INVESTIGATION OF ULTRA-HIGH STRENGTH Fe/4Cr/0.4C MARTENSITIC
STEELS FOR IMPROVED TOUGHNESS

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ABSTRACT

Intergranular cracking in the as-quenched structures of vacuum melted Fe/4Cr/0.4C steel has been investigated in terms of the heat treatment conditions. It has been shown that the amount of carbon in solution and the martensite packet size are the two most important factors influencing cracking tendency. Non-conventional double heat-treatments have been designed in order to take advantage of higher austenitizing temperature and fine austenite grain size. These involved high temperature ($>1100^{\circ}\text{C}$) austenitizing during the first solution treatment followed by either interrupted quenching (M_s-M_f range) or isothermal transformation to produce lower bainite and a subsequent 900°C grain refinement treatment to produce quench crack-free microstructures.

Transmission and scanning electron microscopy and optical microscopy were used to characterize the microstructure. Tensile, CVN impact energy and plane strain fracture toughness tests were conducted to document the mechanical properties.

The predominantly lath martensite in all the as-quenched structures occurred in the form of packets. Higher austenitizing temperatures and coarse grain size resulted in increased amounts of twinned structures whereas low austenitizing temperatures and fine grain size resulted in large amounts of retained austenite.

The strength level of the as-quenched structures was somewhat higher than the reported strength level for similar medium carbon steels while the fracture toughness values corresponded to those reported earlier. Elimination of intergranular cracking produced toughness properties in quenched and tempered martensites which are far superior to those of lower bainitic structure of the same steel. The impact energy and K_{IC} fracture toughness values of the tempered steels are more than double those of lower bainitic steel at the same strength level. The possible role of secondary hardening is examined to explain the arrest in toughness properties upon tempering in the range 200-400°C. A very rapid recovery of impact energy and a somewhat less rapid recovery of elongation occurred after 600°C tempering. It is shown that the toughness properties of double treated steels are superior to those of single treated steels. A duplex structure consisting of appreciable amounts of lower bainite and martensite yielded the highest toughness but the influence of the former in lowering strength is unknown.

The effects of substructural twinning, undissolved carbide morphology, auto-tempering and retained austenite on toughness properties are discussed to explain the observed variation in mechanical properties with heat-treatment.

I. INTRODUCTION

One of the most economical ways of increasing the strength of steel is to raise its carbon content, and utilize heat treatments to form martensite. Unfortunately, above about 0.3-0.35%C, depending on the total alloy content, such high carbon steels are very brittle and in many cases cannot be used because after austenitizing and quenching the steels are already cracked (quench-cracking). Further treatments do not allow crack healing and in any case tend to lower the strength.

In a continuing program on alloy design of experimental structural steels^{1,2} the present investigation of the problem of intergranular quench cracking in Fe/4Cr/0.4C steels is taken up since a similar steel but with a lower carbon content viz., 0.35%, exhibits excellent tensile strength and fracture toughness without even tempering.³ The role of chromium is to achieve suitable hardenability. The increase of carbon from 0.35 to 0.4% has the potential of raising the yield strength to over 300,000 psi and the challenge is to achieve this while at the same time provide good toughness properties and elimination of quench cracking.

Although quench cracking in ferrous martensites, and especially in high carbon steels, has been the subject of many recent investigations,⁴⁻⁸ the phenomenon of intergranular cracking in medium carbon steels remains only partly understood. It has been established^{4,6,8} that microcracking in as-quenched high carbon steels is due to the stresses resulting from impingement of growing martensitic plates. Consequently, Brobst and Krauss⁴ concluded that the microcracking tendency can be decreased by refining the prior austenite grain size so as to limit the martensitic plate length, a parameter which has been directly linked⁶ to the

impingement stresses. It has also been established⁷ that the amount of carbon in solution influences the micro-cracking tendency viz, a higher carbon in solution resulted in more micro-cracking. However, there has been no such systematic investigation of the influence of grain-size, dissolved carbon and quenching medium on the intergranular quench cracking in the as-quenched structures of medium carbon steels. Clark et al.⁹ attributed the intergranular quench cracking to the segregation of impurities at the grain boundaries in a commercial low alloy En 30 steel. On this basis they proposed that a higher austenizing temperature coupled with a fast quenching rate of suppress any segregation of embrittling constituents leads to a decrease or elimination of quench cracks. However, these authors did not investigate the influence of quenching medium nor grain size on the cracking.

It is also well recognized^{10,11} that a mixed microstructure consisting of undissolved carbides is deleterious to toughness, particularly if the carbide phase is present as a network at the original austenite grain boundaries or at interlath α boundaries such as in upper bainite if the steel has inadequate hardenability. Thus, it appears that in order to achieve desirable fracture toughness properties which allow the steels to be utilized in engineering applications, it is essential to austenitize such that all carbides go into solution and, at the same time, in order to avoid intergranular cracking at high levels of carbon in solution (i.e., austenite), it is necessary to obtain a fine grain size. The choice of high purity vacuum melted materials minimizes problems encountered in current commercial steels arising from segregation of embrittling elements^{10,11} e.g., S, P, Sb, etc. Thus, the present study

aims at elimination of intergranular cracking in the as-quenched structures through variation in heat treatment practices.

Though a considerable amount of research in the past^{2,12,13,14} was directed to the design and development of strong tough new steels through often expensive adjustments in composition, comparatively little work was conducted on the effective use of modifications in conventional heat treatment practices to produce desired morphological and substructural features. The potential usefulness of finer austenite grain size in increasing the yield strength,¹⁵ decreasing the severity of segregation of embrittling constituents⁹ and lowering of the ductile brittle transition temperature^{15,16,17} are less thoroughly exploited in medium carbon structural steels. The inferior toughness properties of tempered martensites with C content $\geq 0.4\%$ as compared to lower bainite of the same steel was attributed to twinning and the consequent twin boundary carbide precipitation in martensites.¹³ However, no attempts were made to improve the ever worsening intercrystalline cohesion in martensitic structures due to transformation strains at these carbon levels and then compare the properties of these steels to those of lower bainite. Thus, it is also the aim of the present research to study the influence of heat-treatment variables on the resulting microstructure in a drive to impart reasonable toughness to steel at strength levels exceeding 300KSI.

II. EXPERIMENTAL PROCEDURE

A. Materials Preparation

The ternary alloy steels used in this investigation have a nominal composition of 4Cr, 0.4C, hereby referred to as Fe/4Cr/0.4C steel. The exact compositions (wt%) of the two vacuum melted alloys after homogenization are reported in Table I. Alloy 1 was supplied by Republic Steel Corporation in the form of 0.55 in. thick plate. Alloy 2 was vacuum melted into 20 lb heats and later on hot forged to 0.5 in. thick plates (for charpy specimens), 1 in. and 0.7 in. thick plates (for K_{IC} specimens) and 0.55 in. diameter rods (for round tensile specimens). After sand blasting to clean the surfaces the alloys were all homogenized at 1200°C in vacuum furnace for 48 hrs and furnace cooled. Intergranular quench cracking investigations were conducted using primarily alloy 1.

B. Heat-Treatment

All austenitizing treatments were carried out in vertical tube furnaces under argon atmosphere. Oversized tensile, charpy and K_{IC} specimens were cut from their respective stock and final machining was done under flood cooling after heat-treatment. All isothermal transformations were carried out by quenching into a salt pot located directly underneath the furnace containing agitated salt maintained at the required temperature. Room temperature quenching into agitated oil or water was also conducted in some of the heat-treatments. In most interrupted or isothermal treatments the specimens were allowed to cool to room temperature in air from their transformation temperature before they were subjected to further heat-treatments. A few tempering treatments

at 200°C, 400°C and 600°C for 1 hr were also carried out by immersing the specimen in a salt pot maintained at the required temperature and quickly quenching into water at the end of the treatment.

High temperature austenitizing was found to be necessary in order to take all carbides into solution. This was later confirmed by the absence of undissolved carbides in specimens austenitized above 1100°C for holding times in excess of 45 min. Decarburization studies after high temperature austenitizing treatments indicated that it was never greater than 40 μm from the exterior surfaces of the specimens.

C. X-Ray Analysis

X-Ray analysis was used to measure the percent of retained austenite in those heat-treated specimens wherein large amounts are most likely to occur. When available, specimens for x-ray analysis were cut from the fractured K_{IC} specimens, otherwise, separate specimens heat-treated for this purpose were used. After repeated polishing and etching in solution containing 100ml. H_2O_2 + 4ml HF to obtain a shiny surface, the specimens were scanned in Picker x-ray diffractometer with a fixed horizontal specimen stage using Cu $K\alpha$ radiation. The scan angle (2θ) covered was from 40° to 100° which includes the prominent (111) γ , (311) γ , (220) γ and (222) γ reflections.

