained. Fig. 1(a) and (b) shows the X-ray diffraction data for the cold worked, and 1073 K, 1000 h aged samples and Fig. 2(a) and (b) shows the optical microstructures of the respective samples. There is a definite change in the intensity of the (1 1 1) reflection during annealing at 1073 K. The intensity of this reflection decreased by more than 50% due to recrystallization. The texture change observed by X-ray diffraction caused the drastic decrease in velocity during recrystallization. Hence, the sensitivity of ultrasonic velocity to the occurrence of recrytallization in the 20% cold worked Ti-modified austenitic stainless steel is attributed to the decrease in the intensity of (1 1 1) reflections as observed in the X-ray diffraction pattern.

3.1. Effect of Ti/C ratio

Fig. 3 shows the variation of ultrasonic velocity (10 MHz) with aging temperature for the austenitic stainless steel with Ti/C ratio of 4 isochronally annealed at 1, 100 and 1000 h. After 1 h aging, the velocity values for the samples aged in the temperature range 873–1123 K were

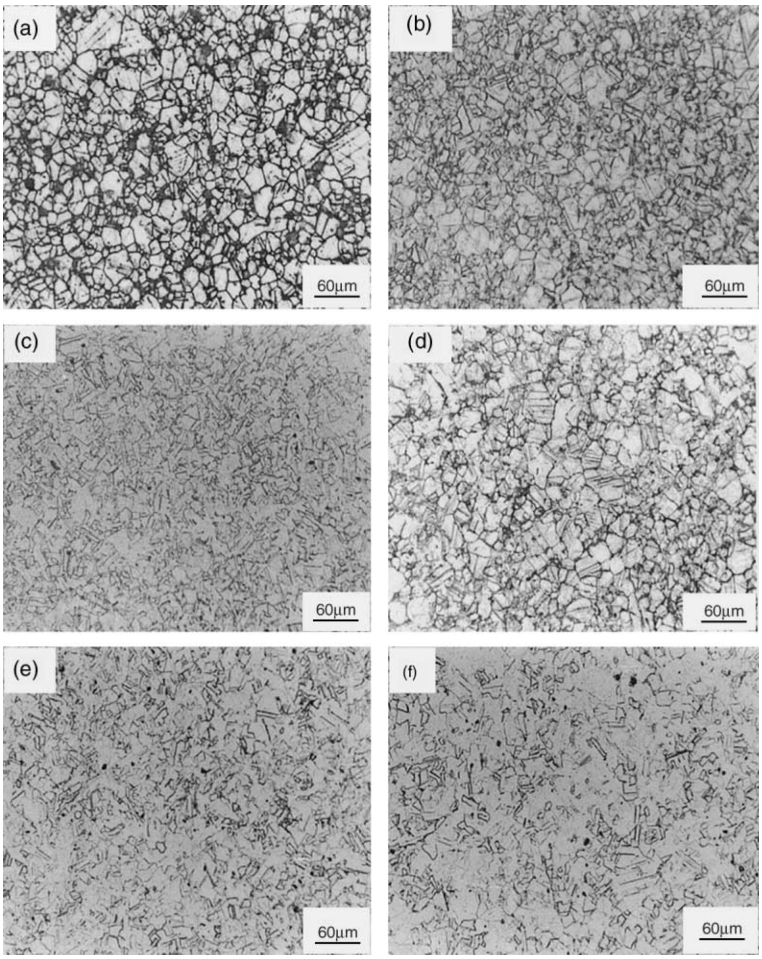

Fig. 4. Optical microstructures of the samples of the alloy with Ti/C ratio of 4 aged at (a) 923 K, 1000 h; (b) 1023 K, 100 h; (c) 1023 K, 1000 h; (d) 1073 K, 100 h; (e) 1123 K, 100 h and (f) 1173 K, 1 h.

