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Aging of a copper bearing HSLA-100 steel

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Abstract. Investigations were carried out on aging of a HSLA-100 steel after varying amounts of cold deformation. Mechanical properties (hardness, tensile properties and toughness) were measured and structural changes were studied using optical, TEM and SEM techniques. As a result of various treatments, the hardness and UTS could be significantly improved, but with drastic fall in ductility and impact strength, especially in peak aged conditions. The parameters affecting impact strength were examined and it was concluded that various microstructural features affected toughness through their influence on tensile properties. In this steel the impact strength could be improved by lowering the UTS and increasing the ductility (pct elongation). The improvement in hardness and UTS was attributed to formation of thick precipitate-dislocation tangles. The aging process caused a slow transformation of lath martensite into acicular ferrite due to occurrence of *in situ* recrystallization. The concentration of Cu in particles precipitating on aging was followed using EDAX technique.

Keywords. HSLA steel; thermomechanical aging; Cu-bearing steel.

1. Introduction

The Cu-containing age hardening steels have been of interest for a number of years owing to their desirable combination of strength and impact behaviour (Ranganathan 1999; Skoufari-Themistou *et al* 1999; Dhua *et al* 2001). The design philosophy underlying such steels is to reduce the carbon content to ~ 0·05 wt pct, to increase toughness and weldability and increase toughness via the precipitation hardening effects associated with high (1–2%) Cu contents. In general, the precipitation strengthening effect of Cu is complimentary to the strengthening induced by other elements which are used for precipitation hardening, such as Nb, Ti and V; these being added to steels to control austenite grain size as well as subsequently to strengthen the transformed structure of ferrite, bainite or martensite. If a Cu bearing HSLA steel is given cold working treatment after austenitization, the dense dislocation–cluster network on aging should provide effective hindrance to the mobility of dislocations. At the same time, cold working should effectively induce aging of an austenitized HSLA steel when subjected to high temperature.

Resistance to brittle fracture has been a major concern in the development of HSLA steels. Studies on toughness of high strength steels have been attempted by various workers (Gordon *et al* 1993; Hamano 1993; Densley and Hirth 1998; Lee *et al* 1998). For development of high strength materials possessing reasonable toughness, it is important to understand the effect of precipitate phases on plastic deformation behaviour. For example, several age hardenable high strength materials show the lowest toughness and the maximum strength in the peak-aged condition (Higashi *et al* 1985). The coherent precipitates are easily cut by moving dislocations (Sanders and Starke 1982; Duerig *et al* 1985; Higashi *et al* 1985; Hamano 1993), which leads to inhomogeneous deformation, early crack initiation, and poor toughness. In the over aged condition, the strengthening precipitates are incoherent and can be bypassed by dislocations leading to homogeneous deformation and increasing toughness with delayed crack initiation. Accordingly, the high strength materials sometimes have to be used in the over aged condition with improved toughness, although at the cost of strength.

The present work was undertaken to study the aging behaviour of a Cu-bearing HSLA-100 steel and to identify microstructural effects on strengthening and impact strength of this steel.

2. Experimental

The steel with a composition (wt pct) C 0·04, Mn 0·86, P 0·004, S 0·002, Si 0·27, Cu 1·58, Ni 3·55, Cr 0·57, Mo 0·60, Al 0·032 and Nb 0·03 was provided by the Naval Research Laboratory, Washington in the form of plate of dimensions 300 × 200 × 50 mm. The steel in the

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as-received (AR) condition possessed a mixed ferrite–bainite microstructure with fine carbonitride and Cu precipitates.

