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Hiroyuki Tokushige
(Ph.D. thesis)

April 1983

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MICROSTRUCTURE-MECHANICAL PROPERTY RELATIONSHIPS
OF MARTENSITE AND LOWER BAINITE
IN A 0.3%C-3%Cr-2% Mn STEEL

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April 1983

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ABSTRACT

The microstructure, crystallography, mechanical properties and fractography of martensite and isothermally transformed lower bainite in a 0.3%C-3%Cr-2%Mn steel have been studied and compared in order to (i) design a strong, tough martensitic steel, (ii) characterize the lower bainitic structure and (iii) examine its applicability for heavy-gauge, high strength uses.

In the as-quenched condition, martensite of the present steel consists of heavily dislocated laths containing fine auto-tempered carbides and bounded by thin films of retained austenite. Similarly, lower bainite after 10 minutes of isothermal holding at 360°C consists of dislocated bainitic ferrite laths, unidirectional intralath carbides and interlath films of retained austenite. However, when the isothermal holding is prolonged to one hour, retained austenite has decomposed into interlath carbides in a similar manner to that observed in martensite tempered at 240-400°C. Crystallographic studies have revealed that bainitic ferrite laths are related to adjacent laths.
as well as to parent austenite through the same orientation relationships as those found for martensite laths, which strongly confirms the shear aspect in the bainite transformation. Bainitic carbides, identified as cementite, are found to have the Isaichev orientation relationship with ferrite, which supports the idea that cementite has formed directly from austenite. The lower bainitic structure is more similar to that of martensite tempered in the tempered martensite embrittlement (TME) range rather than as-quenched martensite.

These microstructural characteristics have been reflected in the mechanical properties. The as-quenched martensite exhibits a high strength and toughness, the latter being greatly enhanced by tempering at 200°C. The strength and toughness of lower bainite are substantially lower than those of as-quenched martensite, and they remain relatively unchanged upon tempering up to 400°C. Fractographic examinations indicate the similarities in the fracture mode between TME and lower bainite; in both cases, the absence of retained austenite films and the presence of coarse carbides appear to cause brittle fracture.

These results show that lower bainite is not favorable for high strength-toughness applications as compared to martensite which has achieved a high strength-toughness combination.
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1. INTRODUCTION

For many years both the martensite transformation and the bainite transformation have been widely used to obtain a variety of strength—toughness combinations in ferritic steels. A great number of martensitic or bainitic structural steels are now commercially available. However, due to the inherent complexities of these non-equilibrium phase transformations, a complete understanding has not been achieved despite tremendous efforts made from metallographic, kinetic, crystallographic and phenomenological theory viewpoints. In particular, steels containing relatively small amounts of carbon and other alloying elements, although frequently used for various engineering applications, have rarely been subjected to the fundamental studies of martensite or bainite transformation until recently. The high transformation temperatures and reaction rates of these steels have presumably rendered direct kinetic studies difficult. In addition, the fine "lath" structures of the transformation products in these low alloy steels seem to have undergone only limited microscopic and crystallographic studies.

However, as will be mentioned later, the advancement of transmission electron microscopy (TEM) in recent years has stimulated intensive research into the microstructure and crystallography of lath martensite in various low alloy steels [1,3-8,22-27]. Through a continuing alloy design program at Berkeley, this microstructural characterization of lath martensite has been effectively combined with investigations into obtaining a strength—toughness combination suitable for high
strength applications, and as a result, a series of medium
carbon-chromium base martensitic steels has been developed (3-8).
In the present research, an alloy design study of strong, tough
martensitic steels has been carried out on a 0.3%C-3%C-2%Mn
steel. Furthermore, using TEM techniques identical to those used
in the previous study of martensite (22-25), the microstructure
and crystallography of lower bainite in the same steel have been
characterized and correlated with the strength and toughness.

1.1 Design of Strong, Tough Martensitic Fe/Cr/C Alloys

Systematic studies of Fe/Cr/C base alloys (3-8) have
revealed that a combination of high strength and high toughness
can be obtained from a microstructure of heavily dislocated
martensite laths containing fine intralath carbides formed by
either tempering (6,7) or auto-tempering (8), with stable films
of retained austenite at the lath boundaries.

The presence of interlath retained austenite is often
associated with the enhancement of fracture toughness (1,3,6-10).
Moreover, the toughness of lath martensite drastically decreases
as the retained austenite decomposes and is replaced by coarse
stringers of carbides forming along the lath boundaries upon
tempering in a temperature range between 240 and 400°C (6-8, 11,
12). This decrease in toughness, known as tempered martensite
embrittlement (TME) (6-8, 11-16), indirectly indicates the signifi-
cance of retained austenite and presents a constant problem
with such martensitic high strength steels. The phenomenon of
TME has been disputed with respect to its mechanism (11-13,
15,17), the fracture path (11-19) and the effects of impurity
segregation (11, 12, 20, 21). This is in contrast with temper embrittlement (TE), which has been generally accepted to be caused by segregation of impurity elements to prior austenite grain boundaries (11, 12, 20, 89, 90).

The discovery of interlath austenite (3) also has drawn attention to the crystallography of lath martensite in low alloy steels (6, 22-27), inasmuch as it renders it possible to directly relate the crystallography of martensite to that of the parent austenite. These crystallographic studies combined with sophisticated techniques of electron microscopy (22-25) and atom probe analysis (25, 28, 29) have allowed detailed information to be obtained of the structure of lath martensite in Fe/Cr/C alloys and its formation.

Efforts to optimize the strength-toughness combination of the Fe/Cr/C base alloys by varying the chemical composition and heat treatment conditions have been carried out, and many measures for improved properties such as the decrease in C content (0.4 to 0.3%) (6, 7), a quarternary addition of Mn (up to 2%) (6-8), the decrease in Cr content (4 to 3%) with partial replacement by 0.5% Mo and the adoption of either double austenitizing (4,6) or high temperature austenitizing (5) have been implemented. After these developments in the alloy chemistry and heat treatments, a 0.3%C-3%Cr-2%Mn-0.5%Mo steel has exhibited a yield strength of approximately 200 ksi (1379 MPa) and a Charpy impact value at room temperature of nearly 30 ft-lb (40.7 J) in the as-quenched condition (8).

This study (8), however, has also reported that the addition
of 0.5% Mo apparently has little effect on the microstructure and the mechanical properties. Since Mo is an expensive element, it would be more cost-effective if Mo could be eliminated without any detrimental effects. Thus, as one of the objectives of the present research, the microstructure and the mechanical properties of martensite in a 0.3%C-3%Cr-2%Mn steel will be examined.

1.2 Demands of High Strength Bainitic Steel for Heavy Gauge Applications

Recently it has been reported that the uniform micro-duplex structure of the Fe/Cr/C alloys is beneficial to the wear resistance of the steel (30,93). Therefore, these alloys seem promising for such high strength structural applications as mining equipment and oil exploration facilities.

As part of the trend in recent technological development, such engineering structures have been constructed or designed to larger scales, greater sizes or more complex shapes because of the requirements for the exploration of deeper mines or wells and the demands for a more economical operation of the system. Consequently, structural components are becoming increasingly heavier in cross-section. This trend introduces a problem in the application of the martensitic Fe/Cr/C base alloys for these structures. With an increase in thickness, the quench cooling rate attained in the center of the material becomes smaller even if the surfaces are cooled rapidly. As a result, it would be difficult to obtain a fully martensitic structure in such heavy-gauge sections, and it would be necessary to assume the presence
of bainite mixed with martensite or even to design a fully bainitic steel as a substitute for martensite.

Ever since it was first reported by Davenport and Bain (31), the bainite transformation in steels has been recognized as one of the most complicated phase transformations, and there is no unique definition of bainite. Nonetheless, it has been generally accepted that microstructurally, bainite is a non-lamellar aggregate of ferrite and carbide (32). In this regard, the two distinct types of bainite, i.e., upper bainite and lower bainite, are distinguished by the morphology of the carbides present. Upper bainite has stringer-type carbide particles precipitated at bainitic ferrite lath boundaries; whereas lower bainite has carbide precipitation within the lath at an angle to its major direction (33). The propensity to internal precipitation, combined with the fact that lower bainite contains a very much higher dislocation density than upper bainite (34), makes this low-temperature product of the bainite reaction attractive because such a microstructure is very similar to that of dislocated lath martensite with fine carbide precipitation. To date, the retention of austenite at ferrite lath boundaries in lower bainite has been reported with several low alloy steels (35-37). Therefore, if in the present Cr/Mn/C steel interlath retained austenite is obtained together with a high density of dislocations and interlath carbides, then it is likely that the lower bainite could be as strong and tough as the martensite, and hence, would be appropriate for use in heavy-gauge structures which require high strength. This possibility is the basis for
the second objective of the present research, which is to examine the applicability of the lower bainitic microstructure in a 0.3%C-3%Cr-2%Mn steel for heavy-gauge, high strength structural components.

1.3 Characterization of the Lower Bainitic Structure

While the structure of lath martensite has been well characterized and correlated with the mechanical properties (7, 8, 11, 12), relatively few studies have been made of the characterization of the lower bainitic structure (4, 38). In order to design a lower bainitic steel, therefore, it is of primary importance to study the microstructure and crystallography of lower bainite.

Because of many features which are intermediate between the pearlite transformation and the martensite transformation, the bainite transformation has been a subject of debate in many regards, in particular in its mechanism (39). Two distinct theories have been proposed to explain the mechanism of bainite transformation. One considers the transformation to be an extension of the martensite transformation to higher temperatures; i.e., austenite transforms to supersaturated bainitic ferrite by a shear mechanism, which is followed by a precipitation of carbides within the ferrite (40). According to this theory, the bainite transformation is analogous to that of auto-tempered martensite (34). The rate of reaction, or the extent of shear is controlled by the diffusion of carbon in austenite away from the austenite/ferrite interface or by some other relaxation processes (40). The other theory considers that
the same mechanism that governs both the pearlite reaction and
Widmanstatten ferrite formation applies to the transformation at
lower temperatures, i.e. the bainite transformation is solely
diffusion-controlled (41). A local equilibrium is accomplished
at the austenite/ferrite interface (42), and the carbides
precipitate at the interface (39). One of the models constructed
from this theory is the ledge mechanism theory (43).

The growth rate of bainite has been directly measured by in-
situ studies using a hot-stage optical microscope (44, 45), and
subsequent attempts have been made to explain the results based
on either a shear transformation model (40) or a diffusion
controlled model (42). However, neither model has satisfactorily
explained the experimental result. A thermodynamic approach (42)
to the kinetics has shown that a diffusion-controlled model of
the reaction is in fair agreement with the growth rate determined
experimentally. However, it has been pointed out that the actual
growth rate is lower by several orders of magnitude, and a
displacive mechanism has been supported (40). Other researchers
have argued that the measured rate is of the same order or
slightly less than that allowed by volume diffusion of carbon in
austenite and that the diffusion-controlled model is not invalid
(41). An electron microprobe study (46) has reported no
supersaturation of carbon in bainitic ferrite, and therefore
concluded that a shear transformation is not involved. However,
recent field ion-atom probe analyses (47, 48) have revealed the
supersaturation of carbon in bainitic ferrite, although not to
such an extent as might be anticipated, and thus favored the
occurrence of a shear-type transformation. Nevertheless, the atom probe analysis (47) has shown a step-like profile of carbon concentration at the ferrite/austenite interface, which suggests that with respect to carbon, a local equilibrium is achieved between bainitic ferrite and untransformed austenite at the interface. The concept of local equilibrium is one of the bases of the diffusion-controlled model (42). As a consequence, the bainite transformation has begun to be considered to be a displacive transformation in which diffusion plays a significant role (94).