greater at least by 50 m/s compared to that of the cold worked condition. The velocity value for the sample aged at 1173 K was lower by 150 m/s compared to that of the cold worked condition. Microstructural observations revealed the occurrence of recrystallization only in the sample aged at 1173 K. The increase in the velocity observed in the samples aged in the temperature range 873–1123 K was attributed to recovery. During recovery, annihilation of point defects and loss of dislocations takes place which reduces the distortion in the lattice caused by CW and also alters the modulus. Hence, there was an increase in velocity due to the reduction in the distortion of the lattice. The decrease in velocity at 1173 K after 1 h of annealing was attributed to the change in the grain orientation (decrease in the intensity of (1 1 1) reflection) caused by recrystallization. After 100 and 1000 h of annealing, the variation of velocity exhibited an increase of 50–100 m/s in the temperature range 873–923 K, followed by a drop at 973 K and distinct peak at 1023 K and continued to decrease until it reached saturation at 1123 K.

Fig. 4(a)–(f) shows the optical microstructures of the samples aged at different temperatures and time for the alloy with Ti/C ratio of 4. Fig. 4(a) shows the microstructure of the samples aged at 923 K for 1000 h. There is no evidence of recrystallization. Fig. 4(b) and (c) shows the microstructures of the samples aged for 100 and 1000 h at 1023 K respectively. There is extensive precipitation in the interior of the grains and on the grain boundaries which retards recrystallization. Fig. 4(d) shows the optical microstructure of the sample aged for 100 h at 1073 K. The alloy has undergone partial recrystallization at this condition. Fig. 4(e) and (f) shows the microstructure of samples aged for 100 h at 1123 K and the sample aged for 1 h at 1173 K. Recrystallization is complete at both the conditions. For the alloy with Ti/C ratio of 4, complete recrystallization occurs only after aging for 1000 h at 973 and 1073 K.

Fig. 5 shows the variation of velocity (10 MHz) with aging temperature for the austenitic stainless steel with Ti/C ratio of 6 for different isochronal annealing times. The noted difference between this material and the material with Ti/C ratio of 4 is that after 100 h of annealing this material undergoes complete recrystallization at 973 K though it also exhibits retardation of recrystallization by TiC precipitation at 1023 K. After 1000 h of annealing the material undergoes complete recrystallization in the temperature range 973–1173 K while the material with Ti/C ratio of 4 exhibited complete retardation of recrystallization at 1023 K. The observed difference in the behaviour is essentially attributed to the amount of secondary TiC precipitation which is higher in the material with Ti/C ratio of 4 [14]. The retardation of recovery and recrystallization by TiC precipitation in this alloy has been reported based on TEM investigations [15–17]. Though precipitation interfered with recovery and recrystallization, the ultrasonic velocity being sensitive to textural changes predicted the onset of recrystallization more precisely. Fig. 6(a)–(f) shows the microstructures of the samples aged at different temperatures and times for the alloy with Ti/C ratio of 6. Fig. 6(a) shows the microstructure of the sample aged for 1000 h at 923 K and there is no evidence of recrystallization. Fig. 6(b) and (c) shows the microstructure of the samples aged for 100 and 1000 h at 973 K. Recrystallization is complete at both the conditions. Fig. 6(d) and (e) shows the microstructure of the samples aged for 100 and 1000 h at 1023 K. There is extensive precipitation in the grain interior and at the grain boundaries. Hence, there is retardation of recrystallization. Fig. 6(f) shows the microstructure of the samples aged for 1 h at 1173 K. The recrystallization is complete. For the alloy with Ti/C ratio of 6, complete recrystallization occurs after aging for 100 hrs at 973 K.