The steel in the AR condition was given the following treatments: (a) OQ: As-received material austenitized at 1000°C for 180 min, followed by oil quenching; (b) QT: OQ treated material tempered at 450°C for 60 min, followed by water quenching; (c) C25A: OQ treated material cold worked to 25 pct reduction by rolling followed by aging at 600°C for various times from 1 min to 1500 min, followed by water quenching; (d) C50A: OQ treated material cold worked to 50 pct reduction by rolling, followed by aging at 600°C for various times from 1 min to 1500 min, followed by water quenching; (e) C80A: OQ treated material cold worked to 80 pct reduction by rolling, followed by aging at 600°C for various times from 1 min to 1500 min, followed by water quenching and (f) TC25A: OQ treated material tempered at 700°C for 120 min, followed by oil quenching, after which the material was cold worked to 25 pct reduction by rolling, followed by aging at 500°C for various times from 1 min to 3000 min, followed by water quenching.

The treatments C25A, C50A and C80A were given to produce particle–dislocation networks of different magnitudes. The TC25A treatment was carried out to make fine/coherent and coarse incoherent precipitates coexist in the matrix.

The Vickers hardness tests were performed to establish the aging curves. Cylindrical tensile specimens were prepared with a gauge length of 25 mm and a gauge diameter of 5 mm. Tensile tests were conducted on Hounsfield tensometer with a crosshead speed of 1 mm/min. Charpy impact tests were performed at room temperature as per ASTM specifications. Extensive structural investigation was conducted using a high resolution light microscope, a 200 kV transmission electron microscope and a 30 kV scanning electron microscope. Detailed examination and tests were conducted in cold worked (CW), peak aged (PA) and over aged (OA) conditions in the various treatments, as well as in the tempered (T) condition in TC25A treatment. In any treatment, OA condition refers to the end of aging process.

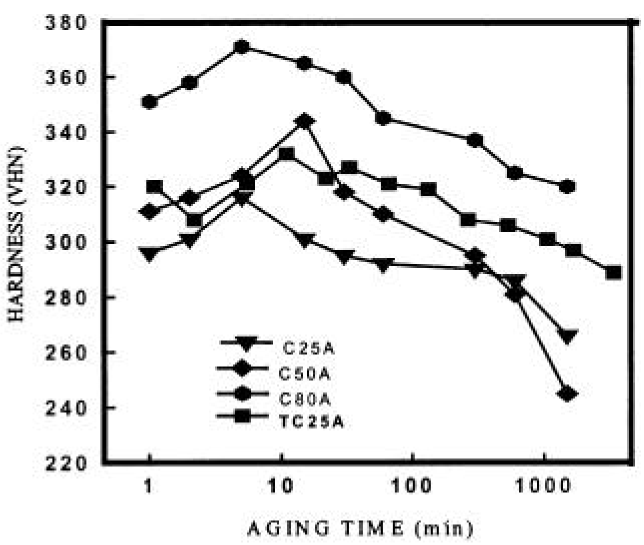

Figure 1. Variation of hardness with aging time.

3. Results

3.1 *Mechanical properties*

The variation of hardness with aging time in the treatments of C25A, C50A, C80A and TC25A is given in figure 1. It was observed that maximum response to age hardening occurred in the C80A treatment, in which a hardness of 371 VHN and UTS of 150·34 kg.mm^{-2} could be obtained in PA condition. The peak occurred after 5 min of aging in C25A and C80A treatments, 15 min in C50A and 10 min in TC25A. Table 1 gives various mechanical properties obtained as a result of the treatments. It was observed that the high hardness and UTS in PA conditions compared to OA were generally accompanied by low ductility (pct elongation) and charpy impact strength, which may restrict the use of this steel. It may be noted that among all the treatments, the TC25A treatment in OA condition yields a high impact strength (30·30 kg·m·cm^{-2}), good hardness (289 VHN) and UTS (94·99 kg·mm^{-2}) together with reasonably good ductility (15·00 pct elongation), which is essential for practical applications.

3.2 *Optical metallography*

The structure in the as-quenched condition consisted of martensite laths together with some bright etching proeutectoid ferrite. Severe cold deformation by rolling caused shearing and fragmentation of laths of martensite. The laths tended to align along the flow lines developed due to cold rolling. Aging of cold deformed samples caused conversion of laths of martensite into acicular ferrite (figure 2). The examination at various stages of aging showed that the transformation of lath structure into acicular ferrite became slower as the amount of deformation prior to aging increased.