Careful x-ray analysis for retained austenite in all the double treated specimens and in a few single treated specimens was conducted and it was concluded that the amount of retained austenite present was too small to be identified by this analysis.

D. Dilatometry

The phase transformation temperatures, viz, M_s , M_f , austenite start temperature A_s and austenite finish temperature A_f of the alloy steels were established by dilatometry and are reported in Table I. For alloy 1, the M_s temperature measurement was also carried out on a specimen which was subjected to initial interrupted quenching treatment. There was a 10°C rise in M_s temperature after the second treatment at 900°C . Thus, while there is a small increase in M_s temperature after the second treatment, the raise itself is not pronounced.

E. Mechanical Testing

1. Tensile Testing

The dimensions of the 1.25" gauge round tensile specimen are shown in Fig. 1A. Tensile testing was done at room temperature in a 300kip capacity MTS testing machine at a cross head speed of 0.04in/min.

2. Fracture Toughness Testing

Plane strain fracture toughness values were obtained by testing standard compact tension crack-line loaded toughness specimens shown in Fig. 1B. After heat-treatment at least 0.020 in. was removed from both the flat faces by grinding under flood cooling. Initially, a very conservative estimate of thickness requirements to satisfy ASTM specification for plane strain condition (thickness $\geq 2.5 \left(\frac{K_{IC}}{\sigma_y} \right)^2$) was made and thus, the first specimens for a number of treatments were much thicker than the second ones. The 300kip MTS machine was again used for fatigue precracking the specimens to a minimum crack length of 0.05 in. which were subsequently tested in the same machine to obtain the fracture toughness data.

3. Charpy Impact Testing

Separate Charpy specimens (oversized) were heat treated. After heat treatment the dimensions of the specimens were reduced to the standard specimen dimensions shown in Fig. 1C. A minimum of 2 and in most cases 3 Charpy-V-notch specimens were tested for each treatment and the tabulated results correspond to the average of these tests. In some plots the scatter band is also shown.

F. Metallography

1. Optical Metallography

Specimens for optical metallography were cut from Charpy specimens, mounted in Koldmount, abraded on silicon carbide papers down to 600 grit, and polished on 1 μ diamond abrasive wheel. For examination of intergranular cracking, freshly cut (i.e., interior) surfaces were examined. To reveal the intergranular cracking and martensite microstructure the specimens were etched in 2% and 5% nital solution. In order to reveal the prior austenite grain boundaries, etching was carried out in an etchant of 5gm of picric acid in 100 cc of water saturated with dodecylbenzene sulfonate.

2. Electron Metallography

Electron metallography of single treated specimens and some double treated specimens in relation to the investigation on intergranular cracking were conducted on alloy 1. The rest of the microstructural study was concentrated on alloy 2. No significant differences in microstructure between the two alloys can be expected since their compositions are very close. For the alloy 2, where available, thin foils for electron

microscopy were obtained from bulk, heat treated fracture toughness testing specimens after completion of testing. About 25 mils thick slices were cut from K_{IC} specimens. If K_{IC} specimens were not available, then a 50 mil thick specimen was heat-treated for this purpose. After removing the oxide scale, if any, the specimens were chemically thinned to less than 5 mils at room temperature in a solution of 4-5 ml hydrofluoric acid per 100ml H_2O_2 . 2.3 mm dia. discs were obtained by spark cutting which were then sanded down to about 2 mils thick. These thin foils were finally electropolished in a twin jet electro-polishing apparatus at room temperature using a chromic-acetic acid solution made of 75gm CrO_3 , 400ml CH_3COOH and 21ml distilled H_2O . The polishing voltage varied from 25 to 35 volts. Thin foils so obtained were then examined in a Siemens Elmiskop IA at an accelerating voltage of 100kV.

G. Fractography

Fractography was conducted on two kinds of specimens. For intergranular cracking studies fractography was conducted on tensile specimens of alloy 1. A few fractographic studies were also conducted on the K_{IC} specimens (alloy 2). A JSM-U3 scanning electron microscope operating at 25kV was used for this purpose. For the K_{IC} specimens, the fracture surfaces were preserved by masking with a tape during cutting. This tape was later dissolved in acetone.

III. RESULTS AND DISCUSSION

A. Investigation of Intergranular Quench Cracking

Conventional heat treatments involving high temperature austenitizing followed by room temperature quenching invariably lead to cracking in the medium carbon steel used in this study. In an effort to understand and characterize the nature of such cracking, the present study involving the influence of heat-treatment conditions on the intergranular cracking is taken up. Table II reports some of the results.

In order to study the influence of austenitizing temperature and quenching medium on the cracking tendency, austenitizing at temperatures of 900°C, 1100°C, 1200°C and 1290°C and quenching into either water or oil were carried out. Higher austenitizing temperatures (>1000°C) were found³ to be necessary to keep all Cr and C (present as chromium carbide) in solution which thus ensures higher hardenability. The mechanical property data on alloy 1 is reported in Table III. In this section, reference to mechanical property and microstructural data will be made in so far as it is relevant to the discussion of intergranular cracking. A detailed discussion of microstructure-mechanical property data will be presented in a later section.

In all the treatments, the cracking when present was intergranular with no evidence of intra-granular microcracking normally associated with high carbon steels. As should be expected, SEM fractographs on the fracture surfaces of quench cracked specimens revealed extensive intercrystalline cleavage as shown in Fig. 3. The poor cohesion at the grain boundaries is also evident from the grain boundary cracks occurring

normal to the fracture surface (Fig. 3a), although some dimpled rupture characteristic of ductile fracture can be seen at areas within the grain (Fig. 3b). Thus, the material shows the promise to yield reasonable toughness once good intercrystalline cohesion is established.

Intergranular cracking was observed in all the steels which had coarse austenite grain sizes when quenched to room temperature in either oil or water. Such cracking was widespread even in steels which were austenitized at 1200°C and 1290°C (Fig. 4a). Quenching in water resulted in more cracking than quenching in oil. Transmission electron microscopy of the steel austenitized at 1200°C resolved large amounts of substructural twinning (Fig. 4b) although optical metallography revealed that the morphology is of lath type occurring in the form of packets¹ (Fig. 4a). In order to understand the influence of reaustenitizing at the same temperature but for a shorter period on the cracking, specimen A [Table II] was reaustenitized at 1100°C for 1 to 15 minutes and oil quenched. In all the steels irrespective of the holding time during the second austenitizing treatment, cracking was present in the final structure. The striking feature of these treatments is that the crack appeared to occur at the boundaries of the grains resulting from the first treatment. Furthermore, SEM fractographs revealed that the cracks from the first treatment did not heal during the second austenitizing treatment. Figure 5a shows the fracture path along the grain boundaries of first treatment in specimen B. Nonetheless, there was a small rise in failure stress after the second treatment [Table III]. Concomitant with this, fractography revealed that there is a large increase in

quasi-cleavage as compared to almost 100% intergranular cleavage in single treated specimens. Figure 5b seems to indicate some kind of nucleation at the cracked boundary probably during re-austenitization. Nevertheless, as is evident from the figure, complete coherency is not established at the cracked boundary. Marder and Benscoter⁵ investigated the influence of tempering on the crack lengths in a 1.39wt. percent C steel and concluded that the number of microcracks decreases sharply above a certain tempering temperature ostensibly due to the bridging of cracks by carbide nucleation. In contrast to this observation on microcracks, bridging of the intergranular cracks is very rare even when the specimen is re-austenitized. Thus, intergranular quench cracking must be eliminated in the first cycle itself in a multicycle treatment.

In order to study the influence of refining the austenite grain size while keeping as much of C and Cr in solution on the cracking, two cycle austenitizing treatments were devised which involve a structure after the first treatment which is almost completely free from cracking. During the first cycle high austenitizing temperature (1100°C) was employed in order to keep all Cr and C in solution while a low austenitizing temperature was employed during the second treatment to refine the grain size. Two heat treatments were designed for this purpose. One of the treatments consisted of an interrupted-quench wherein the steel from the high austenitizing temperature is quenched to 265°C which is below M_s but above M_f . After holding for 3 minutes at this temperature, the steel is air cooled to room temperature. The second heat treatment consisted of isothermally transforming at a suitable temperature to obtain

a 100% lower bainitic structure.¹⁸ Research on the lower bainite in the same steel has shown¹⁸ no cracking occurs by this method. Also, in the temperature range of transformation, the only stable carbide is cementite¹⁹ which is much easier to take back into solution as compared to alloy carbides. Optical metallography did not reveal any grain boundary grooving in steels transformed by interrupted quenching (Fig. 6a). However, the somewhat poor cohesion at the grain boundaries is evident from the small and discontinuous grain boundary cracks as revealed by SEM fractography (Figs. 5c and 5d). Nonetheless, the intergranular cohesion had improved significantly (Fig. 6a). Electron metallography revealed that the microstructure consists of essentially lath martensite (Fig. 6b) with virtual absence of any isothermal transformation products. Also identified in the microstructure are extensive widmanstatten-type tempered carbides and fairly thick films of interlath stabilized austenite (Fig. 7). The material exhibited plastic yielding followed by failure at 270 ksi stress (Table III) which suggests that the critical crack propagation stress lies above the yield strength but below the tensile strength of the steel. SEM fractography (Figs. 5c and 5d) indicated a mixed fracture mode consisting of some dimpled rupture and widespread quasi-cleavage together with some grain boundary cleavage.