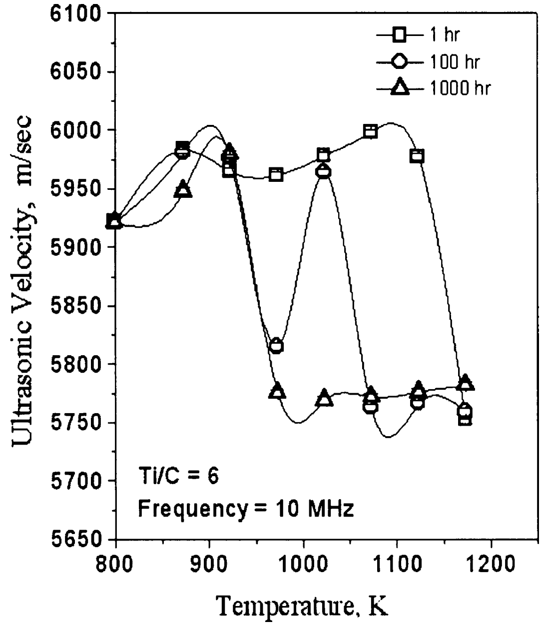

Fig. 5. The variation of ultrasonic velocity with aging temperature for alloy D-9 with Ti/C ratio 6.

3.2. Effect of frequency

Fig. 7(a)–(c) compares the variation of ultrasonic velocity with annealing temperature at different frequencies for the alloy with Ti/C ratio 6. After isochronal aging for 1, 100 and 1000 h, the velocity measured at all the frequencies of 2, 10, 20 MHz on the aged specimens showed the same onset, progress and completion time and temperature of recrystallization. In the frequency range studied, the assessment of the microstructural stability of 20% cold worked Ti-modified austenitic

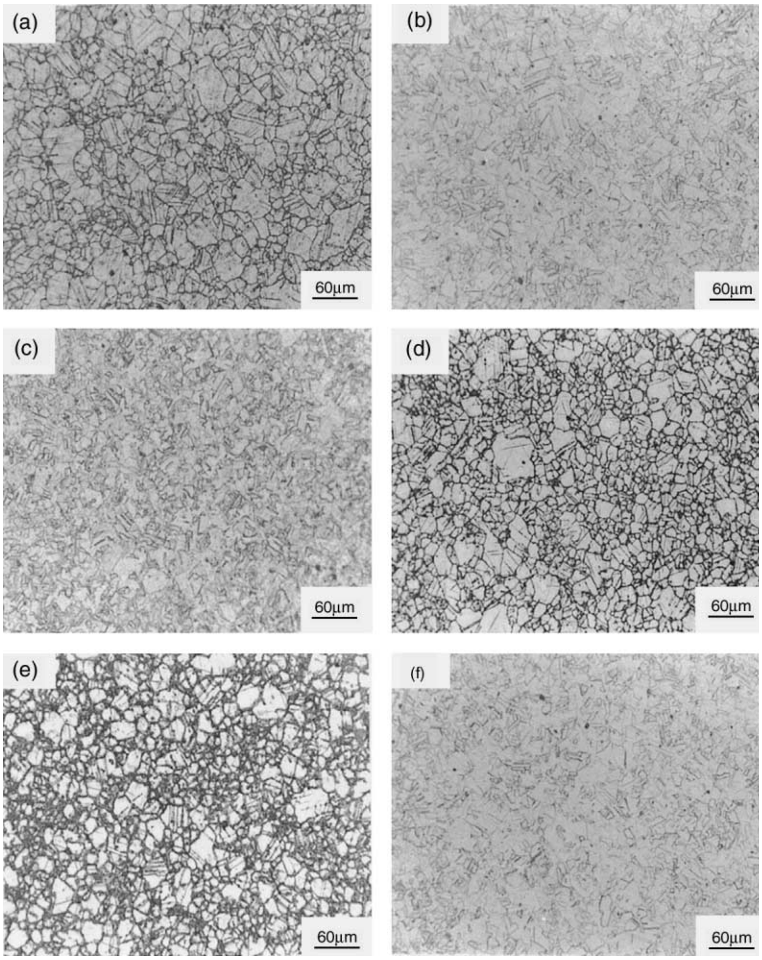

Fig. 6. Optical microstructures of the samples of the alloy with Ti/C ratio of 6 aged at (a) 923 K, 1000 h; (b) 973 K, 100 h; (c) 973 K, 1000 h; (d) 1023 K, 100 h; (e) 1023 K, 1000 h and (f) 1173 K, 1 h.

stainless steel due to aging is independent of the frequency of the ultrasonic waves. This is in agreement with our earlier work [13].