3.3 *Transmission electron microscopy (TEM)*

The TEM study of the oil quenched and cold worked specimens showed laths of high dislocation density. Severe cold working (80 pct) resulted in a much higher dislocation density as compared to 25 pct reduction. At 80 pct reduction thick tangles of dislocations were produced at lath boundaries as well as within the laths. The transmission electron micrographs of the specimens after

Table 1. Mechanical properties of HSLA-100 steel.

Treatment	Designation	Hardness (VHN)	UTS ($kg \cdot mm^{-2}$)	Elongation (pct.)	Impact strength ($kg \cdot m \cdot cm^{-2}$)
AR		231	73·70	27·60	33·80
OQ		289	94·31	15·56	23·25
QT		291	89·30	16·70	26·90
	CW	292	102·61	12·32	15·88
C25A	PA	316	103·77	13·67	16·64
	OA (1500 min)	266	98·49	18·81	25·39
	CW	300	109·83	10·51	16·91
C50A	PA	344	112·82	14·03	12·28
	OA (1500 min)	245	60·24	20·81	33·42
	CW	331	126·97	2·84	4·44
C80A	PA	371	150·34	6·26	9·80
	OA (1500 min)	320	129·58	6·36	9·95
	T	269	87·07	16·22	26·67
TC25A	CW	324	103·03	12·66	18·45
	PA	332	106·12	13·95	19·17
	OA (3000 min)	289	94·99	15·00	30·30

CW: Cold worked; PA: peak aged; OA: over aged; T: tempered.

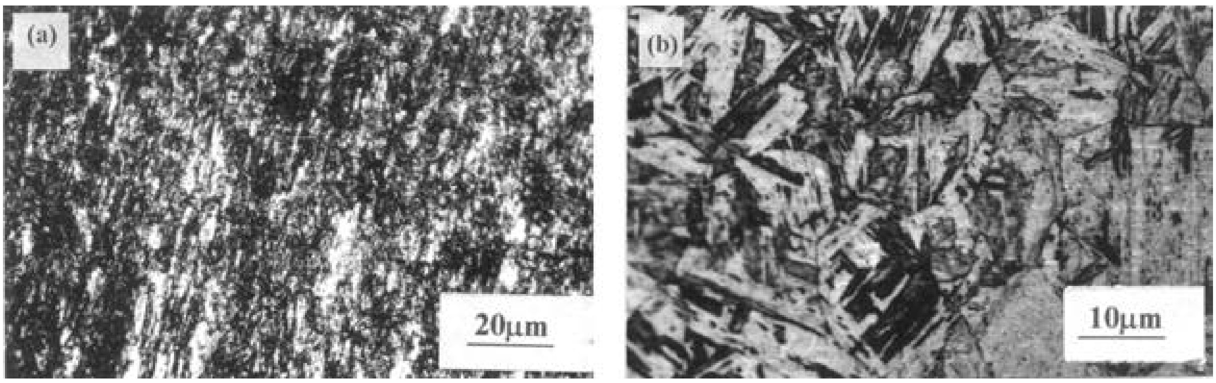

Figure 2. Microstructures after C80A treatment: (a) peak aged and (b) over aged.

various stages of aging are shown in figure 3. Coarse as well as fine precipitates were observed. The precipitates in case of high deformation (C80A) were, in general, more finely distributed. On the basis of Cu content, as determined from EDAX spectra, the precipitates were of two types: (i) having low Cu and (ii) high Cu content. At this stage of investigation it is not possible to say whether the two types of precipitates originated from the matrix differently, or the precipitates formed initially with low Cu content, which gradually increased as the particles grew in size. In pure Fe–Cu alloys it has been reported (Goodman *et al* 1973; Leslie 1981) that, on aging low Cu-bearing clusters are formed initially and as the particles grow, they become richer in Cu. It has been reported by earlier workers (Leslie 1981) that the Cu content in these precipitates could vary from 50 to 100 wt pct.