Starting with one of the two initial microstructures described above specimens were reaustenitized at 1100°C for varying periods of time from 5 to 15 minutes. Specimens which had undergone 5 minutes of reaustenitizing (HI in Table II) showed no evidence of intergranular cracking and the tensile property data for these steels is reported in

Table III. Because of the difficulties involved in controlling grain size at higher austenitizing temperature a low austenitizing temperature of 900°C was employed for the second austenitizing treatment during the subsequent investigation. As a result of the heat treatment investigations (see later sections), the most successful results were obtained starting with one of the two initial microstructures described above and re-austenitizing at 900°C for 30 min, viz., specimens C3 and D6 (Table II).

Rockwell hardness measurements and tensile strength data (Table III) indicated that most of the carbon is in solution after 30 min holding time. The resulting grain size corresponded to ASTM grain size number 8 (Fig. 8a) and the as-quenched structures were free from intergranular cracking (Fig. 8b). A mixed morphology consisting of a small number of plates extending right across the grains as well as parallel martensite laths were identified. Fractographs of specimens C3 and D6 did not reveal any intercrystalline decohesion. Extensive quasi-cleavage fracture with a small amount of dimpled rupture was present in the specimens C3 and D6. In addition, the double treated specimens showed a substantially greater resistance to intergranular cracking upon increasing the severity of quench, i.e., water quench after their second treatment as compared to single 900°C treated specimens.

B. Discussion and Development of a Qualitative Model of Intergranular Cracking

Micro-cracking at the impingement sites in the plate martensites of high carbon steels is totally absent in the present medium carbon steel. Also, it is important to note that there was no intergranular

quench cracking reported³ in a steel containing essentially 4%Cr and 0.35%C when a high temperature austenitizing at 1100°C was practised. The steel in the present investigation which has about 20% more carbon when similarly heat-treated resulted in extensive I.G. (i.e., Intergranular) cracking. Thus, any explanation for the cracking tendency should incorporate amount of carbon in solution as one of the most important factors influencing the cracking tendency.

Intergranular cracking persisted even in specimens subjected to very high temperature austenitizing, viz., at 1200°C and 1290°C followed by fast quenching in water to room temperature. Such a practice was considered⁹ to discourage any segregation of embrittling constituents at the grain boundary and the consequent intergranular cracking. Fast quenching apart from discouraging segregation of embrittling constituents also results in an increased amount of carbon in solution by reducing the carbon segregation to defects in the parent austenite.²⁰ In addition, rapid quenching results in decreased carbon precipitation in martensite, i.e., less auto-tempering by limiting the time for such process. The results suggest that segregation of embrittling constituents at the grain boundary can not be construed as the main reason for the Intergranular cracking in the present steel.

Davies and Magee⁶ considered qualitatively the mechanisms of microcracking and explain that microcracking results from the accommodation of strain at the tip of impinging plates. The authors argue that the strain energy due to the volume change accompanying formation of martensite increases as the transformed volume [proportional to (length)² X thickness of plate]. Then by analogy with a crack, this strain

distribution was proposed to be concentrated near the plate tip.

In contrast to plate martensite in high carbon steels, the martensite at the carbon levels present in the medium carbon steels occurs predominantly in the form of packets consisting of parallel laths, particularly when high temperature austenitizing is employed. It is observed that the packet size is strongly dependent on the prior austenite grain size. Thus, in a steel austenitized at 1200°C, the average packet size was much larger than the average packet size obtained after a 900°C austenitizing treatment.

It was conceived that it may be possible to decrease transformation strains by having retained austenite in the microstructure. Thus, it was attempted to trap thermally stabilized²¹ interlath austenite Fig. 7 in the interrupted quenched specimens C1. Although the Intergranular cracking tendency of the steel after this treatment is substantially reduced, the intercrystalline cohesion is still poor as evidenced from SEM fractographs Figs. 5c and 5d. In packet martensite adjacent laths are so nucleated that they try to minimize the total strain energy (hence the name "self-accommodating"²²) and thus, it appears that the unrelieved strain is concentrated in front of the growing lath in its long direction. Also, in the interrupted quenched steels, some of the strain can be relieved by the tempering that occurs simultaneously with the growth of martensitic laths. As will be discussed later, retained austenite films were identified in all the treatments including in specimen A which was cracked. Thus, it appears that retained austenite which occurs in very small quantities in the present steel might not play any significant role in intergranular cracking. Similar conclusions were made⁸ regarding the

role of retained austenite in microcracking in high carbon steels.

For martensitic inclusions in an untransformed austenite matrix, the transformation strain is predominantly pure shear. For the dilatational contribution to transformation strain, the elastic strain energy is given by Eshelby's treatment.²³ For an inclusion in an infinite matrix when the stress free strain is a pure dilatation, elastic strain energy is given by (assuming Poisson's ratio $\approx \frac{1}{3}$),

$$E_{\infty} = \frac{4}{9} \mu V (e^t)^2 \quad (1)$$

where "V" is the volume of the inclusion, μ is Lamé's constant and e^t is the dilatation strain. For a finite matrix (in this case grain), this equation will be accordingly modified²³ by a suitable function involving the ratio of the volume of inclusion and the matrix. Nonetheless, the above oversimplified equation does establish the dependence of strain energy on volume and the square of the dilatation strain. The precise strain distribution due to this dilatation is not known but because of the nature of formation of packet martensite ("self-accommodative", as discussed earlier) and possibly due to the retention of interlath films of austenite,²⁴ it can be taken to be concentrated near the tip of each growing lath in a packet ahead of its long dimension, i.e., at the packet boundary. Thus, in the specimen subjected to interrupted quenching C1, although there was partial relief of strain due to tempering, the packet size is still large with the result that intercrystalline cohesion was somewhat poor.

It is then possible to identify the two most important factors which influence intergranular cracking behavior, viz., (i) carbon in

solution (through e^t) and (ii) average packet size (through V) of the martensite as determined by the prior austenite grain size. By analogy with the micro-cracking in high carbon steels, continuous intergranular cracking in medium carbon steels can then be proposed to occur as a result of accommodation of strain due to the impingement of one growing packet on the other at the grain boundary. Whereas the strain and the corresponding stress are highly localized in the case of plate martensite, the strain in the packet martensite is distributed throughout the packet boundary and thus, results in continuous cracking. If the carbon in solution is fixed, then, the cracking tendency can be decreased only by limiting the packet size, i.e., prior austenite grain size. Specimens C3 and D6 which had most of the carbon in solution, showed no evidence of intercrystalline cracking because of their much reduced grain size.

C. Possible Influence of Other Parameters on I.G. Cracking

1. Effect of Segregants

Segregation of embrittling elements at the grain boundaries results in lowering of intercrystalline cohesion. Therefore, in the presence of appreciable quantities of segregants at the grain boundaries, intercrystalline cracking could be promoted at lower levels of carbon in solution and smaller martensitic packet sizes. It is suggested⁹ that dissolution of these segregants can be promoted by very high austenitizing temperatures due to enhanced thermodynamic driving force for such dissolution at these temperatures. However, such high austenitizing temperatures more importantly result in complete dissolution of any undissolved carbides as well as a very large packet size, the worst combination of factors for intergranular cracking. While single low austenitizing

temperature treatment (commercial practice) could be effectively used to reduce the packet size, one may not be able to take all alloy carbides in solution³ and thus, one may effectively lose some hardenability. Besides, there is the danger of segregation of embrittling constituents if present in the alloy and also embrittling due to the possible presence of undissolved alloy carbides at the grain boundaries. Something of this sort is discernible from a comparison of toughness properties of single cycle low temperature treated specimens (E2 in Table III) and the corresponding double treated specimens (C3 or D6 in Table III). Specimen E2 showed poor impact toughness and elongation.