3.3. Comparison with hardness measurements

Figs. 8(a)–(c) and 9(a)–(c) show the comparison between the variation of hardness and ultrasonic velocity with aging temperature for the alloys with Ti/C ratio of 4 and 6 respectively. For both the alloys, after 1 h of aging hardness decreased continuously with increasing aging temperature from that of the cold worked condition. The variation of velocity increased with increase in aging temperature from that of the cold worked condition and exhibited a sharp decrease at 1173 K. Microstructural observations revealed the occurrence of recovery in the temperature range 873–1123 K and complete recrystallization only at 1173 K. After 100 and 1000 h of aging, the variation of hardness and the velocity with aging temperature showed one to one correspondence in assessing the microstructural changes in the temperature range 973–1173 K. In comparison to hardness measurements, the velocity measurements could characterize distinctly the recovery and recrystallization regimes. Thus, the onset temperature for recrystallization could be determined more exactly using velocity measurements.

3.4. Correlation with yield strength

Figs. 10 and 11 show the comparison of variation of yield strength and ultrasonic velocity as a function of aging temperature for the alloys with Ti/C ratios of 4 and 6 respectively at different aging times. For both the alloys, after 1 h aging, continuous decrease in yield strength was observed in the temperature range 873–1123 K followed by a rapid decrease at 1173 K. The velocity values were higher in the temperature range

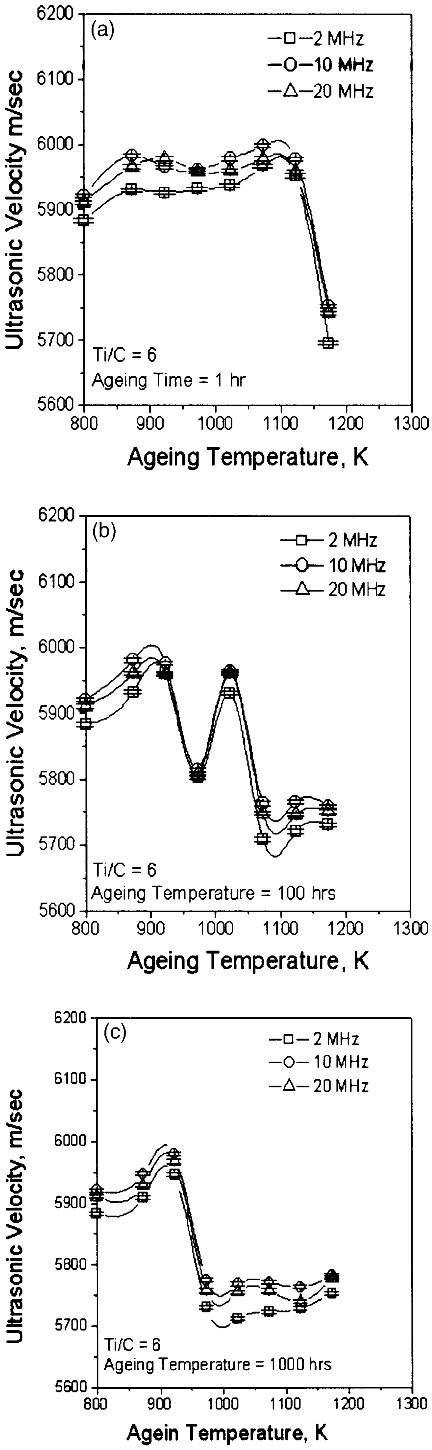

Fig. 7. The variation of velocity with aging temperature after aging for (a) 1 h, (b) 100 h and (c) 1000 h at different frequencies of 2, 10, 20 MHz.