Studies of the crystallography of bainite (37-39, 49-52) have consistently declared the existence of a shear transformation in bainite reaction for either one or both of the following reasons; (i) the orientation relationship and habit planes between bainitic ferrite and parent austenite can be rationalized by the phenomenological theory of the martensite transformation (50, 51), (ii) the orientation relationship between ferrite and cementite in bainite is the same as that in tempered martensite (49, 50, 52). However, some studies have reported an orientation relationship which differs from that of tempered martensite and concludes that the cementite in fact forms at the austenite/ferrite interface rather than within the supersaturated ferrite matrix (35, 36, 51). This conclusion corroborates the suggestion that the bainite transformation may comprise a diffusion component as well as one of shear (35).

An advantage in studying the present steel is that the crystallography of lath martensite in a steel of a similar composition has been well studied (22-25, 95) and that the
formation of lath martensite has been well defined. Therefore, if comparable crystallographic analyses with respect to the three phases of ferrite, austenite and cementite are carried out for the lower bainitic structure of the present steel, then the bainite transformation would be understood in terms of similarities and differences with regard to the martensite transformation, and the contribution of shear, in particular, could be clarified.

In summary, the objective of this research is to study the microstructures and mechanical properties of martensite and lower bainite in a 0.3%C-3%Cr-2%Mn steel in order to:

(i) design a strong, tough martensitic steel without Mo,
(ii) characterize the lower bainitic microstructure, and
(iii) examine its applicability for heavy-gauge, high strength uses.
2. EXPERIMENTAL PROCEDURE

2.1 Material

The chemical composition of the steel studied is shown in Table 1. It is basically 0.3%C-3%Cr-2%Mn with small amounts of P, S, and other impurities.

The steel was supplied by the Japan Steel Works. It was induction-melted and cast into a round ingot in vacuum. The ingot was received after rough forging into a rectangular bar. It was upset- and cross-forged into a plate with a cross-section of 3 inches (76 mm) in width by 1.25 inches (32 mm) in thickness. The homogenization of the steel plate was not carried out because an overheating problem had been found to be associated with the homogenization as shown in Appendix 1.

2.2 Heat Treatment

In order to avoid possible complications in the microstructure produced by continuous cooling after austenitizing, the lower bainitic structure was produced by an isothermal transformation, whereas the martensitic structure was produced by conventional oil quenching. After preliminary experiments to determine the transformation characteristics of the steel, the results of which will be described later, the isothermal transformation temperature to produce lower bainite was fixed as 360°C.

Austenitizing was conducted in a vertical tube furnace under dynamic argon atmosphere; the holding time was 1 hour for all austenitizing temperatures. After austenitizing, the specimens
were dropped directly into either an oil bath for quenching or a salt bath for isothermal holding. Tempering at 600°C or above was carried out in a vertical tube furnace, whereas a salt bath was used for tempering below 600°C. The cooling after either isothermal treatment or tempering was done by means of water-quenching. Specimens for the mechanical tests were heat-treated after being machined into oversized test pieces.

2.3 Dilatometry

A Theta Dilatronic IIIR dilatometer was used to determine the $M_s$ and $M_f$ temperatures and the TTT diagram of the steel. The specimen type used for dilatometry is illustrated in Fig. 1a.

2.4 Metallography and Microscopy

2.4.1 Optical Metallography

Specimens for optical metallography were prepared by first grinding with emery paper down to 600 grit and then polishing with 1 µm diamond paste. A 5% nital etching solution was used to reveal the microstructure. The microstructure was examined with a Carl Zeiss microscope. Observations were made on the cross-section parallel to the main-flow direction of the finish-forging.

2.4.2 Scanning Electron Microscopy (SEM)

An AMR-1000 scanning electron microscope operated at 20 kV and an ISI DS-130 scanning electron microscope operated at 25 kV were used for fractographic studies and metallographic observations at high magnifications.
Surfaces of the broken Charpy specimens were subjected to fractographic studies. For the investigations of crack propagation, some of the broken Charpy specimens were plated with Ni and cut along the direction of crack propagation. The cross-sections were then ground, polished and etched in a 5% nital solution, and examined along the edge of the fracture surface. In order to investigate the correspondence between the microstructure and the fracture surface, some specimens were mounted in Koldmount resin and cut along the direction of crack propagation. After the cross-sections were prepared as mentioned above, the mounting material was removed, and both cross-sections and the fracture surfaces were observed simultaneously.

2.4.3 Transmission Electron Microscopy (TEM)

In order to make thin foils suitable for TEM, slices of about 15 mils (0.4 mm) thick were cut from bulk specimens (broken Charpy specimens or other blanks of a similar size) in such a way that the slices would be parallel to the main-flow direction of the finish-forging. The slices were chemically thinned to about 4 mils (0.1 mm) in a solution of 4% HF in H₂O₂. Discs of 3 mm in diameter were spark-cut from the thinned slices and electropolished in a twin jet electropolishing apparatus at room temperature using a solution of 400 ml CH₃COOH, 75 g CrO₃ and 21 ml H₂O. The electropolishing was conducted at 36-40 mA and 50-60 V.

The thin foils were examined in either a JEM 7A, Philips EM 301 or a Philips EM 400 microscope at an accelerating voltage of 100kV. In addition to the conventional bright field (BF) - dark
field (DF) techniques, microdiffraction analyses were carried out using the Philips EM 400.

2.5 X-ray Measurement

The amount of retained austenite was measured using X-ray diffractometry. The same specimens as for optical metallography were used, and the measurements were performed on the same surfaces as for optical micrographs, but after a different preparation. Surfaces were ground with emery paper down to 600 grit then dipped in a solution of 4% HF in H₂O₂ to remove the mechanically deformed layers.

A Picker diffractometer was operated at 40 kV and 14 mA with a Cu target and a crystal monochromater. The volume fraction of austenite was determined by the relative integrated intensities of (112) ferrite, (220) austenite and (113) austenite peaks according to the following equation (53).

$$V_\gamma = \frac{(R_\alpha/R_\gamma)I_\gamma}{I_\alpha + (R_\alpha/R_\gamma)I_\gamma}$$

where $V_\gamma$ is the volume fraction of austenite, $I_\alpha$ and $I_\gamma$ the integrated intensities of the ferrite and austenite reflections, respectively, and $R_\alpha$ and $R_\gamma$ the atomic scattering factors of the respective reflections. The details of this technique have been well explained elsewhere (11).

2.6 Mechanical Tests

Mechanical properties were evaluated by hardness measurements, tensile tests and V-notch Charpy impact tests. Hardness was measured on broken Charpy specimens in the Rockwell C scale.
Using round tensile specimens of 0.25 inches (6.25 mm) in diameter and 1 inch (25.4 mm) in the gauge length, the tensile test was carried out in a 300 kip capacity MTS testing machine at a cross head speed of 0.03 inches (0.76 mm)/min. The specimens were taken from the steel plate along the forging direction, and the specimen configuration is illustrated in Fig. 1b.

Charpy impact toughness was mainly evaluated by the impact value (the energy absorbed) at room temperature. When the ductile-brittle transition behavior was examined, the specimens were held in a bath of either chilled methyl alcohol or liquid nitrogen for testing below room temperature and in an oil bath for testing above room temperature. The direction of specimen is also incident with that of the finish-forging. The specimen configuration is shown in Fig. 1c.
3. RESULTS

3.1 Transformation Characteristics

Fig. 2 shows typical examples of dilatation curves at several isothermal transformation temperatures from which the TTT diagram was constructed. All the curves show a sinusoidal variation in expansion upon isothermal holding. The expansion reaches saturation within the order of 1000 seconds at all the temperatures, and no appreciable change in the specimen length is observed afterwards. However, the amount of expansion at saturation considerably decreases as the temperature is raised, indicating that the isothermal transformation is not completed.

The higher the transformation temperature becomes, the larger the portion is of austenite that remains untransformed, and at 520 °C no dilatation due to the transformation is observed at all. The incompleteness of the bainite transformation in Cr containing steels has been reported by several researchers (38, 47, 54) and the increase in hardenability due to the addition of Mn in the present steel seems to have enhanced the trend of incompleteness. The temperature at which the bainite reaction occurs to a negligible extent (i.e. 520 °C in this steel) has been referred to as the kinetic Bs (39).

Because of the incompleteness of the reaction, the TTT diagram in Fig. 3 is also partially incomplete, and only the bainite start curve and a curve showing the termination of the relatively rapid portion of the transformation are drawn as was done by Lyman and and Troiano (54). Nevertheless, it shows that the Ms and Mf temperatures of the steel are 330 °C and 206 °C, respectively.
Since the objective of the present study is primarily to understand the structure-property relationships, it is desirable to obtain a fully bainitic structure in order to compare with a fully martensitic structure. Therefore, it is necessary to use a temperature as low as possible but one above the $M_s$ temperature for the isothermal transformation. As a result, $360^\circ C$ has been chosen as the isothermal holding temperature.

Fig. 4 shows the progress of the reaction at $360^\circ C$ observed by optical microscopy. Bainite is recognized as a sheaf-shaped region etched darker than the matrix. (It will be shown to be lower bainite in the following section.) Because of its appearance in optical micrographs, this sheaf-shaped region is often referred to as a "plate" (33). However, it is totally different from a plate in the plate martensite. As will be shown later, the bainite "plate" is further divided into lath-like subunits unlike a martensite plate. In order to avoid the confusion, the term "plate" will not be used in this text. Yet it should be noted here that in lower bainite, a group of laths resembles a plate rather than a packet in lath martensite.

Bainite sheaves grow in number as well in both length and width and gradually fill up the matrix as the holding time increases. However, some islands of austenite remain untransformed even after 24 hours of isothermal holding. The volume fraction of bainite measured by a metallographic intercept method is plotted in Fig. 5 against holding time. It is clear that the reaction proceeds very rapidly between 20 min. and 40 min. of holding time, after a slow start, and reaches saturation almost 1 hour later. According to the results of TEM observations and X-ray...
measurements (in Section 3.2.3), it is unlikely that the untransformed regions remained austenitic after the water-quenching following the isothermal holding. They are believed to have undergone the martensite transformation during the quenching.

3.2 Microstructure and Crystallography

3.2.1 As quenched Martensite

The microstructure of martensite produced by oil-quenching from 900 °C was observed by optical microscopy, SEM and TEM, and typical micrographs obtained from each of three techniques are shown in Fig. 6.