2. Severity of Quench

As pointed out earlier, increasing severity of quench has three principal effects, viz., (i) decreased precipitation of carbon at defects in parent austenite,²⁰ (ii) decreased precipitation of carbides in martensite and (iii) increased thermal stresses. The role of factors (i) and (ii) in increasing the intergranular cracking should be apparent from the above discussion. Thus, it is easy to explain the observed increase of cracking in water quenched specimens. The degree of importance of thermal stresses is in doubt but the fact that the cohesion at the boundaries was still poor in the interrupted quenched specimens C1 even when the thermal stresses are substantially reduced indicates that these stresses can not be very important. Besides, thermal stresses are uniformly distributed (tensile at the outside and compressive inside) and can not explain why cracking takes place only along the prior austenite grain boundaries. Investigating on the microcracking in high carbon steels, Marder et al⁸ concluded that the severity of quench has no effect

on microcracking within the limits of commercial practice.

Finally, it is suggested that intergranular cracking is a manifestation of accommodation of strain resulting from the impingement of two growing martensitic packets at the grain boundary and other suggested parameters such as segregation of embrittling constituents and severity of quench can only aggravate but not initiate such cracking.

D. Structure and Morphology of Martensite

1. Single Treated Steels

a. 1100 and 1200°C Austenitization

As pointed out by Kelly and Nutting²⁵ there is a transition from dislocated lath martensite to twinned plate martensite as the carbon content of steel increases. In the present medium carbon steel the morphology and substructure can best be characterized as mixed in all the as-quenched martensitic structures. Figure 9 represents the typical mixed microstructures of steels austenitized at 1100°C (Figs. 9a and b) and 1200°C (Fig. 9c). Some undissolved carbides, presumably chromium carbides, could be identified in specimens austenitized for only 15 min at 1100°C (Fig. 9a). The minimum holding time at 1100°C to take all carbides in solution was found to vary with specimen thickness but after 45 min holding at this temperature no undissolved carbides could be located in 0.55" thick specimens (Fig. 9b). Twinned regions appeared more frequently in specimens austenitized at 1200°C (Figs. 9c and 4b). The morphology of martensite is predominantly lath type Fig. 4a and several laths containing twins were identified²⁶ (Fig. 4b). The preponderance of twinning in these steels was accompanied by a total absence of any

undissolved carbides. Interlath films of retained austenite have been identified in both 1100°C and 1200°C austenitized specimens.^{24,3} The presence of films of second phase, presumably retained austenite, is discernible in Fig. 9b at the lath boundaries (see also later sections).

b. Structure of 900°C Austenitized Steels

Figures 10a and b represent the typical structures of steel subjected to 900°C austenitization for 60 min (specimen E2). The microstructural investigation for this treatment was carried out on specimens cut from K_{IC} specimens. The essentially lath type martensite occurred in the form of packets (Fig. 10a). The substructure was predominantly dislocated with very little twinning (Fig. 10b). Undissolved carbides were frequently observed in the microstructure. The less frequent observation of twinning in this steel as compared to steel austenitized at 1200°C does not conform to the observation of Lai et al.²⁷ The martensite packet size was much smaller (Fig. 10a) in this steel as compared to 1100°C or 1200°C austenitized specimens. Figure 11 illustrates the unequivocal identification of interlath stabilized austenite. This phase was distributed uniformly throughout the microstructure. Most commonly, the morphology of this phase was in the form of interlath films although, in some places "blotches" or "islands" of this phase were also observed (Fig. 11c). The identification of extensive amounts of austenite in this low temperature treated specimen does not conform with the findings of earlier investigators.^{27,28} There was a negligible amount of auto-tempering in this steel. This observation can be reconciled with considering the decreased supersaturation due to undissolved carbides and the thinner K_{IC} specimen size used [See Table V]. There is a

wide variation in lath widths (Compare Figs. 11 and 10) from about 0.1 μ to about 2 μ .

c. Structure of Interrupted-Quenched Specimen (C1)

Isothermal holding at 265°C (in martensite phase field) resulted in essentially dislocated lath martensite (Fig. 6b) although a very small quantity of twinning was again identified. The twins were identified to run parallel to $\{112\}_m$ trace. Carbides formed in this steel were identified by habit plane analysis^{14,3,12} and were shown to be $\{110\}_m$ widmanstatten type cementite. Some twin boundary carbide precipitation was also noted. The identification of retained austenite in this steel is mentioned earlier (Fig. 7). Importantly, after 3 min holding at 265°C, there was no indication of any isothermal transformation products.

2. Structure of Double Treated Specimens

a. Structure of as-quenched Specimens C3 and D6

After 30 min holding at 900°C during re-austenitization, there was no apparent difference in the as-quenched microstructures of specimens C3 and D6 as shown in Fig. 12. Figures 12a and 12c clearly reveal the lath martensitic structure of specimens C3l and D6l^{*} [See Table V] respectively. Also, these figures clearly illustrate the occurrence of laths in packets. A packet boundary where the laths from two adjacent packets impinge can also be seen in Fig. 12 (note no indication of cracking). In thin foils made from thicker K_{IC} specimens, i.e. D6l and C3l, two types of carbides are identified. Figure 12b illustrates the presence of undissolved carbides (with spherical morphology) and the auto-tempered widmanstatten

* The first two characters identify the specific heat-treatment (Fig. 2) and the third numeral refers to the K_{IC} specimen for that treatment [Table V].

carbides with $\{110\}_m$ habit. It is evident that the 30 min holding time during re-austenitization at 900°C was not long enough for the 0.86 in. thick specimens to dissolve all the carbides produced during heating the massive K_{Ic} specimen. The profuse amount of auto-tempering in this specimen (Fig. 12b and 13) can be explained on the basis of two factors: (i) the degree of auto-tempering was observed to be a function of specimen thickness²⁹. (ii) Due to the decreased amount of carbon in solution, M_s temperature would have been raised promoting auto-tempering.¹ The amount of twinning in thicker specimens was also much lower (Fig. 12d).

With a view to confirm that the presence of undissolved carbides was due to the large thickness of K_{Ic} specimen, microscopic examination of specimen C32 (thickness 0.581") was carried out and the important findings can be summarized as virtual absence of undissolved carbides, a concomitant increase in the amount of twinning (Fig. 13c) and a substantial decrease in the amount of auto-tempering.

Retained austenite was observed both in thick and thin specimens although the austenite films appeared to be thicker in thinner specimens. Figures 14 and 15 illustrate the identification of austenite in these specimens. The difficulties involved in imaging austenite reflections from the as-quenched martensitic structures are pointed out elsewhere²⁴ but here it is sufficient to say that, often, even with the smallest aperture, closely lying carbide reflections can also be included (Fig. 14d) particularly if $\{111\}_\alpha$ reflections are used for imaging.

b. Structure of Double Treated Interrupted-quenched Specimens,
C7 and D5

With a view to investigate the possiblity of improving the

toughness of the steel while not suffering a serious drop in strength, interrupted quenching to a temperature in the M_s - M_f range was carried out in the final treatment (C7 and D5). Holding at 265°C for 3 min during the final quench resulted in a duplex structure consisting of lower bainite and lath martensite as shown in Figs. 16a and 16b. By careful visual examination the amount of lower bainite in the thicker specimens, i.e., D51 and C71 [Table V] is estimated to be about 40%. Fig. 16c reveals the presence of some twinning in these specimens although it is by no means widespread or present in large quantities. There was widespread precipitation of carbides in martensite and this will be discussed in a later section. Again, the thicker specimens contained scattered amounts of undissolved carbides. In contrast, thin foils made from thinner bulk specimens (C72) contained negligible amount of undissolved carbides. Concomitant with this, the amount of auto-tempering increased considerably within the martensite as shown in Fig. 16d and precipitation of carbides along the twin boundaries was also identified. These carbides are identified as cementite both from the diffraction information and also by trace analysis. In addition, there was a substantial drop in the amount of lower bainite in the thinner specimens. A rough estimate of this is put at about 10%. Within the martensite, frequently, adjacent laths in a packet were twin related²⁹ (Fig. 24).

Another significant observation is that while retained austenite could be unambiguously identified in foils made from thinner K_{Ic} specimens, the amount present in thicker specimens was too small to be detected.

3. Structure of Tempered Steels

A few tempering treatments of C3 and D6 at 200°C, 400°C and 600°C were carried out in an attempt to compare the toughness properties of these steels with those of lower bainitic steels¹⁸ at the same strength level. However, microstructural investigation was limited to 200 and 400°C tempered steels.