From figure 3 it is also seen that even in over aged condition (600°C, 1500 min), the precipitate distribution was much finer in specimens cold worked to high deformations (C80A) as compared to C25A. A dense dislocation substructure still existed in C80A. The TEM study also showed that aging of cold worked specimens was accompanied by subgrain growth leading to *in situ* recrystallization (figure 4). No significant formation and migration of high angle boundaries was observed. EDAX analysis showed that the subgrain growth process occurred by exodus of Cu atoms from some relatively unstable sub grains leading to their collapse.

3.4 *Scanning electron microscopy (SEM)*

The scanning electron micrographs of fracture surfaces of impact specimens in PA conditions are given in figure 5. In the case of TC25A and C25A treatments, which yielded high impact strength, the fracture surface showed

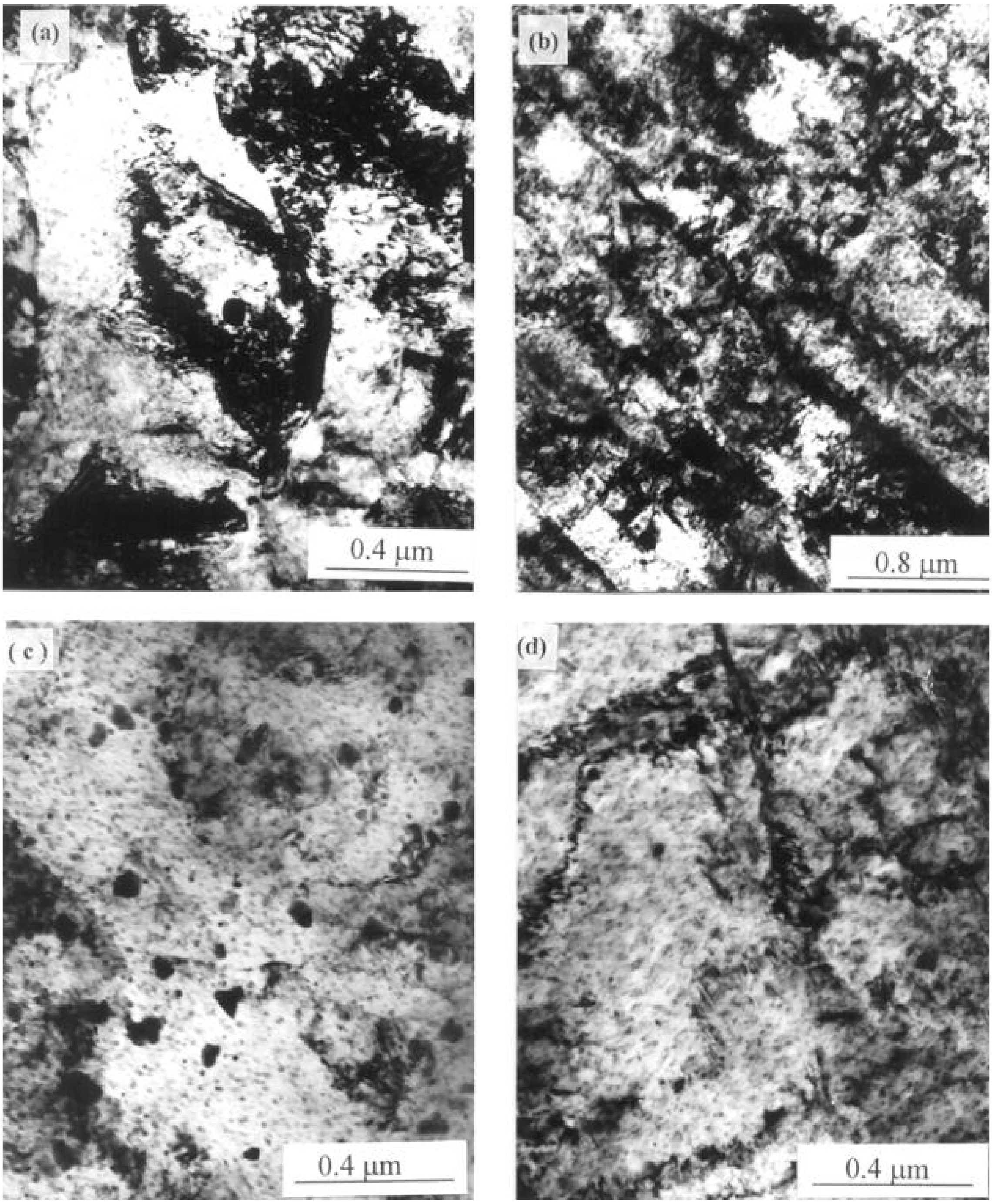

Figure 3. Transmission electron micrographs showing Cu precipitates: (a) peak aged, C25A; (b) peak aged, C80A; (c) over aged, C25A and (d) over aged, C80A.

very fine distribution of micro dimples indicating a high resistance to cleavage fracture. It was observed that, in general, the fracture occurred by formation and coalescence of micro voids. The nucleation of microvoids was assisted by the presence of precipitate particles or inclusions as shown by arrows. In case of treatments involving high deformations (C50A and C80A), the fracture surface indicated quasi-cleavage appearance which is in conformity with low impact strength values observed in these cases.

4. Discussion