It is found that the structure is heavily dislocated lath martensite, and that the laths are bounded by thin, continuous films of retained austenite (See Fig. 7). The presence of fine auto-tempered carbides within the laths is also observed. The SAD pattern in Fig. 7c shows the \( \langle 111 \rangle_{f1} // \langle 100 \rangle_{f2} // \langle 110 \rangle_{a}^* \) triplet, which is frequently observed in microstructures containing both lath martensite and retained austenite. It appears that both the Kurdjumov-Sachs (K-S) (55) and the Nishiyama-Wassermann (N-W) (56, 57) orientation relationships are obeyed upon the martensite transformation. It is noted that the two variants of the martensite laths have their \( [110]_f \) direction, which is perpendicular to the lath boundaries, in common, and that this direction is parallel to the \( [111]_a \)

* Suffices \( f \) and \( a \) in the text denote ferrite and austenite, respectively.
direction of the retained austenite. Thus, $(110)_f$ and $(111)_a$
planes are parallel to the broad faces of the lath boundaries.
All of these observations are in good agreement with the previous
studies (6, 22, 23, 25) and therefore the elimination of Mo appears
to have little effect upon the as-quenched microstructures.

3.2.2 Lower Bainite after 10 minutes of Isothermal Holding

As shown in Fig. 4, the bainite transformation has barely
started after 10 minutes of isothermal holding at $360^\circ$C. Fig. 8
shows the microstructure revealed by a 5% nital solution,
observed by both optical and scanning electron microscopy. The
matrix is mostly martensite which transformed from the remaining
austenite during quenching from the isothermal holding tempera-
ture. Bainite sheaves etched darker than the martensite, are
growing mainly from prior austenite grain boundaries. The high
resolution micrograph (Fig. 8b) shows the presence of carbides
inside the deeply etched regions, which indicates that these
regions are lower bainite.

Bainite regions present within the martensite matrix were
also observed in TEM through careful examinations of thin foils.
In Fig. 9a, the region marked by a dashed line has a contrast
slightly different from other areas in the grain and is divided
into lath-like subunits. Inside the subunits, coarse precipi-
tates, whose morphology is apparently different from the
Widmanstatten carbides which have been frequently observed in
lath martensite (2, 4-8, 11, 12, 15, 58, 61-63), are observed.
From these features, this region is considered to be a growing
lower bainite sheaf, while the rest of the grain is considered to
be martensite. The dark field image (Fig. 9b) shows that both the martensite and the lower bainite have continuous films of retained austenite at lath boundaries. The austenite films in the lower bainite seem thicker than those in the martensite.

Another bainite region observed is shown in Fig. 10, and its magnified view is shown in Fig. 11. In the bright field micrographs (Figs. 10a, 11a) it is clear that carbides are aligned in one direction inside the bainitic ferrite laths at an angle of 55-60° with the lath boundaries. These unidirectional carbides are characteristic of lower bainite (33-35,37-40). Again, continuous films of retained austenite are observed at the lath boundaries. The SAD pattern taken from this bainite plate shows two ferrite zones, [011]₉ and [113]₉; the former has the N-W orientation relationship, and the latter has the K-S relationship with the [112]ₐ austenite zone. The DF micrographs taken from the ferrite reflections 2, 3 (Figs. 10c, 10d) indicate that the bainitic ferrite laths in this region belong to either one of these two zones, and that the change in the orientation of laths frequently takes place. It should be noted that these two zones of ferrite laths have one of <011>₉ directions, which is perpendicular to the lath boundaries, in common, and that this direction is parallel to the [111]ₐ direction of the retained austenite. This is the same situation as that observed in the as-quenched martensite (Fig. 7).

A magnified view of this region (Fig. 11) shows a high density of dislocations within the bainitic ferrite laths. The DF micrograph of retained austenite shows that segments of the
austenite films lie across the laths. These segments are almost parallel to the unidirectional carbides.

The features of this microstructure are very similar to those of lath martensite except for the morphology of the intralath carbides, which are unidirectional and coarse as compared to thin platelets of cementite with the Widmanstätten morphology typical of lath martensite (6-8, 11, 58, 63).

Fig. 12 shows the microstructure near the tip of the same bainite region. In the DF micrograph taken from an austenite reflection (Fig. 12b), the austenite films are partially discontinous. At A a segment of the film across the lath appears to terminate inside the lath, and at B there are discrete sections of the austenite film. It is, therefore, apparent that the interlath austenite films are not stable at this temperature even at such short holding times as 10 minutes.

3.2.3 Lower Bainite after 1 Hour of Isothermal Holding

Optical, SEM and TEM micrographs of the lower bainitic structure produced by isothermal holding at 360°C for 1 hour are shown in Figs. 13 and 14. The unidirectionality of the carbides is clearly recognized with SEM and TEM. Bainitic ferrite laths are heavily dislocated as in martensite laths; however, the unidirectional bainitic carbides (Fig. 14) are much coarser than the auto-tempered carbides in the lath martensite (Fig. 7). Moreover, they are more coarse and stringy than those observed after only 10 minutes of isothermal holding (Fig. 11). Another interesting result is that unlike in the lath martensite or in lower bainite after 10 minutes of holding, no retained austenite
is found at the lath boundaries. Instead, there are some carbides present along the lath boundaries. It should be noted that these interlath carbides are often considered to be characteristic of upper bainite (33,34).

In thin foils, small regions of irregular shapes, which have larger thickness than the surrounding area were frequently found. Such regions as are shown in Fig. 15 are considered to be martensite which has formed from untransformed austenite present between bainite sheaves (hence corresponding to the unetched regions in optical micrographs). Although these martensite islands were originally austenite, they seem to have all transformed to martensite during quenching from the isothermal holding because an examination by electron diffraction did not reveal the presence of austenite within the region shown in Fig. 15.

Nevertheless, X-ray measurements indicated the presence of austenite as illustrated in Fig. 16. The volume fraction of austenite appears to increase with the holding time at 360°C until 1 hour, then starts to gradually decrease. After 1 hour of holding, the amount of austenite is even higher than observed in martensite produced by oil-quenching. However, this increase is considered due to the increase in the volume fraction of bainite. If lower bainite is more apt to retain austenite than is martensite of the same volume, the increase in the amount of bainite could lead to the increase in the total amount of austenite even though the amount of austenite in individual bainite regions decreases. Since the change in the volume fraction of bainite
with the holding time is shown in Fig. 5, it is possible to estimate the amount of austenite only in bainite regions if the amount of austenite retained in the martensite produced by water quenching is known. It is also shown in Fig. 16. The estimated volume fraction of austenite in bainite is illustrated in Fig. 17. Although these estimated values after short-time holding are questionable because of large errors, this figure qualitatively indicates that the volume fraction of austenite in bainite regions decreases with the holding time.

The actual decrease of austenite was confirmed by a TEM experiment using an intense electron beam in the EM 400. In Fig. 18 the SAD pattern involves reflections from austenite, and its DF image (Fig. 18c) shows interlath films of austenite which are no longer continuous. These discrete segments of austenite appear to be in the process of decomposition from continuous films as shown in the preceding section, and it is reasonable to assume that those interlath carbides observed in Fig. 14 are the product of the decomposition. It is interesting to point out that this is the same microstructural change as occurs upon tempering the lath martensite of Fe/Cr/C alloys in the temperature range from 240 to 400°C (6-8, 11-13, 25). However, the number of interlath carbides is relatively small, and the size and shape of them do not differ much from those of the intralath unidirectional carbides. Therefore, these carbides at the lath boundaries do not appear to be a representative microstructural feature in this condition.

Fig. 18 also presents crystallographic information. Two zone axes of the ferrite matrix are observed, i.e., [2\bar{1}1]_f and
[311]_f, both of which are related with [112]_a austenite zone by the K-S orientation relationship. Again, the lath boundaries are parallel to (011)_f and (111)_a planes, and [011]_f direction is common to the two ferrite zones. The analysis of the carbide pattern has revealed that these carbides are cementite. This is reasonable because this transformation temperature (360 °C) is too high for ε-carbide to form (6-8, 33, 39). These cementite particles appear here almost parallel to the lath boundaries but still show the unidirectional morphology. The orientation relationship between cementite and the [211]_f ferrite matrix is found to be the Isaichev orientation relationship [59]:

\[
\begin{align*}
(111)_f &// (010)_c^* \\
(011)_f &// (103)_c
\end{align*}
\]

This orientation relationship is illustrated in a stereographic projection in Fig. 19. Planes of low indices and closely oriented planes in the respective phases are plotted.

A microdiffraction technique which has recently been applied to the crystallographic study of lath martensite (25, 95) was carried out to examine relative orientations of adjacent bainitic ferrite laths. Fig. 20 shows the microdiffraction patterns obtained from six neighboring laths in the vicinity of the area in Fig. 18. It is clear that the zone axes of these laths are all very close to [311]_f, and that very little change in the orientation is observed.

Fig. 21 presents another microdiffraction result in which

*Suffix c denotes cementite.
neighboring laths are alternately oriented in either the \(<100>_f\)
or \(<111>_f\) zone having one of the \(<011>_f\) axes in a common direction. These two patterns are both observed in Lath 5 because of overlap of the electron beam into adjacent laths. Although the retention of austenite is not observed here, it would be the same situation as the \(<111>_f//<100>_f//<110>_a\) triplet which is found in the martensitic structure (Fig. 7), if the parent austenite crystal is assumed to have been in the \(<110>_a\) zone. This assumption is made based on a speculation that these laths have formed from the same parent austenite, but showing two variants of different orientation relationships, i.e., the K-S and N-W relationships, respectively. This speculation is reasonable because a similar situation has been already observed in Fig. 10.

The SAD pattern obtained from this area (Fig. 22) shows that cementite and the matrix in the \(<111>_f\) zone are related by the Isaichev orientation relationship. The analysis of another set of BF–DF–SAD also gives the same orientation relationship (Fig. 23). It is also noticed that in Fig. 23 \([011]_f\) axis is perpendicular to the lath boundaries, which is similar to other bainitic ferrite laths (Figs. 10, 18, 22). This \([011]_f\) type lath boundary is often found in the lower bainitic structure, and thus it is considered to be one of the characteristics of lower bainite as it is in lath martensite (6, 8). It is also pointed out that when the Isaichev orientation relationship is present between ferrite and cementite, the \([103]_c\) plane is always parallel to the \([011]_f\) plane which in turn is parallel to the lath boundaries.
In lower bainite, however, lath boundaries which are not a (011)_f type are occasionally found. Fig. 24 shows lath boundaries parallel to the (121)_f plane. Two ferrite patterns in Fig. 24b indicate the presence of (112)_f type twin relation. The DF micrographs, Figs. 24d and 24f, taken from ferrite reflections in the respective patterns give contrast to every other lath, whereas the DF micrograph, Fig. 24e, taken from the (121)_f reflection, common to both patterns, shows a continuous fringe across the laths. Therefore, these bainitic ferrite laths are twin-related. This is confirmed by a microdiffraction experiment (Fig. 25). Twin-related laths have been reported for lath martensite (6, 25), although in the martensite the twins are a (110)_f type as compared to a (112)_f type observed here. In Fig. 24, no retained austenite is observed at the twin-related lath boundaries, which is the same result as that obtained for the twin-related martensite laths. Fig. 26 shows another example of the twin-related laths. The twin plane is also one of (112)_f planes.

The orientation relationship between the ferrite matrix and cementite is again the Isaichev relationship in both Figs. 24 and 26. Although the (103)_c reflection is not present in Fig. 24, it is observed that the (101)_c plane is almost parallel to the (121) plane, and by referring to Fig. 19, the orientation relationship can be identified as the Isaichev relationship. Among three DF images taken from three cementite reflections, Figs. 24g and i involve double diffracted images due to twinning; however, Fig. 24h shows the presence of unidirectional cementite particles.
which are parallel to the lath boundaries.