Tempering at 200°C resulted in substantial $\{110\}_m$ widmanstatten cementite precipitation shown in Fig. 17a. The retained γ from the as quenched structures remained untransformed (Figs. 17 b and c) after tempering at 200°C. These films of γ apparently prevented lath boundary carbide precipitation. Twin boundary cementite precipitation was, however, identified and is similar to that reported earlier³.

Upon tempering at 400°C, the intralath widmanstatten carbides appear to be dissolving (Figs. 18 a and b) while a new approximately spherical carbide (about 150Å large) precipitation at the lath boundaries from the existing austenite films (Figs. 18c, 18d and 18e) seems to have already started. This new carbide could not be identified in this investigation, but Seal and Honeycombe³⁰ identified it as Cr_7C_3 . They explained that the formation of Cr_7C_3 at the lath boundaries even before general intralath Cr_7C_3 precipitation is due to the higher carbon concentration of retained austenite. General precipitation of Cr_7C_3 within the grains subsequently takes place during the re-solution of cementite which provides the carbon.

E. Mechanical Properties

The mechanical properties of alloy 1 are reported in Table III and were earlier referred to in connection with the discussion on intergranular

cracking. The tensile and CVN impact properties of alloy 2 are reported in Table IV. It is seen that the mechanical properties of common heat-treatments of alloys 1 and 2 are similar although there was some general deterioration in ductility in alloy 2 particularly in the specimen C3. These differences can be related to the somewhat higher total alloy content of alloy 2 (Table I). Table V reports the valid plane strain fracture toughness results of some of the heat-treatments and Figs. 19 and 20 plot the K_{Ic} fracture toughness values against yield and ultimate tensile strength respectively of the steel. Figure 21 shows the variation of CVN impact energy with yield strength for specimen C3.

The effect of holding time at 900°C during re-austenitization of steels subjected to initial interrupted quenching on their tensile properties is shown in Fig. 22. From this, it is evident that a minimum of 10 min holding time is required (for 0.55" thick specimens) for keeping appreciable amounts of carbon in solution. Also, from the plot it appears that a 30 min holding time produces the best combination of properties.

The tempering behavior of steels C3 and D6 are shown in the plot of Fig. 23. It is clear that their behavior is similar except for a rapid increase in the elongation after 200°C tempering in specimen C3. However, both the specimens C3 and D6 exhibited an arrest in their elongation, CVN impact energy and little drop in yield strength in the tempering range 200°-400°C. Tempering in 400°-600°C resulted in a very rapid recovery in impact energy and elongation and a concomitant drop in yield and ultimate tensile strength values (Fig. 23). Rapid coarsening of the intra-lath and interlath Cr_7C_3 carbides in this temperature range

results in depletion in ferrite matrix of both Cr and C.³⁰ This results in rapid lowering of transition temperature^{31,32} and drop in strength level.³

From the fracture toughness data it is seen that there is no appreciable difference in the toughness properties of specimens C3 and C9. Increasing the austenitizing temperature to 1200°C during first treatment did not reflect in any pronounced changes in the properties the K_{IC} fracture toughness is almost the same as C3 while there is a small drop in tensile strength as compared to C3. In addition, it appears that once the cracking is eliminated during the first cycle, any variation in later treatments results in little variation in mechanical properties.

Interrupted quenching to 265°C in the M_s - M_f region resulted in a rather significant improvement in the elongation and K_{IC} fracture toughness values while suffering a drop in the strength properties. Thicker specimens containing a higher percent of lower bainite (C71 in Table V) showed the highest fracture toughness. However, it is doubtful that these fracture toughness values correspond to reported strength values obtained from thinner (~0.55" thick) specimens. Interrupted quenching to 200°C (specimen G5) did not improve the toughness of the steel over the directly room temperature oil quenched specimen. Quenching to this temperature would have resulted in 100% martensite and little tempering. Fracture toughness properties of initially bainitic steels (D series) were inferior to those of initially interrupted quenched specimens, although the difference is not marked. It is seen that the conventionally treated specimens, i.e., E2, showed the lowest toughness properties while their strength level compares with others.

F. Discussion on Structure of Martensite

The increase in the amount of twinning with higher carbon contents in solution have been reported earlier.^{25,1,3,13} There has been considerable debate^{14,33} over the factors responsible for promotion of twinned substructures in martensite. M_s temperature was suggested to be the governing factor for twinning, i.e., a lower M_s temperature results in a higher amount of twinning. However, such an approach failed to explain the apparent increase in transformation twinning with Co addition although it was shown¹⁴ that Co addition to steel raises M_s . In the absence of a general satisfactory explanation for twinning, a recent proposal by Thomas³³ that the magnitude of CRSS for slip and twinning govern the dominant inhomogeneous shear appears to be successful in explaining the observed trends. It is known that increasing carbon content in steels makes slip quite difficult in martensites in two important ways: (i) by forming strain centers in martensite wherein dislocations become pinned and (ii) by lowering the formation temperature of martensites and thus resulting in indirectly increasing the difficulty for slip. Although it is not known how increasing alloy content and lowering the formation temperature affects the CRSS for twinning, the presence of appreciable amounts of twinned substructures in all the as-quenched specimens can be explained on the basis of the difficulty of slip alone. The carbon content in this steel corresponds to a level said¹ to be the limit in order to avoid excessive amounts of twinning. Leslie et al³² pointed out that twinning is also inhibited by a dispersion of second phase particles. Thus, in the present steel, the substantial increase noted in the amount of twinning as the austenitizing temperature is raised from 900°C to

1200°C can be attributed to not only the increased alloying content in solution (due to better dissolution) but also to the absence of second phase particles.

In low alloy low carbon steels martensite occurs in the form of parallel laths with $\langle 110 \rangle_\gamma$ habit.²⁵ At low carbon levels, the strain energy due to transformation is small and hence adjacent laths may be separated by low angle boundaries.²⁹ As carbon content increases, the increased strain energy results in high angle and often twin related laths. The preponderance of twin related laths (Fig. 24) in some of the double treatments can be explained on this basis.

Another observation that needs to be explained is that lower bainite was observed in interrupted quenched specimens that have been double treated (C7 and D5) but not in the single treated interrupted quenched specimens (C1). The presence of undissolved carbides in the thicker specimens (C71, D51) would have effectively lowered the carbon content of γ and thus, promote faster lower bainite reaction kinetics. However, the presence of small percent of lower bainite even in thinner specimens wherein undissolved carbides were virtually absent requires that an additional parameter should be causing the rapid reaction kinetics for the isothermal products. The only major difference then, between single and double treated interrupted quenched specimens is their grain size. Thus, it is possible that the finer grain size of C7 would have promoted lower bainite formation and this effect of grain size on the kinetics of lower bainite is similar to that reported by Barford and Owen.³⁴ They also reported that nucleation of lower bainite occurs at the austenite grain boundary.

C. Formation of Carbides in Tempered Structures

There is a striking difference in the morphology of carbides in quenched and tempered steels, and interrupted quenched steels (C7, D5). The morphology of carbides in 200°C tempered steels is similar to that reported earlier^{3,14} and mainly consisted of $\{110\}_m$ widmanstatten carbides with some twin boundary precipitation. Twin boundary precipitation was also observed in all interrupted quenched specimens (Fig. 26). The carbides precipitated in specimens C71, C72 and D51 [Table V] were more numerous and hence direct identification by diffraction analysis was possible. As discussed before, in these steels frequently twin related laths were present. The carbides precipitated during isothermal holding occurred within the laths and surprisingly, showed a preference for twin related lath boundaries (Fig. 24). Twinning in the matrix occurred on $\{112\}_m$ and the orientation relationship between orthorhombic carbide (Fe_3C) and ferrite shown was

$$\begin{array}{l} [2\bar{1}0] \text{ carbide} \parallel [311] \text{ matrix} \\ (001) \text{ carbide} \parallel (112) \text{ matrix.} \end{array}$$

This is verified to correspond to one of the variants of the Bagaryatski orientation relation between ferrite and Fe_3C in tempered martensite as reported earlier.^{35,29} These carbides appear to be spheroidized in the 3 min holding at 265°C.