The cold working and aging treatments may cause precipitation of calcium oxysulfides, manganese sulfides and niobium carbides in addition to formation of Cu-rich particles (Densley and Hirth 1998). Copper improves hardenability and introduces precipitation hardening. Although two precipitation hardening elements (Cu and Nb) are present in this steel, the occurrence of single age hardening peaks suggests that the precipitation hardening is primarily due to the presence of Cu. Extensive TEM study was conducted to follow the precipitation of Cu-bearing particles on aging/tempering of this steel. The study could not establish any relationship of coherent precipitates with the matrix. There is profuse precipitation of Cu-bearing particles on aging/tempering and TEM is unable to detect any coherent particle with clarity. Although coherent and incoherent precipitation in Cu-bearing HSLA steels was reported by some earlier researchers (Fox *et al* 1992; Mujahid *et al* 1998; Dhua *et al* 2001), they also could not provide sufficient TEM support for existence of coherent particles. Fox *et al* (1992) could not image Cu precipitates on tempering at 450°C and concluded that Cu precipitates must be of coherent type. Mujahid *et al* (1998) observed the presence of fine Cu clusters manifested by Moiré patterns. Dhua *et al* (2001) have reported formation of very fine Cu-bearing precipitates of 10–15 nm size on quenching and on tempering at 400°C. According to them these fine particles play the role of coherent particles in affecting the various mechanical properties.

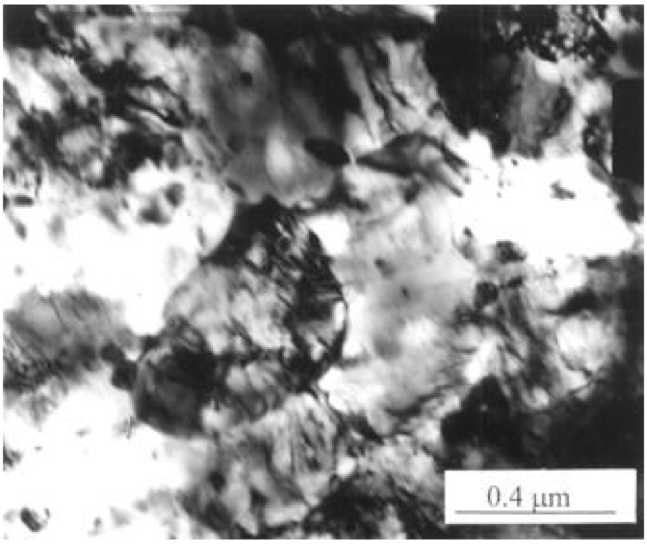

Figure 4. Transmission electron micrograph showing polygonization and *in situ* recrystallization (over aged, C80A).