Fig. 27 shows a relatively wide lath containing coarse cementite particles. The lath boundaries are almost parallel to the (103)_f plane, and this type of lath boundary is rarely observed. The SAD pattern (Fig. 27c) appears complicated due to quite a few satellite spots produced by double diffraction. Noting that the transmitted beam has two satellites on each side, and hence accounting only for the spot in the middle of every row of five diffraction spots, the analysis has resulted in the indexed pattern in Fig. 27d, which again shows the Isaichev orientation relationship.

It should be noted that the Bagayatskii orientation relationship (60) which has been verified for tempered martensite (61-63) and lower bainite (37, 49, 50) has not been observed throughout the present crystallographic study.

A trace analysis was carried out with the SAD patterns in order to determine the habit plane of cementite in the ferrite matrix. The result, illustrated in Fig. 28, shows that the habit plane is close to one of {112}_f planes. This is in good agreement with the previous reports on the cementite in lower bainite (35-37, 61, 64, 92), but, different from the {110}_f type habit plane of the Widmanstatten cementite in tempered martensite (2, 4-8, 58, 63, 92).

3.2.4 Martensite Tempered at 300°C (TME Structure)

The decomposition of austenite into interlath carbides has been found to occur in a similar manner in the tempering of martensite (6-8, 11-13) and in the isothermal transformation of
austenite to lower bainite. Because of this similarity it might be interesting to examine the microstructure of martensite tempered at 300°C. Fig. 29 presents optical, SEM and TEM micrographs of the microstructure observed in the present steel tempered at 300°C for 1 hour. The TEM micrograph (Fig. 29d) shows martensite laths without any interlath retained austenite films. Instead, the lath boundaries are decorated by stringy carbides. The intralath carbides, which have a typical Widmanstatten morphology with a [110]_f type habit plane, have become noticeably coarsened as compared to the as-quenched condition (Fig. 7). These coarse carbides in particular, the interlath carbides are clearly revealed by nital etching in the SEM micrographs (Figs. 29b and 29c). With respect to the presence of coarse carbides, the lower bainitic structure is more similar to the microstructure of tempered martensite than that of the as-quenched martensite. However, there is also a microstructural difference between lower bainite and tempered martensite. In tempered martensite, coarse carbides are mainly along the lath boundaries, whereas in lower bainite, not only the interlath carbides but also the intralath unidirectional carbides are coarse.

A detailed study of tempered martensite and its embrittlement has recently been carried out with this steel (11, 12), and therefore, this section will not be extended beyond a general description of the microstructure.
3.3 Mechanical Properties

Although the microstructure has to be fully martensitic or lower bainitic in order to determine the mechanical properties of each phase and to compare them with each other, it was hardly possible to obtain a fully lower bainitic structure as shown in 3.1. Figs. 4 and 5 show little increase in the volume fraction of bainite upon isothermal holding over an hour; therefore, for the examination of the mechanical properties of the lower bainitic structure, the specimens were isothermally treated for 1 hour at 360°C. Those materials which will be hereafter referred to as lower bainite were all treated in this condition.

The results of mechanical tests in the untempered conditions after single austenitizing at either 1100°C or 900°C are summarized in Table 2. For comparison, the results for the martensite quenched from 900°C and tempered at 300°C (in the TME condition) is also shown in Table 2. Also, in order to compare the mechanical properties of the as-quenched martensite in the present steel with those of the Mo containing steel, the results for the Mo containing steel in the as-quenched conditions (8) are shown in the same table. The effects of double austenitizing on hardness and Charpy impact toughness are shown in Table 3. In addition, Fig. 30 shows the Charpy impact transition curves of untempered martensite and lower bainite, both austenitized at 900°C. The changes in hardness and impact toughness upon tempering (after austenitizing at 900°C) are summarized in Table 4 and illustrated in Figs. 31 and 32. These figures also include the previous data for the Mo containing steel.
3.3.1 As-quenched Martensite

As shown in Table 2, the tensile properties of martensite in the as-quenched conditions are almost unaltered from previous results obtained for the Mo containing steel (8). After single austenitizing, the yield strength is maintained at ~200 ksi (1379 MPa) in spite of the elimination of Mo. The Charpy impact toughness at room temperature after quenching from 900°C varies little between the present steel and the Mo containing steel. However, the impact toughness after austenitizing at 1100°C is substantially lower in the present steel than in the Mo containing steel. As shown in Fig. 33, high temperature austenitizing is associated with the coarsening of prior austenite grains as well as martensite packets. It is, therefore, reasonable to attribute the low toughness of the present steel after 1100°C austenitizing to grain coarsening. In the case of the Mo containing steel (8), austenitizing at 1100°C was preferred to dissolve coarse carbides and other precipitates initially present in the material, and this effect appeared to exceed the deleterious effect of grain coarsening. Although the presence of coarse undissolved carbides has not been metallographically observed in the present steel, double austenitizing, which was designed to dissolve coarse precipitates at 1200°C and then to refine prior austenite grains at 900°C, has provided the steel with further improvement in toughness as shown in Table 3. Hence, the present steel may contain a certain amount of undissolved precipitates before the final heat treatment, so that the toughness after single austenitizing at 900°C is not as high as that previously obtained for the Mo
containing steel quenched from 1100°C. However, unlike in the Mo containing steel, 900°C austenitizing still gives higher toughness than 1100°C austenitizing in the present steel. As shown in fig. 30, the ductile brittle transition temperature (DBTT) is well below room temperature. Austenitizing at 900°C also gives higher tensile ductility as compared to austenitizing at 1100°C. For these reasons, in the case of the present steel, austenitizing at 900°C is preferred.

3.3.2 Lower Bainite

Both the strength and hardness of lower bainite in the as-transformed condition are substantially lower than those of the as-quenched martensite. Despite the general rule of the inverse relationship between strength and toughness [1], the impact toughness of lower bainite is considerably lower than that of martensite. The impact value at room temperature changes little with the austenitizing temperature. Optical micrographs (Figs. 33 c and d) show that bainite sheaves grow into such various directions that prior austenite grains are further divided more finely as compared to prior austenite grains in martensite which are divided by packets, and hence the difference in the prior austenite grain size is not as distinct in lower bainite as in martensite. The Charpy transition curve in Fig. 30 indicates that the DBTT is above room temperature for lower bainite. Also, unlike in the case of martensite, the toughness was further dropped by double austenitizing showing that the dissolution of
coarse precipitates does not seem to be beneficial to the toughness of lower bainite.

In overall comparison, lower bainite is not as strong or as tough as the as-quenched martensite. Similarly, when the tempered martensite embrittlement takes place, both strength and toughness become lower than the as-quenched martensite. Although the impact value in the TME condition is not quite lower than that of the as-quenched martensite (Table 2), the fracture appearance of the former is mainly quasi-cleavage, whereas that of the latter is mainly in a ductile mode as will be shown in Fig. 34. Therefore, the former is more brittle than the latter, even though the impact values are at the same level, and this is because the upper shelf energy value in the TME condition is higher than that in the as-quenched condition owing to the decrease in strength. The low strength and low toughness of lower bainite are more similar to those of the TME structure than to those of the as-quenched martensite, although both the strength and toughness of the former are still lower than those of the latter (Table 2). This implies that even though these two microstructures are somewhat similar, there should be some microstructural differences which further lower the toughness of lower bainite.

3.3.3 Effect of Tempering

It has been shown that the microstructure and mechanical properties of lower bainite are more similar to those of the TME structure than to those of the as-quenched martensite. If it is true that the microstructure and mechanical properties of lower
bainite are virtually similar to those of tempered martensite, the mechanical behaviors of lower bainite upon tempering would be different from those of martensite.

This is clear in Fig. 31. The hardness of martensite monotonically decreases over a temperature range up to 650°C. It is almost equal to the hardness of the Mo containing steel up to 400°C then starts falling more rapidly. This is probably due to enhanced temper resistance by the Mo addition through its strong tendency for secondary hardening. The hardness of lower bainite, in contrast to martensite, remains almost constant up to 400°C except for the slight decrease upon 200°C tempering and falls in the same level (or even higher) as the hardness of martensite at or above 400°C. This suggests that little change in the microstructure of lower bainite occurs in a temperature range up to 400°C.

The impact toughness of martensite (Fig. 32) displays a typical tempering curve for this kind of martensite steel (6-8), although changes are more rapid as compared to those in the Mo containing steel (8). A drastic increase is observed upon 200°C tempering, which has been attributed to the depletion of carbon from the supersaturated ferrite matrix by the precipitation of fine carbides (6,82) and by the diffusion into retained austenite (6,25). The impact value in this condition is much higher than that of the Mo steel. However, a considerable decrease in toughness takes place at 300°C. At this temperature, it has already been observed that the decomposition of retained austenite and the formation of interlath carbides take place (Fig. 29), and
therefore, this toughness drop is considered to be TME. The drop
is followed by the second trough at 500°C, which will be proved
to be temper embrittlement (TE) by fractography (Section 3.4).
Although toughness begins to recover above this temperature, it
remains at a lower level than the Mo steel, which indicates the
effectiveness of Mo in suppressing TE.

The impact value of lower bainite is relatively unaffected
by tempering up to 500°C though it shows a slight increase at 200
°C and a slight decrease at 300°C, i.e., the same trend as
martensite. Then without showing a further decrease, it rises to
an extremely high value upon 650°C tempering. This result also
supports an idea that little change takes place in the lower
bainitic structure upon tempering up to 400°C.

3.4 Fractography

3.4.1 Comparison of Brittle Fracture

Both microstructural observations and mechanical tests have
demonstrated the similarities between lower bainite and tempered
martensite. Therefore, in the first part of this section, the
fractographs of broken Charpy specimens especially in brittle
conditions will be compared for the as-quenched martensite,
tempered martensite (TME) and lower bainite.

Fig. 34 shows the fractographs of Charpy specimens broken at
room temperature. In the as-quenched martensite (Fig. 34a) the
fracture mode is mainly ductile mixed with quasi-cleavage, which
is a typical fracture appearance above DBTT. On the other hand,
both TME (Fig. 34b) and lower bainite (Fig. 34c) specimens
exhibit a quasi-cleavage mode. From Fig. 34b it is clear that in
this condition (tested at room temperature) the TME specimen is
fractured in a brittle manner even if the impact value is not
quite low. The TME specimen shows elongated ridge shaped cleavage
facets, which have been often reported for martensitic steels
which are tempered in the TME range (6,8,11-14).

In order to examine the effects of microstructure upon
fracture in particular brittle fracture, the comparison of
fracture appearance should be made at the same level of impact
toughness. Fig. 35 shows fracture surfaces of the three micro-
structures tested near the lower shelf (i.e., -196 °C for the as-
quenched martensite and TME, -75 °C for lower bainite). The
impact values range from 3.4 to 6.0 ft-lb (4.6 to 8.1 J). All
the microstructures show a quasi-cleavage mode, and little
difference is observed in the fracture appearances. However,
fractographic examinations combined with a metallographic
technique have revealed differences in crack propagation.