The unequivocal identification of orthorhombic cementite precipitated along twin boundaries in martensite becomes difficult in $\langle 110 \rangle$ matrix orientation since the twin reflections together with double diffraction produce a pattern which approximately superimposes on the carbide pattern

obeying Bagaryatski relation with the matrix. Only careful dark-field imaging of several reflections can establish the presence of such carbides. On the other hand if (001) carbide reflection is also present in the diffraction pattern, then these reflections cannot be accounted for by double diffraction and hence it is possible to establish uniquely the presence of cementite in such case from SAD pattern alone. Figures 25 and 26 illustrate these ideas clearly. Figure 25 shows SAD pattern and analysis from an area containing twinning and carbides wherein (001), (003) etc. reflections from carbides are also visible. The sequence of pictures shown in Fig. 26 confirm that the carbide present is cementite and the $\{112\}_m$ habit plane of these carbides agrees with that already reported³⁵ in the presence of twins. The Bagaryatski orientation relationship, i.e.,

$$\begin{aligned} (001)_{\text{Fe}_3\text{C}} &|| \{211\}_{\alpha} \\ [010]_{\text{Fe}_3\text{C}} &|| \langle 1\bar{1}\bar{1} \rangle_{\alpha} \\ [100]_{\text{Fe}_3\text{C}} &|| \langle 0\bar{1}\bar{1} \rangle_{\alpha} \end{aligned}$$

is shown to be obeyed in Figs. 25 and 26.

H. Stability and Formation of Retained Austenite

The presence of retained austenite in steels has been noted earlier^{37,38,30} but it is only recently³ that its systematic identification by electron metallographic techniques has been documented in a 0.35%C steel. In the present steel retained austenite has been observed in all the as-quenched structures regardless of grain size and austenitizing treatment.

However, in fine grained specimens viz., E2, C3, D6 its presence was noted more frequently and often in larger amounts than in coarse grained

steels austenitized at higher temperature (see Fig. 11). Similar variation in the amount of retained austenite with grain size was noted by Brobst and Krauss⁴ in a 1.22C steel.

In explaining the stability of austenite in the present steel the three possible stability mechanisms already proposed can be investigated viz., (i) chemical stability due to partition of alloying elements and the resultant enrichment of austenite, (ii) thermal stabilization^{20,21} arising from precipitation of carbon at dislocations in γ due to slow cooling and (iii) mechanical stabilization^{25,39} due to production of high density of tangled dislocations in γ to accommodate the transformation strain. In the present as-quenched structures, the carbon supersaturation is largely relieved by carbide precipitation within the laths (widmanstätten precipitation) and the time to reach interlath boundaries to dissolve in austenite may not be sufficient during quenching. Besides, if the austenite is chemically stabilized, then, it should undergo transformation if the temperature is sufficiently lowered. Even after refrigeration in liquid N_2 there is no noticeable decrease in the amount of retained austenite in the present steel (Fig. 15a). A similar observation was made in Fe-4Cr-0.35C steel.³ Refrigeration in liquid N_2 should also result in a substantial decrease in the amount of retained austenite if the major stability of γ is thermal stabilization. On this basis it is possible to conclude that the major stability of γ in this case is due to mechanical stabilization. Thus, if a transformation yields a duplex structure of lower bainite and martensite, (such as in specimen C71) then, the transformation strains are markedly lowered and

likewise substantial reductions in the amount of retained γ can be expected which was the case.

The Kurdjumov-Sachs orientation relationship between martensite and austenite observed in all steels is shown in Fig. 27.

IV. CORRELATION OF MICROSTRUCTURE AND MECHANICAL PROPERTIES

Lower bainitic structure represents one of the most uniform mixtures of carbide and ferrite. The initial interrupted quenching treatment (C1) resulted in considerable widmanstatten type precipitation of carbides. This again is a uniform mixture of carbides and ferrite, although in this case the carbides are much thinner. Thus, with either of the two initial structures, the preferential nucleation of austenite at carbides⁴⁰ would be high resulting in a uniform, fine austenite grain size at the end of second low temperature austenitizing treatment. Since in both cases the carbides are Fe_3C and not chromium carbide¹⁹ the dissolution rate in γ would be much faster. Thus, it is not unexpected that the tensile properties of steels C3 and D7 are almost identical.

The strength level of these specimens is somewhat higher than the reported strength level of similar medium carbon steels.²⁷ This can not be attributed to the presence of Cr since Cr is known to be an ineffective solid solution strengthener in ferrite. In the present steels partial contribution to yield strength could have resulted from their fine grain size.¹⁵ Although Cr is an ineffective solid solution strengthener, it does promote substructural twinning.³ In the present investigation twinning was present in all the steels although its amount varied from treatment to treatment. It was observed quite frequently in steels C3 and D7. Many earlier investigators^{25,14} have already pointed out the strengthening resulting from twinning. Similarly, the deleterious effects of twinning on toughness properties of steel have been discussed by some authors.^{1,25} However, as pointed out by Raghavan²⁹, to

produce a higher percent transformation twins the alloy content of the steel has to be increased with the result it becomes difficult to isolate the effects of twinning alone on one hand and the effect of these additional alloying elements on the slip characteristics of ferrite on the other hand. Indeed, Raghavan⁴¹ in a later study concluded that twins per se did not have any influence on toughness of steel. Nonetheless, if the martensite deforms by twinning, it is clearly documented that the deformation twins interact with transformation twins in the matrix and result in microcracks at the junctions.⁴² If twinning is indeed detrimental to toughness properties of steels in the medium carbon steels, then the properties of martensitic microstructure in these steels should be far inferior to the properties of the same steel in lower bainitic condition.¹³ However, as is clear from the plots of Figs. 19 through 21, the mechanical properties of martensitic microstructure are far superior to the single treated coarse grained lower bainitic steel and the CVN impact energy and K_{IC} fracture toughness values of martensitic microstructure are more than double the corresponding values for lower bainite at the same strength level. Before specific conclusions can be made, one other important factor has to be considered. The plotted properties of Figs. 19 through 21 correspond to fine grained martensite whereas the plotted values for lower bainite correspond to very coarse austenite grain size.¹⁸ Following Cottrell¹⁶ and Petch's¹⁷ analysis the Ductile Brittle Transition Temperature (DBTT) can be related to metallurgical variables through the following identity,

$$DBTT \propto \ln \left\{ \frac{1/2}{\frac{Bd}{\beta G \gamma} - \frac{K_y}{K_y Z}} \right\} \quad (2)$$

where 'd' is the prior austenite grain diameter, K_y is the Hall-Petch slope, G is the shear modulus, γ is the surface energy and B and t are constants. On the basis of this identity, it is possible that because of the much finer grain size in specimen C3, the impact energy values correspond to upper shelf values whereas for the coarse grained lower bainite, the CVN energy values are reported¹⁸ to correspond to lower shelf values. However, the transition temperature and upper shelf values for the lower bainite were also determined and the upper shelf energy corresponded to about 13 ft. lbs. Thus, even if we use these upper shelf values for lower bainite, the toughness properties of these steels are far inferior to those of tempered martensite in the present Fe/4Cr/0.4C steel for a given strength. At about the carbon levels where lower bainite is shown to be superior to tempered martensite by earlier investigators¹³ intercrystalline cohesion at the prior γ grain boundaries becomes increasingly poor. The crack propagation will in those cases tend to be along this low energy crack path, i.e., intergranular. Thus, unless efforts are made to re-establish this cohesion in martensitic steels, any comparison made on the basis of internal microstructure will be in doubt.

It has been shown³⁰ that secondary hardening in the temperature range 250 to 400°C is a function of both carbon and Cr content of the steel. At 0.4%C it is suggested³⁰ that significant secondary hardening occurs at as low as 3% chromium content and that even higher Cr and C contents are needed in order to actually observe a raise in yield strength upon tempering in this temperature range. In the present

investigation there was an arrest in the CVN energy and elongation values upon tempering in 200-400°C range (Fig. 23) and it is argued that secondary hardening is not quite prominent in the present steel on the basis that there was actually a small drop in yield strength in this temperature range. In this tempering range it is observed that while the intralath widmanstatten carbides appear to be dissolving, there was a simultaneous precipitation of spheroidized carbides, presumably Cr_7C_3 , at the interlath boundaries. This might mean that the total alloy content of ferrite remains unaltered in this temperature interval and thus the DBTT is maintained.

The surface energy term γ in the relation (2) above can be increased by elimination of carbides at the boundaries. Specimens E1 and E2 which are single treated at 900°C showed much inferior CVN energy values as compared to specimen C3 even though their grain sizes are very much alike. In the former, grain boundary decoration by precipitates, presumably undissolved alloy carbides has been noted. On the same basis interlath precipitation of carbides can partially account for the arrest in CVN energy values in the 250-400°C tempered specimens.