The existence of a dislocation substructure in the presence of various alloying elements presents a very complex situation. As a result, it is observed that the formation

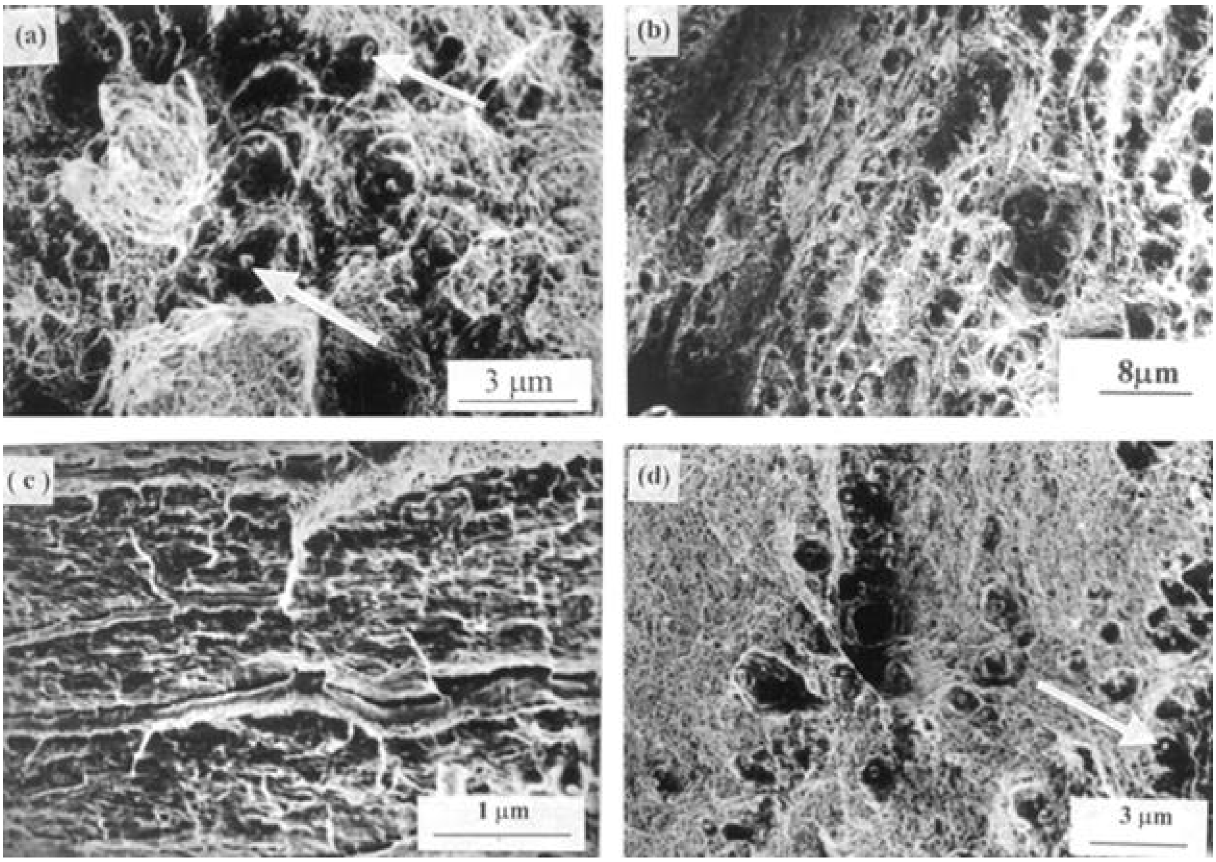

Figure 5. Scanning electron micrographs of impact fracture surface in peak aged conditions: (a) C25A, (b) C50A, (c) C80A and (d) TC25A.