Fig. 36 shows the correspondence between the martensitic
microstructure in the as-quenched condition and the fracture
surface which is plated with Ni. As indicated by the arrows, lath
boundaries apparently show signs of resistance to fracture. The
resistance to crack propagation provided by lath boundaries is
also demonstrated at the secondary crack shown in the same figure
(upper left-hand corner); the crack changes its direction at
every lath boundary. These observations suggest the likely
beneficial effects of retained austenite films at lath boundaries
upon the toughness.
A similar advantageous effect of lath boundaries is not found with the TME structure which does not have interlath austenite films. Fig. 37 shows the microstructure and corresponding fracture surface in a Ni plated TME specimen. In this case secondary cracks in Fig. 37a propagate across or partially along lath boundaries. In addition, the main crack passes across laths without being noticeably arrested by lath boundaries and partially goes along a lath boundary (shown by the arrows in Fig. 37b). It should be noted that the majority of crack paths does not appear to be associated with lath boundaries containing coarse carbides as previously reported. (6,8,12,14).

This is confirmed by observing the fracture surface and the microstructure simultaneously. In Fig. 38 cleavage facets are perpendicular to lath boundaries showing that in a brittle mode the fracture paths are not interlath. Therefore, it is concluded that the occurrence of cracks appearing to run along lath boundaries is rather a fortuitous phenomenon than a primary event, although as shown in Fig. 37, those boundaries which contain coarse carbides can provide cracks with easy paths.

Figs. 39 through 41 are the results of the same experiments performed on lower bainite. A secondary crack in lower bainite also proceeds over a distance much larger than lath width with no signs of resistance from the lath boundaries (Fig. 39). Two-surface fractographs in Fig. 40 show that one unit of cleavage facet sometimes covers a whole bainitic region (Fig. 40a) and that in other cases more than one cleavage facet is present in one bainitic region (Fig. 40b). In any case, however, cleavage fracture appears to be confined within bainitic regions, and
martensite islands as shown in Fig. 40a do not seem to yield to cleavage. In Fig. 41 the main crack which is covered with plated Ni propagates across the unidirectional carbides - not along the direction of the long axis of the carbides. Therefore, these carbides do not seem to contribute to providing easy crack paths.

3.4.2 The Change in Fracture Mode on Tempering

The fractographs of tempered Charpy specimens tested at room temperature are shown in Fig. 42, and these fractographs explain the change in the toughness of tempered martensite and tempered bainite.

For martensite, a transition in the fracture mode from quasi-cleavage (Fig. 42a) to an intergranular mode (Fig. 42c) is clearly observed as the tempering temperature is raised from 300 °C to 500 °C. The second trough in impact toughness (Fig. 32) coincides with this transition, and thus, can be attributed to temper embrittlement. Intergranular fracture remains a major fracture mode upon tempering at 650 °C although its contribution has declined (Fig. 42e).

The change in the fracture mode of lower bainite with tempering temperature is essentially the same as that of martensite, although the susceptibility to temper embrittlement appears to be smaller than in martensite. The fracture mode on 300 °C tempering is quasi-cleavage (Fig. 42b), which does not differ from that in the untempered condition. This is reasonable because the results of mechanical tests (Figs. 31 and 32) have suggested little microstructural change in lower bainite upon
tempering up to 400 °C. Upon tempering at 500 °C, the predominant fracture mode becomes intergranular, although, there are still many regions in which quasi-cleavage has occurred (Fig. 42d). The change in the toughness of lower bainite with tempering does not show the second trough at 500 °C (Fig. 32) probably because of this smaller fraction of intergranular fracture. At 650 °C, when the toughness level has significantly increased, about 80% of the fracture is ductile (Fig. 42f), the remaining region still being intergranular (Fig. 42g). Therefore, even lower bainite still exhibits the influence of temper embrittlement.
4. DISCUSSION

4.1 Characterization of the Lower Bainitic Microstructure

4.1.1 Crystallography of Bainitic Ferrite and Austenite

As mentioned earlier, the crystallography of lath martensite in the Fe/Cr/C base alloys (22, 23), particularly a 0.3%C-3%Cr-2%Mn-0.5%Mo steel, has been studied in detail and rationalized in terms of a mechanism of shear transformation (22, 23, 25). It has been hypothesized (6,22,23,25,95) that a martensite lath forms by shear in a particular $\{11\}_a$ plane maintaining its particular close-packed plane, i.e., $[011]_f$, parallel to this $\{11\}_a$. Strains built up in the surrounding austenite by the formation of the first lath, promote subsequent lath formation while stabilizing the austenite trapped between the laths. During the successive formation, adjacent laths arrange their relative orientations so that the overall transformation strain will be minimized.

As a consequence of the shear transformation, lath martensite has a $\{11\}_a$ type habit plane which is parallel to a $[011]_f$ at the interface between martensite and austenite, i.e., the broad face of the lath boundary (6,25,95). Therefore, adjacent laths in a packet have one $<011>_f$ direction perpendicular to the lath boundaries in common. The relative orientations of adjacent laths which have resulted from the strain minimization are observed in various modes such as a rotation about the common $<011>_f$ axis by either small angles ($0^\circ < \theta < 10^\circ$) or large angles ($\theta > 20^\circ$) (24, 25, 95), or $[011]_f$ type twin-related laths (6,25). The orientation relationship between austenite and martensite varies from the Kurdjumov–Sachs (K–S) relationship.
(55) through the Greninger-Troiano (G-T) relationship (64) to the Nishiya-Wassermann relationship (56, 57). As shown earlier, Fig. 7 presents a typical example of lath martensite.

As far as the austenite and ferrite phases are concerned, the crystallographic characteristics observed in the lower bainitic structure of the present steel are mostly in good agreement with the above results obtained for the lath martensite. The presence of a $<011>_f$ direction perpendicular to lath boundaries has been frequently observed for lower bainite (Figs. 10, 18, 21, 23). This direction is common to adjacent laths even if the relative orientation is changed (Figs. 10, 18, 21). With the presence of retained austenite, this $<011>_f$ direction has been shown to be parallel to a $<111>_a$ direction (Figs. 10, 18) which would imply that a $[111]_a$ type habit plane is present in bainite transformation.

Three types of arrangement of adjacent laths have been revealed by microdiffraction experiments, viz., (i) clustering orientations around the $[311]_f$ pole (Fig. 20), (ii) abrupt change in the zone axis from $<111>_f$ to $<001>_f$ (Fig. 21) and (iii) twin-related laths (Fig. 25). Such a small difference in the orientations of adjacent laths as case (i) has been observed for martensite laths (25) with the difference that in martensite it is associated with a large angle rotation around a $<011>_f$ axis in the vicinity. The presence of a large angle rotation is not shown in Fig. 20. However, as stated, the observed area is very close to the area in Fig. 18; hence, as shown in the SAD pattern (Fig. 18d), there is a rotation from the $[311]_f$ zone axis to the
[211]_f zone axis in the vicinity of the area in Fig. 20. The abrupt change in the orientation of neighboring laths in case (ii) appears to be due to the coincident operation of both the K-S and the N-W variants, hence the same situation as Fig. 7, and it can be interpreted as a large angle rotation around the common [011]_f axis resulting from the variation in the G-T orientation relationship (24,25,95). Therefore, these two cases are the same situation as the lath rotation around a <011>_f axis by either small angles or large angles in lath martensite. The relative orientations of the bainitic ferrite laths in Fig. 10 are also an example of the rotation around a <011>_f axis.

The case of twin-related laths (Figs. 25, 26) in lower bainite is, however, not the same as that found in martensite in that the former twin plane is {112}_f whereas in the latter it is {011}_f (25). In the case of the {011}_f type twin plane, it is considered to be the simplest arrangement of adjacent laths for minimizing the transformation strain because the net shear over two laths is zero (25). As a result, no retained austenite is observed at twin-related lath boundaries (25). Also, a {011}_f type twin does not contradict the observed {111}_a habit plane. Although why a {112}_f type lath boundary is formed in lower bainite may have to be subjected to further study, these laths are still considered to be the result of strain minimization by adjacent laths because no interlath carbides are observed in Figs. 24 and 26, which suggests that no retained austenite was previously present at the lath boundaries as in the case of twin-related laths in lath martensite. The reason for the occurrence of a {103}_f type lath boundary (Fig. 27) is not clear yet.
However, both \{112\}_f and \{103\}_f type boundaries are rarely observed, and the majority of the lath boundaries in lower bainite is of the \{011\}_f type.

In summary, although a few differences are observed, crystallographic characteristics such as the orientation of lath boundaries, the orientation relationship between ferrite and austenite and the mutual arrangement of adjacent laths in the lower bainitic structure are very similar to those in lath martensite. Therefore, the existence of shear in the bainite transformation is strongly indicated.

4.1.2 Crystallography of Bainitic Cementite

Although the results in the previous section have shown clearly the similarity between the crystallography of bainitic ferrite and retained austenite and that of lath martensite and retained austenite, this is not completely the case when considering the crystallography of cementite in bainite and cementite in tempered martensite.

The orientation relationship between cementite and bainitic ferrite is the Isaichev orientation relationship (59), viz.,

\[
\{111\}_f \parallel \{010\}_c \\
\{011\}_f \parallel \{103\}_c
\]

instead of the Bagaryatskii orientation relationship (60) between cementite and lath martensite, viz.,

\[
\{110\}_f \parallel \{100\}_c \\
\{111\}_f \parallel \{010\}_c \\
\{11\bar{2}\}_f \parallel \{001\}_c
\]
However, these orientation relationships are not totally different from each other. As can be observed in Fig. 19, the Bagaryatskii orientation relationship is obtained by rotating the cementite phase around the [010]_c axis by approximately 4°. The principal axes of the cementite phase are oriented in such close directions that these two orientation relationships have been regarded as one relationship which is typical of tempered martensite in some of the previous studies (49, 64). However, as shown later, the small difference between these orientation relationships should not be ignored.

It should be noted that when the Isaichev orientation relationship is obeyed, the [103]_c plane is parallel to the (011)_f plane which in turn is parallel to the lath boundaries (Figs. 18, 22, 23) except for the cases when the lath boundaries are parallel to [112]_f or [103]_f, (Figs. 26 and 27 respectively). As discussed in the previous section, (011)_f is very likely to be the habit plane of bainite transformation, and hence is parallel to one of [111]_a planes of the parent austenite (e.g., (111)_a). Then, there is a parallelism between these phases, viz.,

\[(\bar{1}1\bar{1})_a // (01\bar{1})_f // (103)_c\]

This is indeed observed in Fig. 18. In this case, the K-S (55) orientation has been found between ferrite and retained austenite, viz.,

\[(1\bar{1}1)_a // (0\bar{1}1)_f\]
\[(110)_a // (111)_f\]
\[(\bar{1}12)_a // (\bar{2}11)_f\]

Accounting for this, another parallelism between three phases is
derived, viz.,

\[ (110)_a // (111)_f // (010)_c \]

The first parallelism indicates that the close-packed planes of austenite, ferrite and cementite are parallel to each other, whereas, the second parallelism indicates that the close packed directions of the three phases (i.e. the plane normals of the respective plane) are parallel to each other \([59, 64]\).