As has already been pointed out, a very small percent of retained austenite has been conclusively identified in all steels. While it is possible that it could have a positive contribution to the CVN energy values, it is not clear at the moment how exactly the fracture toughness properties of various treatments are influenced by such small quantities of retained austenite. Webster²⁸ reported a positive improvement in the K_{Ic} values with increasing amounts of retained austenite based on the observation that the crack path is effectively increased. However, one

has to be very cautious in directly using these results since the amount and morphology of austenite could be of very great importance. The morphology of austenite in the present case, i.e., interlath films, is quite different from the morphology reported by Webster²⁸ wherein islands of austenite trapped in between growing martensite plates was the main morphology. While the austenite is tested to be stable to subzero refrigeration, it seems likely that it is unstable to externally applied strains. It was not attempted to follow the transformation characteristics of this austenite as the crack propagates during K_{Ic} testing and it seems that the variables are so numerous and the available information on the characteristics of this γ is so little that no valid conclusions can be made regarding its influence on toughness properties.

Figures 28a and b illustrate the fractographs (from K_{Ic} specimens) of specimens D6 and C7 respectively and a mixed fracture mode consisting of packets of high energy ductile rupture and low energy quasi-cleavage can be seen. The amount of ductile rupture is higher in specimen C7 (Fig. 28b).

V. CONCLUSIONS

Based on the present study on Fe/4Cr/0.4C steels, the following conclusions can be drawn:

1. It has been established that carbon in solution and the martensite packet size are the two most important factors influencing intergranular cracking in the present steel. An increase in either of them increases the probability of such cracking.
2. Since small amounts of mechanically stabilized austenite of inter-lath film morphology have been identified in the crack riddled as well as crack free structures the role of this phase in intergranular cracking is suggested to be of small significance. Precipitation of carbon by either deliberate interruption in quenching or auto-tempering reduces the impingement stresses.
3. Conventional grain refining involving repeated austenitizing and quenching cannot be used to refine the grain size since intergranular cracks produced after the first high temperature treatment can not be eliminated during subsequent heat-treatment.
4. Based on this research, heat-treatments have been developed which ideally combine the beneficial effects of higher austenitizing temperature and fine grain size. In addition to eliminating intergranular cracking, good mechanical properties are also obtained utilizing these treatments.
5. The amount of twinning in single treated specimens increased with austenitizing temperature due to better dissolution of alloy

carbides at higher temperature. However, retained austenite was observed more frequently in fine grained, low temperature treated specimens.

6. The strength level of the crack-free as-quenched structures is somewhat higher than that of similar 0.4C martensitic steels while the fracture toughness values corresponded to those reported earlier.

7. The toughness properties of double treated specimens are superior to those of single conventionally treated specimens. The impact toughness was more than doubled while there was about 25% gain in K_{IC} fracture toughness of some of the double treatments.

8. Elimination of intergranular cracking produced toughness properties in quenched and tempered martensites which are far superior to those of lower bainitic structure of the same steel. The impact energy and K_{IC} fracture toughness values of the tempered steels are more than double those of lower bainitic steel at the same strength level.

9. Tempering in the range 200-400°C resulted in an arrest in the impact energy and elongation, and tempering at 600°C resulted in a very rapid recovery of these properties.

10. Once intergranular cracking is eliminated, increasing austenitizing temperature during the first solution treatment did not result in any improvement in mechanical properties.

11. A duplex structure consisting of appreciable amounts of lower bainite and martensite appears to yield good fracture toughness while lower bainite if present in small amounts (5-10%) did not improve toughness.

12. The quenched and tempered steels in general appeared to possess better combinations of properties than steels interrupted quenched to the same strength level.

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TABLE CAPTIONS

Table I.	Compositions (wt%) of the two vacuum melted alloys used in this investigation together with their transformation temperatures.
Table II.	Intergranular quench cracking characteristics of various heat-treatments.
Table III.	Room temperature mechanical properties of alloy 1.
Table IV.	Room temperature tensile and impact (CVN) energy properties of alloy 2.
Table V.	Room temperature fracture toughness properties of alloy 2.

TABLE I.

Alloy	Cr	C	Fe	M _B (°C)	M _F (°C)	As (°C)	Af (°C)
Alloy 1	4.2%	0.42%	Balance	320	~220	810	860
Alloy 2	4.6%	0.43%	Balance	310	~220	-	-

TABLE II

Specimen	Heat-treatment Description	ASTM grain size no.	Presence of g.b. cracking (Nital etch)	Fractography
A	1100°C-60min+oil	<0.5	yes	Intercrystalline cleavage.
B	A+1100°C-15min+oil	-	yes	Some quasi-cleavage
F	1200°C-50min+oil	<0.0	yes	-
G	1200°C-50min+water	<0.0	yes	-
E2	900°C-60min+oil	6.5	no	-
C1	1100°C-60min-265°C-3min-Aircool	<0.5	no grooving was observed	small gr. bd. cracks are detected
H1	C1+1100°C-5min+oil	3.3	no	-
C3	C1+900°C-30min+oil	8	no	largely quasi-cleavage
C7	C1+900°C-30min+265°C-3min-Aircool	8	no	quasi-cleavage + some dimpled rupture
D6	1100°C-60min-360°C-15min-Aircool-900°C-30min+oil	8	no	largely quasi-cleavage
D5	1100°C-60min-360°C-15min-Aircool-900°C-30min-265°C-3min-Aircool	8	no	quasi-cleavage + some dimpled rupture

TABLE III

Specimen	0.2% offset yield strength KSI*	UTS KSI*	% Elongation (1.25in gauge length)	CVN Impact Energy Ft.lbs°	Rc Hardness
A	200	← quench cracked →			60
B	235	← quench cracked →			-
C1	210	270	1.0 ^Δ	9.4	53
H1	235	315	0.6 ^Δ	-	-
C3	250	335	6.0	6.5	58
C7	225	300	7.0	10.5	53
D6	280	335	2.5 ^Δ	9.0	57
D5	225	310	7.0	10.5	53
E2	270	330	2.0 ^Δ	4.0	58

* 1 KSI = 6.89 MNm⁻²

° 1 ft-lb = 1.36 Nm

ΔFailed before the onset of necking.

TABLE IV

A. Initial Interrupted Quenching to $M_s - M_f$ Range

Heat-treatment	0.2% offset Yield Strength (KSI)	Ultimate Tensile Strength (KSI)	% Elongation in 1.25" gauge		CVN Impact Energy (Ft-lbs)
			Uniform	Total	
C3, as-quenched	260	340	1.5	1.5	10.0
C3, 200°C tempered	236	297	4.4	9.0	15.0
C3, 400°C tempered	222	245	2.7	9.5	15.5
C3, 600°C tempered	128	148	3.2	10.5	80.0
C7	233	316	3.8	6.1	11.6
C9	265	330	1.2	1.2	--
G6	215	307	4.4	7.8	--
G5	247	326	3.6	5.1	--
K2	255	324	3.0	3.5	6.1

B. Initial Lower Bainitic Treatment

D6, as-quenched	260	330	2.5	2.5	6.8
D6, 200°C tempered	243	295	2.8	6	15.0
D6, 400°C tempered	225	246	1.6	6.5	15.5
D6, 600°C tempered	128	146	3.5	11.0	80.0
D5	242	302	2.6	4.4	11.0

C. Single Cycle 300°C Austenitizing Treatment

E1 (30 min holding)	245	330	2.8	2.8	3.8
E2 (60 min holding)	260	330	1.8	1.8	3.7

TABLE V

Heat-treatment	Test Specimen #	Thickness of Specimen (inches)	P_m/P_Q	Plane Strain Fracture Toughness K_{IC} (KSI-in $1/2$)	$K_{IC}/\sqrt{Y \cdot s}$ (in $1/2$)
C3	C31	0.862	1.00	42.3	0.16
	C32	0.581	1.00	33.00	0.13
	D61	0.830	1.11	36.5	0.14
C3 (200°C tempered)	T1C31	0.861	1.00	52.5	0.22
	T1C32	0.581	1.00	51.0	0.22
D6 (200°C tempered)	T1D61	0.830	1.02	52.1	0.21
	T2C31	0.862	1.00	61.2	0.28
C3 (400°C tempered)	T2D61	0.832	1.00	58.1	0.25
	C71	0.862	1.00	77.6	0.33
C7	C72	0.581	1.00	39.1	0.17
	D51	0.830	1.00	58.8	0.24
K2	K21	0.581	1.00	33.6	0.13
	K22	0.581	1.03	32.9	0.13
G5	G51	0.581	1.00	33.2	0.13
	G52	0.581	1.00	34.7	0.14
C9	C91	0.581	1.00	34.8	0.13
	C92	0.581	1.00	36.0	0.14
G6	G61	0.581	1.00	45.8	0.21
E2	E21	0.504	1.00	27.8	0.11
	E22	0.504	1.00	28.5	0.11