of age hardening peaks is not necessarily due to the existence of coherent Cu precipitates alone. The TEM study also shows (figure 3) that in peak aged specimens coarse Cu precipitates also exist in addition to fine precipitates. The dislocation networks introduced by cold working retard the transformation of martensite laths into ferrite. In C80A treatment a very slow transformation of lath structure into ferrite occurs due to high density of dislocations. With increasing deformations, the dislocation-precipitate tangles become highly immobile leading to conditions favourable for *in situ* recrystallization. As a result, high angle boundaries are formed only locally and annealing occurs without their large-scale movement. This process leads to formation of acicular ferrite from lath structure, especially in the case of C80A treatment. The low rate of phase transformation also helps in slowing down the over aging process significantly. In addition to Cu precipitates, the dissolved Cu atoms in the matrix also retard the polygonization and subgrain coalescence during aging in various treatments. As seen in this study, the subgrain growth occurs by migration of dissolved Cu solute atoms from relatively unstable subgrains towards more stable ones.

Cold working prior to aging of precipitation hardening alloys has many practical applications. In general, cold working of Fe–Cu alloys and Cu-containing steels enhances the practical applications. In general, cold working of Fe–Cu alloys and Cu-containing steels enhances the precipitation of Cu; the time to reach peak hardness being reduced and higher hardness levels being achieved (Leslie 1981). At various degrees of cold working, the positions of hardening peaks may change due to competition between the recrystallization and aging processes. Accordingly, in this study, peak aging in C25A and C80A was observed after 5 min, in C50A after 15 min and in TC25A after 10 min.

Observation of low impact strength in peak aged conditions appears to be a matter of concern. The various treatments enhance the hardness and UTS, but significantly lower the impact strength in peak aged conditions. The C80A treatment, which results in the highest hardness and UTS, gives poorest charpy impact property. The TC25A treatment gives an optimum combination of tensile properties and impact strength. In the PA condition in all the treatments the charpy impact strength went down to very low values. The strengthened matrix is obviously detrimental for notch toughness. Mujahid *et al* (1998), Wilson *et al* (1988), Dhua *et al* (2001) and Hamano (1993) also observed similar dips in the CVN values in peak aged condition in their studies on HSLA steels. There have been various attempts to study the causes of poor impact strength in peak aged alloys (Sanders and Starke 1982; Duerig *et al* 1985; Higashi *et al* 1985). According to Hamano (1993) and Skoufari *et al* (1999), poor ductility and impact strength are due to existence of coherent precipitates in peak aged condition. The coherent precipitates are easily cut by moving dislocations, which leads to concentration of slip in only a few slip bands. This localization of deformation causes an early initiation of cracks and poor toughness. On over aging the phenomenal improvement in toughness is due to the over aged microstructure of partially recovered matrix and coarse Cu precipitates, which might have helped in arresting the propagation of cleavage cracks. In the C80A treatment the coexistence of high dislocation density and fine/coherent Cu precipitates in the highly cold worked matrix causes rapid nucleation and growth of matrix cracks leading to very low CVN values. Even on over aging the recovery and recrystallization in the matrix are extremely slowed down due to dislocation-precipitate pinning. Thus in C80A the charpy impact value does not significantly improve even on over aging up to 1500 min. The SEM study of impact fracture surfaces indicates that the fracture occurs by formation of microvoids at precipitate sites. The coherent/fine particles develop high stress concentrations at particle–matrix interfaces. These stresses, when combined with the stress fields of high density dislocation cells, cause localization of slip, nucleation of cracks and microvoids leading to poor impact strength. Accordingly, existence of coarse and incoherent precipitates causes an improvement in impact strength, as observed in over aged conditions.

All such microstructural features which trigger initiation of cracks and formation of microvoids, also affect the resultant tensile properties of the material. Miglin *et al* (1983) have attempted to correlate the effect of microstructural features on the impact strength through their effect on tensile properties. Figures 6(a) and (b) show respectively the relation between charpy impact strength with UTS and ductility. In these figures, data obtained from all the impact and tensile tests carried out at various stages of different treatments have been plotted. It is observed that the impact strength decreases with increase in UTS and increases with increase in ductility.