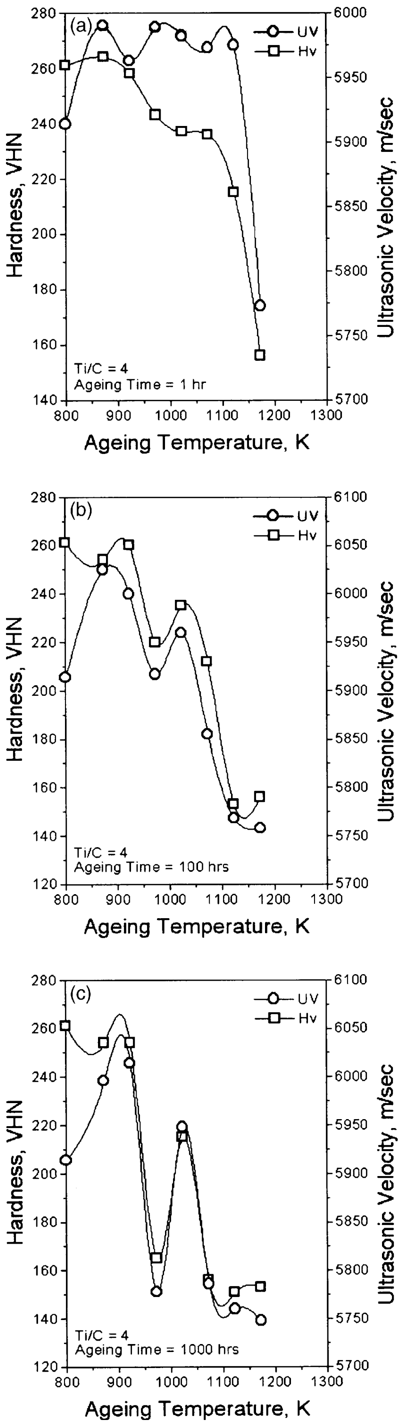

Fig. 8. The comparison of the variation of ultrasonic velocity and hardness with aging temperature for the alloy with Ti/C ratio of 4 at different isochronal annealing times: (a) 1 h, (b) 100 h and (c) 1000 h.

873–1123 K than the cold worked condition followed by a rapid decrease at 1173 K. For the alloy with Ti/C ratio of 6, a minimum in yield strength value is observed at 973 K (Fig. 11(a)) after 1 h of aging.