Assuming these parallelisms, the orientation correspondence between austenite, ferrite and cementite is illustrated in the stereographic projection shown in Fig. 43. It is interesting to note that a new orientation relationship is now established between austenite and cementite, which is close to:

\[ (554)_a // (100)_c \]
\[ (110)_a // (010)_c \]
\[ (225)_a // (001)_c \]

This orientation relationship is the Pitsch orientation relationship \([66]\), which was first found between austenite and proeutectoid cementite in a hypereutectoid C-Mn steel. Therefore, it can be concluded that bainitic cementite was initially formed from austenite. The same conclusion has been derived by Huang \([35, 36]\) for 2%Si steels and a 12%Ni–4%Co steel.

When the N-W orientation relationship is present (Figs. 10 and 21), however, the SAD patterns obtained from these areas are not clear enough to allow the indexing of cementite reflections. Therefore, it is not certain whether the Pitsch orientation relationship is observed between cementite and austenite in the case of the N-W orientation relationship between ferrite and
austenite. Nonetheless, it is still possible to predict an
orientation relationship between ferrite and cementite, assuming
that cementite precipitates from austenite through the Pitsch
orientation relationship and that the N—W relationship, viz.,
\[(111)_a \parallel (011)_f\]
\[(101)_a \parallel (100)_f\]
\[(121)_a \parallel (011)_f\]
is obeyed between austenite and ferrite. In this case the
parallelism between the close packed directions of the three
phases no longer exists. However, the first parallelism between
the close packed planes may be still present because \[(111)_a \parallel (011)_f\] is still true. Therefore,
\[(111)_a \parallel (011)_f \parallel (103)_c\]
is again assumed. In order for the above N—W relationship to be
met as this parallelism is maintained, the ferrite phase in Fig.
43 has to be rotated with the \([011]_f\) direction by 5° while the
austenite and cementite phases are fixed. The predicted orienta-
tion relationship is shown in Fig. 44, in which no parallel
relations between particular low index planes in the two phases
are observed except that \([011]_f \parallel (103)_c\]. Therefore, the SAD
patterns of ferrite with low-index zone axes would not be
associated with the SAD patterns of cementite with low-index zone
axes. This may have been the reason that clear SAD patterns
could not be obtained from the cementite phase when an N—W
variant was present.

Huang’s analysis (36) has been criticized that the result
was deduced using separately determined austenite/ferrite and
ferrite/carbide orientation relationships (37). The present
result, however, is derived based on a direct analysis of the orientation relationships between coexisting three phases, i.e., ferrite, cementite, and retained austenite (Fig. 18) and therefore supports Huang's previous results.

The conclusion that, in lower bainite, cementite has precipitated from austenite, obeying the Isaichev orientation relationship, is compatible with the fact that there are differences in the habit plane and the morphology of cementite between lower bainite and tempered martensite, the latter showing the Bagaryatskii orientation relationship. Previous studies (35, 37, 64) as well as the present one have reported that the habit plane of cementite in lower bainite is near \( \{112\}_f \), and as a well-known fact, the morphology of cementite in lower bainite is unidirectional (33-38, 49-52, 58, 64), i.e., the precipitation occurs in only one of the planes which are possible for the habit plane. On the other hand, in tempered martensite, cementite has \( \{011\}_f \) type habit plane and exhibit a Widmanstatten morphology (2,4-8, 11, 12, 15, 58, 61-63). An example of Widmanstatten cementite is shown in Fig. 29d, and it is obvious that the precipitation has occurred in two or three habit planes. If the formation of bainitic cementite is the same phenomenon as the formation of Widmanstatten cementite in tempered martensite, the resulting habit plane and morphology should also be the same. Therefore, these differences also support the idea that the orientation relationship between bainitic ferrite and cementite and hence the parent phase of cementite precipitation does differ from that between tempered martensite and Widmanstatten
cementite.

Some of the previous crystallographic studies (34, 37, 49, 64) have argued that cementite in lower bainite forms from supersaturated ferrite after the bainite transformation takes place by a shear mode, based on the observation of the Bagaryatskii orientation relationship. The Bagaryatskii relationship has been found in the lower bainitic structure of 1%Cr-0.5%Mo-0.002%B steels (with 0.1-1%C) (34, 49), a 0.3%C-4%Cr steel (37) and a 0.66%C-3.3%Cr steel (64). However, the accuracy of the analyses is questionable. For example, Shackleton and Kelly (49) examined the orientation relationship after indexing both the cementite and the ferrite patterns using the stereograms previously prepared upon an assumption that the orientation relationship would be either the tempering relationship (i.e., the Bagaryatskii) or the pearlite relationship (67). They did not consider the Isaichev relationship; it is likely, therefore, that they may have mistaken the Isaichev relationship for the Bagaryatzkii relationship. Bhadeshia (37) has concluded that bainitic ferrite is related to the parent austenite through an N-W variant and that cementite has the relationship of Bagaryatskii with the ferrite matrix. However, as already shown, individual bainitic ferrite laths can be related to austenite through either the K-S or the N-W variants. Also, in one of the SAD patterns shown in his paper, an angle between a cementite reflection and a ferrite reflection is closer to that in the Isaichev relationship than that in the Bagaryatskii. Srinivasan and Wayman (64) have shown only a limited number of data with respect to a 0.66%C-3.3%Cr steel. Their only SAD pattern,
however, indicates the presence of twinning hence is likely to involve double diffraction spots in the cementite pattern.

Shackleton and Kelly (49) have reported that the upper bainite in their steels exhibited another orientation relationship in addition to the Bagaryatskii relationship, and thereby concluded that cementite begins to precipitate directly from austenite as the transformation temperature is raised. Although the number of available data is limited, it appears to be generally accepted that in upper bainite the carbide forms from austenite trapped between the bainitic ferrite laths (33). The present results have shown that the origin of cementite precipitation changes from ferrite to austenite upon the transition from martensite to lower bainite and not upon the transition from lower bainite to upper bainite.

4.1.3 Mechanism of Bainite Transformation

With respect to the mechanism of the bainite transformation, two apparently contradictory conclusions have been deduced from the preceding discussions on the crystallography of lower bainite. The crystallography of bainitic ferrite and austenite suggests that the ferrite matrix forms by a shear transformation; whereas the crystallography of bainitic cementite indicates that the carbides precipitate directly from austenite and hence suggests a diffusion-controlled transformation. Therefore, the bainite transformation must combine the nature of both shear, and diffusion-controlled transformations (94).

From the present experimental results, it seems obvious that
a shear mechanism is involved in the bainite transformation. This can be shown not only by the crystallographic similarities between lower bainite and lath martensite, but also by the substructure of a bainitic ferrite lath, which is found to consist of a high density of dislocations (Figs. 11, 18, 22-24, 26, 27) similar to that in lath martensite. Moreover, in Fig. 10, it is observed that the orientation of ferrite laths changes alternatingly between [011]_f and [113]_f zones. This orientation change is due to the change from a K—S variant to an N—W variant. It should be noted that the lath orientation sometimes changes along the growth direction of the bainite sheaf. If the growth of bainite is solely due to diffusional mechanism, the orientation would hardly change along the growth direction.

In order to discuss the role of diffusion in the bainite transformation, the precipitation of cementite should be considered. Although it has been shown that the cementite particles precipitate from austenite, they may not precipitate exclusively within austenite. If this were the case, it would be a similar situation to that found in the pearlite reaction; therefore, the cementite would be likely to have the pearlite orientation relationship with the ferrite matrix rather than the observed Isaichev. Also, the observations made with SEM and TEM, which show no signs of carbide precipitation in the martensite islands (e.g. Figs. 15, 40, 41), means that there is no carbide precipitation within the untransformed austenite.

Therefore, the bainitic cementite must precipitate at the ferrite/austenite interface. The unidirectional morphology of
the carbides in lower bainite can be explained for this reason. Aaronson et al. [68] have shown that if precipitation takes place at interphase boundaries, the precipitate must be in contact with both phases. Following this theory, the cementite particles are considered to form in contact with both the austenite and ferrite phases. This is reasonable because in this manner, the three orientation relationships between either two of the ferrite, austenite and cementite phases can be easily maintained. As far as iron atoms are concerned, the lattice structure of cementite does not differ very much from that of ferrite [69], therefore, it is likely that the ferrite surface in contact with austenite may act as a potent nucleation site for cementite [51].

Thus, during the bainitic reaction, the carbides form at the austenite/ferrite interfaces as the ferrite phase grows (i.e., the interface moves) by a shear motion. It would be reasonable, then, to assume that the extent of shear is controlled by the precipitation of carbides at the moving interface. In this sense the role of the carbide precipitation in the bainite transformation is similar to that in the case of interphase precipitation [70], except that the progress of the interface is controlled by shear in the former instead of the formation of ledges in the latter [70]. Therefore, bainite transformation is a diffusion-controlled shear transformation.

Fig. 11 provides this hypothesis with strong support. During the transformation, it may occur that the local carbon concentration ahead of the austenite/ferrite interface is lowered to such an extent that the shear motion can resume even if the
carbides have not formed at the interface. The segments of austenite films lying across the laths in Fig. 11 are considered to have been retained after the interface moved away. The fact that the unidirectional carbides are almost parallel to these segments of austenite suggests that the carbides have been formed at the austenite/ferrite interface when the shear was momentarily halted.

The explanation of the morphological change of bainitic carbides between lower bainite and upper bainite, i.e., intra-lath, unidirectional carbides vs. interlath carbides (33), can be given in terms of the driving force for a shear transformation and the diffusivity of carbon. When the transformation temperature is low, the driving force is so high that shear can start wherever the carbon content is locally lowered. The prior austenite grain boundaries are likely to exhibit local depletions of carbon and hence may be preferential sites for the nucleation of bainite. However, the diffusion of carbon in austenite away from the moving austenite/ferrite interface is so slow that carbon cannot maintain a complete partitioning between the austenite and ferrite. This results in the formation of unidirectional carbides at the interface; the shear transformation can continue after the local carbon concentration is lowered by the precipitation. On the other hand, at a high temperature, although the carbon diffusivity is raised, the driving force for shear is lowered, and therefore, carbon can diffuse away from the austenite/ferrite interface before the bainitic ferrite laths form. As a result, the carbon concentration in the austenite trapped between the laths
increases, eventually leading to the formation of carbides at the lath boundaries.

In reality, these two types of carbides are present concurrently in the lower bainitic structure at the transformation temperature of 360°C; the unidirectional intralath carbides and the interlath carbides formed after the decomposition of retained austenite are both observed, although the latter is much less in number. The coexistence of the two types of carbides has been reported as an intermediate microstructural feature between lower bainite and upper bainite (34, 40). Since the transformation temperature in the present study is close to 350°C, the temperature at which, according to Hehemann (33, 39, 40), the transition from lower bainite to upper bainite occurs regardless of the chemical composition of steel, it may not be unusual to observe the two types of carbides. It should be noted, however, that in the present case, the decomposition of retained austenite is a secondary sequence of the bainite transformation, viz., the interlath carbides form not upon the formation of lower bainite, but, as a result of the subsequent isothermal holding. In this sense, the lower bainite transformation can be defined to involve the "delayed upper bainite reaction." It is interesting to point out that the lath martensitic structure of the same steel has been termed as "untransformed upper bainite" (87,95) because of the presence of retained austenite films which are stable even after the martensite transformation is completed.
4.1.4 Comparison of the Lower Bainitic Microstructure with the Lath Martensite

To conclude the characterization of the lower bainite in the present steel, its microstructure will be compared with that of the lath martensite. The similarities and differences will be pointed out in this section, and they will be correlated with the mechanical properties that are discussed in a later section.