FIGURE CAPTIONS

- Fig. 1. Various test specimens used to obtain mechanical property data.
- Fig. 2. Schematic description of various heat-treatments.
- Fig. 3. SEM fractograph of specimen A revealing intercrystalline fracture mode.
- Fig. 4. (a.) Coarse prior austenite grain size and the resultant intercrystalline cracking. Note large martensite packets. (b.) Electron micrograph of the same steel showing mixed, i.e., twinned and dislocated substructure. (c.) Schematic illustration of heat-treatment.
- Fig. 5. (a.) Fractograph of specimen B revealing intergranular cleavage along the grain boundaries produced from first treatment. (b.) Fractograph of the same specimen indicating some kind of nucleation at the cracked boundary reminiscent of the first treatment. Fig. 5c and d. Fractographs of specimen C1 revealing the somewhat poor cohesion at the grain boundaries.
- Fig. 6a and b. Optical and electron micrographs of specimen subjected to interrupted quenching. No grain boundary cracks can be found in (a.). (c.) Schematic description of heat-treatment.
- Fig. 7a and b. Identification of retained austenite in Specimen C1. (a.) is the BF picture and (b.) is DF picture obtained from austenite reflection, whereby the interlath austenite films reverse contrast. Tempered $\{110\}_M$ carbides can also be seen in (a.).
- Fig. 8a. Optical micrograph of specimen D6 etched in nital to reveal microstructure. (b.) Optical micrograph of the same specimen etched to reveal prior austenite grain boundaries.

Fig. 9. (a.) Mixed microstructure of the steel austenitized for 15 min at 1100°C followed by iced brine quenching. Twinned plates and some undissolved carbides (indicated by arrows) can be seen.

(b.) Microstructure of the same steel austenitized for longer time, i.e., 45 min at the same temperature and oil quenched.

(c.) Mixed microstructures of the steel austenitized at 1200°C for 45 min followed by oil quenching. Preponderance of twinned martensite can be seen.

Fig. 10a and b. Typical microstructures of specimen E2.

Fig. 11. Identification of retained austenite in specimen E2. (a.) is the BF and (b.) is the DF obtained from an austenite reflection whereby the austenite phase reverses contrast. (c.) is the DF of austenite reflection from an adjacent area. (d.) is the SAD pattern revealing fairly intense austenite reflections.

Fig. 12a and c. Lath martensitic structure of specimens C3 (a.) and D6 (c.). (c.) also illustrates a packet boundary where two adjacent packets impinge.

Fig. 12 (b.) The two kinds of carbide morphologies observed in the as-quenched structures of C3 and D6. Spherical undissolved carbides (marked by arrows) and widmanstätten $\{110\}_M$ auto-tempered carbide platelets can be clearly seen.

Fig. 12 (u.) Electron micrograph showing presence of twinning in specimens C3 and D6.

Fig. 13a and b. BF and DF pictures revealing the $\{110\}_M$ widmanstätten cementite platelets in as-quenched structures of specimens C3

and D6. (c.) Large amounts of substructural twinning present in thinner C3 specimens can be seen. Undissolved carbides are virtually absent in these steels.

Fig. 14. Identification of retained austenite in specimens C3 and D6.

The BF and DF pictures of (a.) and (c.) reveal the presence of thin films of stabilized austenite at the lath boundaries. The BF and DF pictures of (b.) and (d.) were obtained from a different area of the specimen. Closely lying austenite and carbide reflections were imaged in Fig. (d.) whereby both interlath γ and intralath $\{110\}_M$ widmanstätten cementite platelets reverse contrast.

Fig. 15 (a.) Presence of untransformed γ in specimen C3 after refrigeration in liquid N_2 . (b.) and (c.) BF and DF pictures of another area of as-quenched specimen D6 revealing thin films of austenite. The SAD pattern (Fig. 15b insert) reveals presence of faint streaking in a direction normal to γ films indicating that these films are indeed thin.

Fig. 16a and b. BF and DF pictures showing lower bainite structure in foils obtained from thicker K_{IC} specimens of C7 and D5 treatments. The amount of lower bainite was estimated to be about 40% in thicker specimens and about 10% in thinner specimens. (c.) Presence of small amount of substructural twinning in martensites of interrupted quenched specimens C7 and D5. (d.) BF picture revealing extensive tempering in martensite and the numerous, thick $\{110\}_M$ widmanstätten cementite platelets in thinner specimens of treatments C7 and D5.

- Fig. 17. These three figures illustrate typical microstructure of steels C3 and D6 which were tempered at 200°C. The BF picture (a.) illustrates substantial $\{110\}_M$ widmanstatten carbide precipitation whereas the BF and DF pictures of (b.) and (c.) reveal untransformed austenite in these specimens.
- Fig. 18. These five figures illustrate typical microstructure of steels C3 and D6 which were tempered at 400°C. The intralath $\{110\}_M$ carbide platelets in the BF and DF pictures of (a.) and (b.) appear to be dissolving and the interfaces of the carbide platelets are quite irregular. Fig. (c.) illustrates lath boundary carbide precipitation (indicated by arrow), presumably, from the break-up of existing austenite films. Fig. (d.) and (e.) also reveal preferential precipitation of spherical carbides at lath boundaries.
- Fig. 19. Plot of K_{IC} fracture toughness vs. yield strength of some of the treatments.
- Fig. 20. Plot of K_{IC} fracture toughness vs. ultimate tensile strength of some of the treatments.
- Fig. 21. Plot of CVN impact energy vs. yield strength of specimen C3.
- Fig. 22. Effect of holding time at 900°C during reaustenitization on the tensile properties of initial interrupted quenched specimens.
- Fig. 23. Plot of mechanical properties of C3 and D6 vs. tempering temperature.
- Fig. 24. Carbide precipitation in martensite in interrupted quenched steels C7 and D5. Fig. (a.) is the BF, and (b.) is the DF from a twin reflection. (c.) is the DF from a carbide reflection. (d.) is the SAD pattern. (e.) is the analysis of the

pattern. The twinning plane of the twin related laths is $\{112\}_M$.

Fig. 25. SAD pattern (a.) obtained from an area containing $\{112\}_M$ twinning and carbides and analysis (b.) Somewhat strong extra spots are due to the superposition of double diffraction from twinning and carbide reflections. The faint extra spots are carbide reflections as indexed in (b.). This pattern is typical of all the double treated steels.

Fig. 26. Twin boundary precipitation in interrupted quenched steels C7 and D5. (a.) is the BF, (b.) is the DF from the superposition of carbide and double diffraction (from twinning) spots whereby the twin boundary carbides reverse contrast and twins can be seen in the background, (c.) is the DF from superposition of twin and carbide reflections whereby the twins appear in better contrast, (d.) is the DF of carbide and double diffraction superimposed spot of an adjacent area and (e.) is the SAD pattern.

Fig. 27. The common orientation relationship observed between lath martensite and retained austenite in all the steels. The SAD pattern (a.) and the analysis (b.) show the Kurdjumov-Sachs orientation relationship viz., $[110]_\gamma \parallel [111]_M$, $(1\bar{1}1)_\gamma \parallel (1\bar{1}0)_M$ observed between bcc martensite and fcc γ .

Fig. 28. Fractographs of specimens D6 (a.) and C7 (b.).

A. ROUND TENSILE SPECIMEN

B. FRACTURE TOUGHNESS SPECIMEN

C. CHARPY V-NOTCH IMPACT SPECIMEN

XEL 754-6176

Fig. 1

XBL754-6177

Fig. 2

XBB 746-4216

Fig. 3

XBR 746-4219

Fig. 4

RE-AUSTENITIZED AT 1100 °C - 15 MIN - OIL QUENCH

UTS 235 KSI

TREATMENT 2A

UTS 270 KSI

NBB 7410-7017

Fig. 5

XBB 746-4218

Fig. 6

XBB 755-3490

Fig. 7

XBB 755-3479

Fig. 8

XBE 755-3492

Fig. 9

XBB 755-3491

FIG. 10

Fig. 11

XBB 755-3484

Fig. 12

XBB 755-3489

Fig. 13

XBB 755-3485

Fig. 14

Fig. 15

XBB 755-3487

Fig. 16

XBB 755-3488

Fig. 17

Fig. 18

Fig. 19

XBL754-6179

Fig. 20

XBL754-6181

Fig. 21

XBL 754-6182

XBL 754-6178

Fig. 23

- M Matrix reflections
- T Twin reflections
- C Carbide reflections
- X Powder diffraction
- Y Grain walls

e

NIB 755-3482

M: Matrix reflections
T: Twin reflections
C: Carbide reflections
■: Superposition of Carbide and double diffraction spots

XBB 755-3494

Fig. 25

NBB 755-3481

Fig. 26

XBB 755-4493

Fig. 28