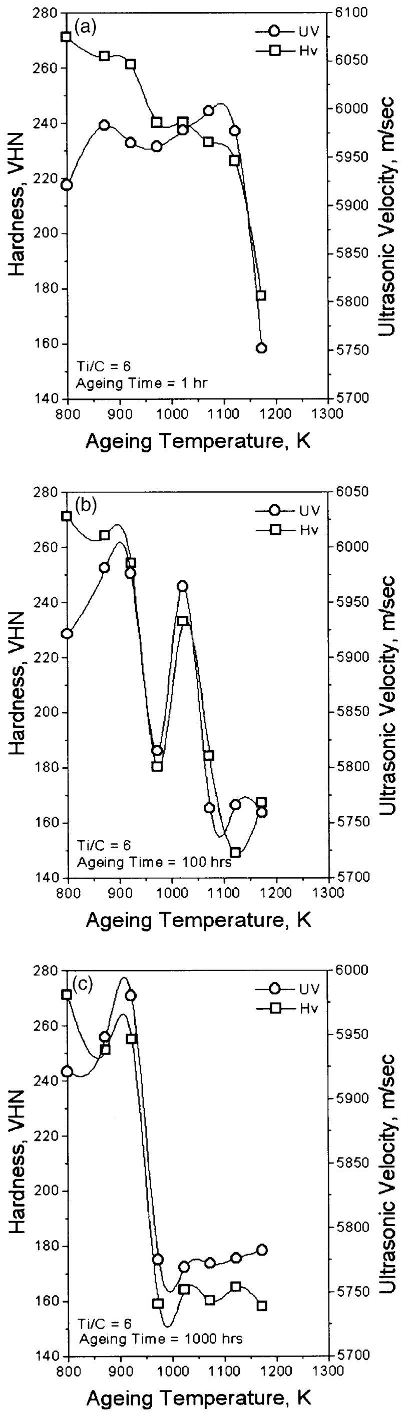

Fig. 9. The comparison of the variation of ultrasonic velocity and hardness with aging temperature for the alloy with Ti/C ratio of 6 at different isochronal annealing times: (a) 1 h, (b) 100 h and (c) 1000 h.

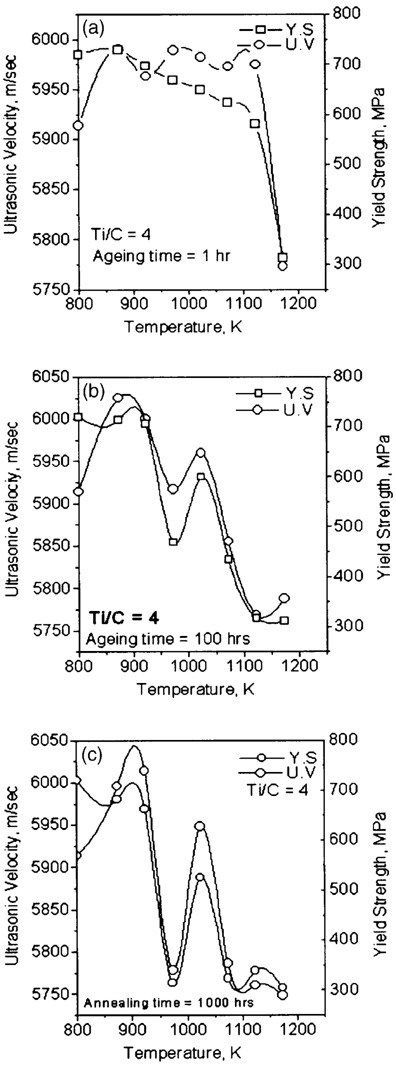

Fig. 10. The comparison of the variation of ultrasonic velocity and yield strength with aging temperature for the alloy with Ti/C ratio of 4 at different isochronal annealing times: (a) 1 h, (b) 100 h and (c) 1000 h.

Microstructural observations revealed that after 1 h of aging, recovery had occurred in the temperature range 873–1123 K while recrystallization occurred at 1173 K. Thus, velocity measurements could distinctly characterize the recovery and recrystallization regimes. A minimum in yield strength observed at 973 K after 1 h of aging for the alloy with Ti/C ratio of 6 may be attributed to earlier onset of recrystallization at this condition. For both the alloys, the yield strength decreased by about 200 MPa at 973 K followed by a peak at 1023 K and then decreased continuously till it reached saturation at 1123 K after 100 h of aging. From the observation of yield strength, it appears that recrystallization has progressed considerably at 973 K. However, microstructural observation and velocity measurements indicate that only onset of recrystallization has occurred