As a result of shear transformations, both the lower bainite and martensite exhibit lath structures, which are crystallographically similar to each other. However, a group of bainitic ferrite laths form a sheaf, which resembles a plate as compared to a packet in the martensite. When adjacent bainite sheaves grow in the same direction, their appearance is similar to a martensite packet (Fig. 13). Generally, however, the growth direction of a bainite sheaf is so arbitrary that the microstructure of lower bainite does not exhibit an obvious packet structure (Fig. 33). The lower bainitic structure also contains ~10 volume % of martensite in the form of islands. The presence of these islands may not be desirable for uniformity of the microstructure and hence uniformity of the properties.

The substructure of the laths consists of a high density of dislocations in both the lower bainite and the martensite as shown in the TEM micrographs. However, the dislocation density may be higher in the martensite because the dislocation density is known to increase as the transformation temperature decreases due to recovery (71).

Notable differences are observed in the morphology and shape
of the carbides within the laths. As already discussed, the intralath carbides in the lower bainite show a unidirectional morphology in comparison with the Widmanstätten carbides in the martensite laths. In addition, it can be noticed that the bainitic carbide is coarser and thicker than the Widmanstätten carbide (See, for example, Figs. 7 and 14). The shape of the bainitic carbide is rather similar to that of the carbide which is observed at the lath boundaries of the martensite tempered at 300°C [Fig. 29c and d]. This intralath carbide has been believed to form as a product of the decomposition of retained austenite [3, 6-8, 11-16]. Recent atom probe analyses [12, 25, 28, 29] have revealed a high degree of carbon buildup at the interface between the martensite matrix and the retained austenite, and as a consequence, the interlath carbide has been considered to nucleate at the austenite/martensite interface [12, 25]. The bainitic carbides, as discussed earlier, also nucleate at the austenite/martensite interface [12, 25]. The bainitic carbides, as discussed earlier, also nucleate at the austenite/ferrite interface. The similarity in the shapes of these two types of carbides, therefore, can be attributed to the fact that the interface is the nucleation site in both cases. In addition, the bainitic carbides form on a \(112_f\) plane. The Widmanstätten cementite particles in tempered martensite, on the other hand, form on \(011_f\) planes, which reportedly have a small misfit with the \(100_c\) plane of cementite [63], and hence are likely to develop the shape of a thin platelet. This difference in the habit plane can also be the reason for the thickening of the
bainitic carbides.

The decomposition of retained austenite films observed in the lower bainite is a similar structural change to that observed in tempered martensite. Shortly after the bainitic ferrite laths are formed, austenite is retained at the lath boundaries (Figs. 9, 10). The degree of retention seems even higher than as-quenched martensite (See Figs. 9 and 17). It is likely that the austenite is stable because of the buildup of transformation strains (mechanical stabilization) (6,25) and the partitioning of carbon (chemical stabilization) (6, 25, 47) as in the case of martensite. However, these stabilizing mechanisms would not be as intense as in the case of martensite. Firstly, because of the high transformation temperature, the strain buildup would not be so high. Secondly, the concentration of carbon in the austenite may not be so high because extensive precipitation of intralath carbides has already occurred. Although Bach (47) has reported on a 0.3%C-3%Cr-1.5%Mo steel that as much as 6-8 at. % of carbon was detected by an atom probe analysis in the retained austenite films at the boundaries of bainite laths, he has also admitted that the films are not continuous. Therefore, the austenite may have been enriched in carbon because its decomposition had already started. The fact that in this study the number of interlath carbides in the lower bainite is much less than observed in the tempered martensite indirectly shows that the carbon concentration in the retained austenite is initially low. In view of the fact that the austenite films in the lower bainite have become partially discontinuous after only a holding time of 10 minutes (Fig.12), it appears that the austenite can be stable
for a short time. After the isothermal holding is prolonged so that the matrix would become mostly bainitic, a large portion of the bainite has been "tempered," and hence austenite is no longer present. Only a small amount of bainite that has formed near the end of the isothermal holding is able to retain austenite. Therefore, a certain amount of austenite is still detected by X-ray measurements (Fig. 16). Because these fresh bainite regions are likely to be located near the martensite islands (i.e., previously untransformed austenite), they may not be revealed by thin-film electron microscopy; the bainitic microstructure is polished more rapidly than the martensitic structure during the foil preparation. In other words, in the predominantly bainitic microstructure (viz., after 1 hour of holding), the retained austenite is not distributed as uniformly as in the as-quenched lath martensite (Fig. 7).

Thus, the lower bainitic microstructure is different from the as-quenched martensitic microstructure, but, becomes more similar to that of martensite tempered in the TME range. The heat treatments leading to these microstructures are schematically illustrated in Fig. 45. These microstructures are summarized as follows:

**Lower bainite:** bainitic ferrite laths with coarse intra- and interlath carbides and non-uniformly distributed retained austenite.

**Tempered martensite:** martensite laths with the Widmanstätten intralath carbides plus coarse interlath carbides and no retained austenite.
As-quenched martensite: martensite laths with fine auto-
tempered intralath carbides and stable
interlath films of retained austenite.

4.2 The Correlation Between Microstructure and Mechanical
Properties

The yield strength of martensite in the as-quenched
condition is as high as that of the Mo containing steel.
Although the impact toughness in this condition is not quite as
high as that obtained for the Mo containing steel, the toughness
has been improved drastically by tempering at 200°C at little
expense to the hardness. However, tempering at a higher
temperature of 300°C has resulted in a considerable decrease in
the toughness as well as in a decrease in the hardness (TME). In
the case of lower bainite in the as-transformed condition, both
the toughness and strength are substantially lower than those of
the as-quenched martensite. The toughness decrease in these two
cases has occurred in opposite to the general rule of the inverse
relationship between strength and toughness (1). The cause of
the brittleness will be discussed in this section after the
toughening mechanisms of the lath martensitic structure are
specified.

4.2.1 Toughening Mechanisms of Lath Martensite

For the Fe/Cr/C base alloys, Rao [6] has pointed out the
following as the key factors for improved toughness in the lath
martensite: (i) dislocation substructure, (ii) small packet size,
(iii) elimination of undissolved carbides, (iv) presence of
stable retained austenite films and (v) precipitation of fine carbides.

As a result of the accommodation of the inhomogeneous shear upon the transformation, the martensite substructure generally consists of either dense tangles of dislocations or fine internal twins or both (1,2). The microtwins limit the number of available slip systems upon deformation, and hence lowers the toughness of martensite (1, 2, 58, 72). Internal twins are also preferred precipitation sites for coarse carbides upon tempering (1, 2, 6-8, 58, 72). For these reasons a dislocation substructure is greatly favored. As shown in Fig. 7, a heavily dislocated, almost twin-free substructure has been obtained in the present steel.

Grain refinement is one of the most effective means to enhance the impact toughness (73, 74). Rao and Thomas (75) have emphasized that the martensite packet boundaries are as capable as prior austenite grain boundaries in absorbing the energy of running cracks. It is clear in Fig. 33 that the refinement of prior austenite grains by lowering the austenitizing temperature is also effective in refining the packet size, and the effect of refinement is reflected in the improvement in the impact toughness values (Table 2).

Carlson et al. (5) have reported that in a Ti bearing steel, the toughness is enhanced by raising the austenitizing temperature and have attributed this effect to the dissolution of coarse Ti carbides. Chromium is also a strong carbide former (91), although not as strong as Ti; therefore, it has been recommended
that the Fe/Cr/C alloys should be austenitized at a high temperature, e.g. 1100°C. However, because this may result in grain coarsening, Rao et al. (4) have proposed the use of double austenitizing, viz., the first austenitizing at 1100°C to dissolve coarse carbides followed by the second austenitizing at 900°C for grain refinement. Although in the present steel, none of the microscopic observations have revealed the presence of coarse carbides after being austenitized at 900°C, they are likely to be present because the double austenitizing has shown to improve the toughness (Table 3).

The effect of retained austenite upon the toughness has been a subject of debate for years (3-10, 13, 76-80). For the present steel however, evidence for its beneficial effect has been observed. Figure 36 provides rather direct evidence, in which the retained austenite films at the lath boundaries are shown to be resistant to crack propagation. This resistance appears to be due to the effect of crack blunting (81), viz., plastic flow in austenite results in a decrease in the stress concentration at a crack tip. A recent in-situ deformation study of lath martensite on a high voltage electron microscope (87) has shown that in a tough condition plastic deformation can occur across the lath boundaries. The present observation is in good agreement with this result. As mentioned earlier, ferrite and retained austenite have a [111]a // [011]f relation through the lath boundary, and in this manner, the two phases maintain high coherency (6,22,87). Therefore, it is believed that with the interlath austenite films present, plastic flow at a crack tip can be easily transmitted from ferrite to austenite as
schematically shown in Fig. 46a. It has been reported that in cryogenic steels the retained austenite is mechanically unstable and transforms to martensite upon dynamic loading. (78, 79). However, the so-called retained austenite in cryogenic steels is actually produced by the precipitation (or inverse transformation) during tempering (76-80), and its morphology is in the form of discrete particles (78-80). On the other hand, the retained austenite in the present steel has survived the martensite transformation, and its morphology is thin, continuous films at the lath boundaries; therefore, the retained austenite may be highly stabilized. As shown in Fig. 36, the lath boundaries have stopped the propagation of the secondary crack even at temperatures as low as −196 °C. The retained austenite also has an indirect effect of toughening. As discussed earlier, the retained austenite films are enriched in carbon (12, 25, 28, 29, 87). Hence, it is likely that the actual carbon content in the matrix is lowered, and the toughness is increased. This is the same as the scavenging effect (80) in cryogenic steels.

It has been reported that the retained austenite in the present steel remains stable upon tempering at 200 °C (11, 12). However, the rapid increase in the toughness by this tempering cannot be explained in terms of retained austenite because no major microstructural changes have been observed with respect to the austenite in this condition (6-8, 11, 12). This implies that the improvement in the toughness due to the precipitation of fine carbides is also important. In his 0.3%C-4%Cr-2%Mn steel, Rao has observed a drastic toughness increase from almost no
ductility in the as-quenched condition to an upper shelf level after tempering at 200 °C. He (6, 7) has attributed the poor toughness in the as-quenched condition to the lack of auto-tempering and the improved toughness after tempering to the extensive precipitation of Widmanstatten cementite. When the \( M_f \) temperature is low, there is not sufficient time to allow carbon to diffuse and form clusters or precipitates. These carbon atoms locate themselves in the interstitial sites of the bcc crystal, distort the lattice (82) and hence make the dislocations immobile. The carbon atoms are also likely to interact with the dislocations by pinning (82), which renders the dislocations immobile. Therefore, the toughening due to auto-tempering is considered to be a process of increasing the mobile dislocation density through the relief of internal strains induced by the supersaturation of carbon (87). The degree of auto-tempering is obviously enhanced by raising the \( M_s \) and \( M_f \) temperatures (6-8), and in the present steel, a substantial amount of auto-tempered carbides is observed (Fig. 7) by raising the \( M_f \) to above 200 °C. Nevertheless, it has been also shown that the toughness can be further improved by the additional tempering at 200 °C, which suggests that the carbon atoms cannot be converted to a form of fine carbides by auto-tempering alone.