The influence of various mechanical properties on charpy impact strength is quite different in high strength precipitation hardenable materials. Since the hardness level in such materials is already quite high, any further

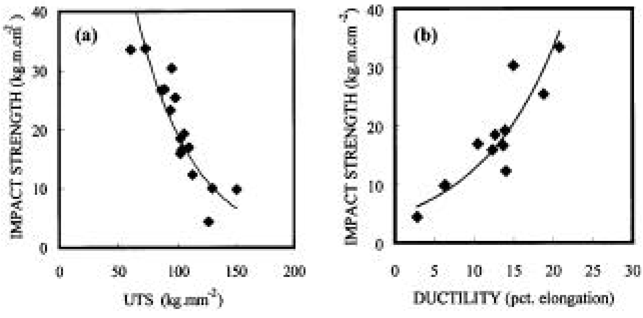

Figure 6. Variation of impact strength with (a) UTS and (b) ductility.

increase in hardness results in embrittlement and poor impact strength. In general, the charpy impact strength in such cases is promoted by (i) ease of plastic flow, (ii) low response to work hardening and (iii) increasing ductility to avoid localized build up of stresses. Thus low UTS, which favours (i) and (ii), and high ductility should cause improvement in impact strength as is seen in figures 6 (a) and (b). It has been observed by Gladman *et al* (1976) in Nb- and V-bearing HSLA steels that the ductile to brittle transition temperature increases by 0·3 to 0·5°C for each 1 MPa increase in yield strength. Miglin *et al* (1983) have shown in a HSLA steel that the J_{1C} for ductile fracture decreases monotonically with increasing strength level. In a recent study (Sharma 1996) on high strength martensitic stainless steel, it has been observed that the erosion resistance, which improves with increasing toughness, is also improved by lowering of hardness and increase in ductility.

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References

Densley J M and Hirth J P 1998 *Scr*. *Metall*. **39** 881

Dhua S K, Mukerjee D and Sarma D S 2001 *Metall*. *Mater*. *Trans*. **A32** 2259

Duerig T W, Allison J E and Williams J C 1985 *Metall*. *Trans*. **A16** 739

Fox A G, Mikalac S and Vassilaros M G 1992 *Speich Symp. Proc*. (Warrendale, PA: ISS) pp 1554–1561

Gladman T, Duilieu D and Mclvor I D 1976 *Microalloying' 75* (Niagara Falls, NY: Union Carbide Corp.) p. 32

Goodman S R, Brenner S S and Low Jr S S 1973 *Metall*. *Trans*. **A4** 2363

Gordon J A, Hirth J P, Kumar A M and Moody Jr N E 1993 *Metall*. *Trans*. **A23** 1013

Hamano R 1993 *Metall*. *Trans*. **A24** 127

Higashi K, Hirari Y and Ohnishi T 1985 *J. Jpn. Inst. Light Met.* **35** 353

Lee S, Kim B C and Kwon D 1998 *Metall. Trans.* **A24** 1133

Leslie W C 1981 *The physical metallurgy of steels* (NY: McGraw Hill Book Co.) p. 133

Miglin M T, Hirth J P and Rosenfield A R 1983 *Metall*. *Trans*. **A14** 2055

Mujahid M, Lis A K, Garcia C I and DeArdo A J 1998 *J. Mater. Eng. Perf*. **7** 247

Ranganathan S 1999 *Mater*. *Sci*. *Technol*. **15** 523

Sanders Jr T H and Starke Jr E A 1982 *Acta Metall*. **30** 927

Sharma M K 1996 *Effect of welding parameters on the structure, properties and silt erosion in 13/4 martensitic stainless steel*, Ph D Thesis, University of Roorkee, Roorkee

Skoufari-Themistou L, Crowther D N and Mintz B 1999 *Mater*. *Sci*. *Technol*. **15** 1069

Wilson A D, Hamburg E G, Colvin D J, Thompson S W and Krauss G 1988 *Proc. int. conf on microalloyed HSLA steels, Microalloying' 88* (Metals Park, OH: ASM International) pp 259–275