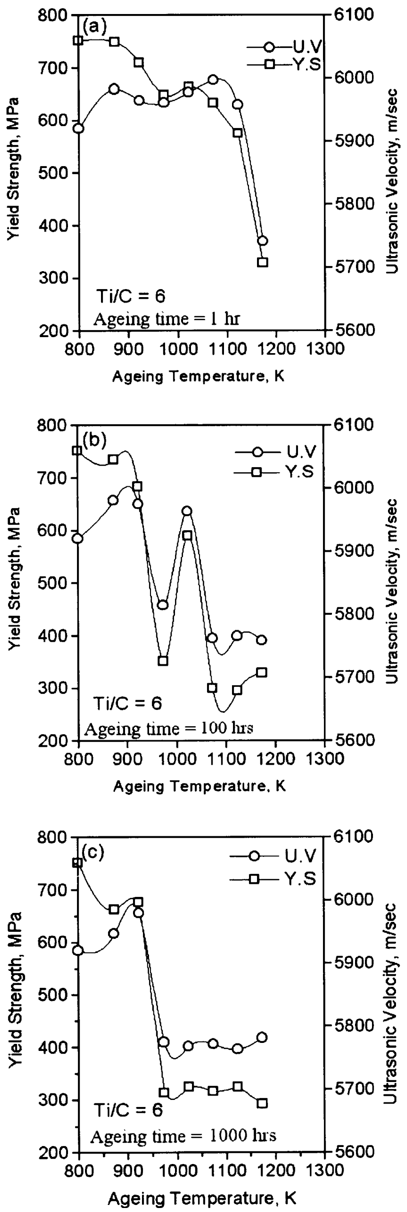

Fig. 11. The comparison of the variation of ultrasonic velocity and yield strength with aging temperature for the alloy with Ti/C ratio of 6 at different isochronal annealing times: (a) 1 h, (b) 100 h and (c) 1000 h.

at 973 K. The considerable decrease in yield strength was attributed to the coarsening of TiC precipitates caused by the onset of recrystallization. The coarsening of these precipitates during the onset of recrystallization in this type of alloy has been reported in the literature [15–19]. From the yield strength values it is difficult to know the extent of recrystallization. After 1000 h of aging, for both the alloys, the variation of yield strength and velocity exhibited a sharp drop at 973 K followed by a peak at 1023 K and then reached saturation at 1073 K. Both yield strength and velocity predict recovery in the temperature range 873–923 K, complete recrystallization at 973 K and in the temperature range 1073–1173 K and retardation of recrystallization at 1023 K. The velocity and the microstructural observations indicate complete retardation of recrystallization at 1023 K while yield strength values indicate occurrence of recrystallization. So, interaction of the TiC precipitation with recrystallization has led to wrong indication from yield strength values while there was good correlation between the velocity values and the microstructural observations. Hence, tensile property measurements are not a good technique to study the stability of cold worked microstructure when there is secondary precipitation associated with recrystallization. We should use techniques which are sensitive to texture like ultrasonic velocity measurements in those alloys. Though the present study was carried out only on the thermally aged samples, ultrasonic velocity measurements should be tried during post irradiation examination of this alloy to assess the microstructural degradation caused by the elevated temperature and irradiation exposure. This technique may be useful along with TEM to characterize the microstructural evolution in this alloy during irradiation service.

4. Conclusions

1. The cold worked alloys with Ti/C ratio of 4 and 6 undergo recovery in the temperature range 873–923 K after aging for 1, 100 and 1000 h. Both the alloys recrystallize completely after aging for 1 h at 1173 K. The alloy with Ti/C ratio of 4 recrystallizes completely after aging for 100 h at 1123 K and after aging for 1000 h at 973 K and 1073 K. The alloy with Ti/C ratio of 6 recrystallizes completely at 973 K after aging for 100 and 1000 h and exhibit retardation of recrystallization at 1023 K after 100 h of aging.

2. The ultrasonic velocity measurements correlated well with the microstructural observations of the onset temperature for recrystallization due to variation in the Ti/C ratio of the alloy.

3. There was one to one correspondence between the variation in the hardness, strength and velocity measurements in assessing the microstructural changes during aging of the cold worked alloy except under some conditions described in the text. When there is not good correlation among these properties we speculate that there is dissolution of TiC precipitates due to the onset of recrystallization. The variation in the ultrasonic velocity measurements correlated well with the microstructural changes and the extent of recrystallization.

4. The ultrasonic velocity measurements could be employed to precisely measure the time and temperature

for onset of recrystallization compared to that of the mechanical property measurements in this type of alloy.

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