In reality, the toughness value is determined by the relative contribution of all the toughening factors mentioned above, some of which are contradicting each other. For example, a high temperature austenitization to dissolve coarse carbides causes grain coarsening in austenite. When the steel contains a strong carbide forming element like Mo (91), the effect
of the elimination of coarse carbides exceeds the detrimental
effect of grain coarsening. For the present steel, however, the
effect of grain refinement is more pronounced. Even though the
undissolved coarse carbides may have been present, the toughness
is shown to be enhanced by other factors, viz., grain refinement
and the presence of retained austenite and fine tempered
carbides. Grain refinement appears to increase the ductility as
well (Table 2) and possibly for this reason, the impact value in
the upper shelf region is further increased by grain-refinement
(e.g. at 200°C tempering, Fig. 32).

4.2.2 Tempered Martensite Embrittlement (TME)

The mechanism of TME has recently been studied using the
present steel (11). In this study (11), the phenomena of TME and
TE have been unambiguously differentiated, and it has been
concluded that TME is caused by the decomposition of retained
austenite films into coarse carbides at the lath boundaries
whereas TE is caused by the segregation of S to the prior
austenite grain boundaries.

The causes of TME can be understood in terms of the
initiation and propagation of cracks. The coarse interlath
carbides which has replaced the retained austenite films have
been identified as M₂₃C type. The structure of these carbides
is the same as cementite, but with ~15% of iron atoms being
replaced by chromium (11, 12). The role of a brittle carbide
for the initiation of a brittle fracture has been reported by
McMahon and Cohen (83), and it has been shown that the
probability of microcrack formation increases with the increasing
thickness of the carbides. Knott and co-workers (17, 84) have shown that the critical stress required to propagate a cleavage fracture from a cracked carbide is inversely proportional to the square root of the carbide width. Since the fracture upon the Charpy impact test is stress-controlled (85, 86), an increase in the number of coarse carbides leads to a decrease in the critical stress for the crack propagation as well as the increase in the number of crack nuclei. The coarse interlath carbides in the TME structure are thus considered to promote the initiation of brittle fracture.

Thomas et al. (87) have observed in an in-situ deformation experiment on a high voltage electron microscope that in the TME condition, a crack propagates in a zig-zag pattern, and its path is confined to a particular lath. The coarse stringers of interlath carbides have been considered to prevent plastic flow from crossing over the lath boundary although the Widmanstatten intralath carbides which form along the principal slip planes of the matrix, i.e., \(\{110\}_f\) planes, may not restrict the slip within the lath (12, 87). The lack of coherency at the lath boundary due to the absence of the retained austenite films would also render the cross-over of plastic flow across the boundary difficult. As a result, the plastic zone size at a crack tip would be limited by the width of lath as schematically shown in Fig. 46b. This leads to higher stress concentration at a crack tip than that in the as-quenched martensite, which would promote further crack propagation along the laths. The ridges observed in Fig. 34b are considered to be the result of this
crack propagation confined to a particular lath.

Several previous researchers (6-8, 13, 14, 16, 19) have argued that the interlath carbides also provide running cracks with an easy path, based upon the observation of ridge patterns in the fracture surface. In the present study, however, interlath fracture seems to be a fortuitous event and not a predominant fracture mode of TME (Figs. 37, 38). Therefore, it is concluded that the role of interlath carbides for crack propagation is restricting local plastic deformation rather than providing an easy crack path.

Thus, the causes of TME can be summarized to the crack initiation enhanced by the coarse interlath carbides and to the crack propagation promoted through the limited local plastic flow due to the absence of retained austenite and the presence of carbide stringers at the lath boundaries.

4.2.3 Lower Bainite

The lower bainitic microstructure has at least two advantageous characteristics for toughness as compared to the martensitic structure. Firstly, because of the formation of sheaves instead of packets, the matrix of bainite is divided more finely than that of martensite. Indeed, the impact toughness of the lower bainite shows little dependence on the prior austenite grain size (Table 2). Secondly, because of the higher transformation temperature, the dislocation density in the lower bainite is expected to be lower than that in the martensite (34).

Nevertheless, the impact toughness of the lower bainite is lower than that of the as-quenched martensite. Even the TME
structure exhibits higher impact values with slightly higher strength as compared to the lower bainite. Therefore, other differences in the microstructure must have detrimental effects upon the toughness.

The lower bainite contains ~10 volume % of martensite islands. However, Fig. 41a strikingly shows that the martensite island is tougher than the lower bainitic region. Therefore, these martensite islands cannot be the reason for the low toughness of lower bainite.

The remaining factors are, as in the case of TME, the absence of retained austenite films and the presence of coarse carbides. In fact, retained austenite is present in the lower bainite. It is, however, not uniformly distributed as already discussed. Therefore, it would not effectively act in preventing crack propagation.

Earlier, it was shown that coarse carbides promote the initiation of brittle cracks, and in the lower bainite, coarse carbides are present not only at lath boundaries but also within the laths. As discussed earlier, these unidirectional carbides are thicker than the intralath carbides in the martensite probably because they initially form at the austenite/ferrite interface and have a (112)$_f$ habit plane rather than a (110)$_f$ type. Therefore, in the lower bainite, the number of possible sites for the crack initiation is much greater than in the TME structure.

Irvine and Pickering (88) have reported that in their low carbon bainitic steels, the toughness of upper bainite is lower
than that of lower bainite and have attributed it to the difference in the morphology of carbides. The intralath carbides in the lower bainite are finer than the interlath carbide films in the upper bainite, and therefore, the former is tougher than the latter. If the same comparison is made between lower bainite and dislocated lath martensite, the carbides in the martensite are still finer than those in the lower bainite. Therefore, it may not be surprising that the martensite is tougher than the lower bainite.

In addition to crack initiation, the lower bainitic structure appears to promote crack propagation more readily than the as-quenched or tempered martensitic structure. The intralath unidirectional carbides in the lower bainite form along a particular \( \{112\}_f \) plane, and hence they are likely to block local slips on \( \{110\}_f \) planes unlike the Widmanstätten carbides along \( \{110\}_f \) planes in the martensite. Therefore, as illustrated in Fig. 46c, the plastic zone size at a crack tip in the lower bainite may be limited by the interparticle spacing of the intralath carbides, which is smaller than the lath width. This explains why the toughness of lower bainite is even lower than that of the TME structure. The stress concentration at the carbides may lead to the cracking of carbides for further crack propagation. This is suggested by Figs. 39 and 41, where the cracks proceed across the unidirectional carbides.

Thus, the coarse unidirectional carbides in the lower bainite play a role similar to that of the interlath carbides in the TME structure. The fact that the double austenitizing treatment has decreased the toughness of lower bainite (Table 3)
is explained in terms of an increase in the number of coarse carbides, i.e., the extra carbon, which went into solution during the first austenitizing, has contributed to the formation of coarse bainitic carbides after the second austenitizing.

In summary, the low toughness of lower bainite can be attributed to the absence of retained austenite and the presence of coarse carbides. These reasons are eventually the same as those for TME, although the location of coarse carbides is different between the lower bainite (both inside the laths and at the lath boundaries) and the TME structure (at the lath boundaries).

4.3 Alloy Design Perspectives for Engineering Applications

4.3.1 Martensite

From the current results, the following can be suggested with respect to the alloy design viewpoint, i.e., (i) the elimination of Mo and (ii) the selection of low austenitizing temperature.

The present steel (0.3%C-3%Cr-2%Mn steel) has shown no detrimental effects upon the mechanical properties due to the elimination of Mo. The only pronounced effects of Mo are the enhancing the temper resistance and the suppressing of TE above 400°C. However, this steel will not be used in this temperature range because of the decrease in the strength as well as the presence of two types of embrittlement, viz., TME and TE. Therefore, the addition of Mo is not necessary.

For the increased toughness, the optimum heat treatment
condition is quenching from 900°C and subsequent tempering at 200°C. The Mo containing steel is able to provide high impact toughness only by single austenitizing at 1100°C (8), and the requirement of tempering for the present steel does not seem to be advantageous. However, in the light of the cost of energy, this low temperature austenitizing-tempering process is more economical than heating the furnace up to 1100°C. Moreover, from the quality assurance point of view, it is better to obtain both stable retained austenite and fine tempered carbides by tempering at 200°C rather than relying upon auto-tempering in the as-quenched condition. Although the low temperature austenitizing cannot dissolve coarse carbides, the toughness increase by the tempering can overcome the effect of undissolved carbides. Tensile ductility is also improved by low temperature austenitizing.

However, when a low austenitizing temperature is chosen, precautions must be taken in order to avoid an overheating problem. As shown in Appendix 1, the formation of intergranular precipitates due to high temperature soaking and subsequent slow cooling has to be prevented.

A process, which prevents overheating and is even more economical, is to combine the heat treatment with hot rolling, i.e., direct quenching after hot rolling, thereby, the steel does not have to be reheated for austenitizing. The applicability of direct quenching is studied briefly in Appendix 2. Although more studies are needed, the present result appears promising.
4.3.2 Lower Bainite

In the present steel, lower bainite exhibits lower toughness as well as lower strength than lath martensite. Therefore, for high strength applications, the lower bainitic structure is not favorable.

The only way to make use of the lower bainitic structure would be to alter the alloy design criterion from the high strength oriented to the high toughness oriented. One of the advantages in using high strength structural steels is that they are able to withstand high load per unit area and hence allow the wall thickness of structural components to be reduced. This leads to the reduction in the net weight of materials and consequently in the cost of materials. However, when high toughness which exceeds the capability of martensitic high strength steels is required, the concept of alloy design has to be changed in such a way that the high toughness is achieved by drastically lowering the strength and that the loss of strength is compensated by increasing the wall thickness. In this case, the lower bainitic structure is more adequate than that of martensite for two reasons. Firstly, it is obviously difficult to attain a fully martensitic microstructure throughout a large cross-section. Secondly, upon tempering at 650 °C, the impact toughness of lower bainite is much higher than that of martensite as shown in Fig. 32.

This high toughness of lower bainite is considered to be due to the coarsening of carbides [82], the recovery of the matrix (71, 82) and relatively low susceptibility to temper embrittle-
ment (89). Although the hardness is rather low in this condition (Fig. 31), uniformity in the bainitic microstructure can be accomplished more readily than the martensitic structure, and hence, uniform mechanical properties may be achieved over a wide range of wall thicknesses. Thus, for heavy-gauge, low strength, high toughness applications, the lower bainite tempered at high temperatures (e.g. 650°C) is favorable.
5. SUMMARY AND CONCLUSIONS

In the scope of the present study on the microstructure-mechanical property relationships in a martensitic and lower bainitic 0.3%C-3%Cr-2%Mn steel, it is possible to conclude the following:

1. Microstructure and mechanical properties of the martensite
   a. The microstructure in the as-quenched condition consists of heavily dislocated martensite laths containing autotempered carbides with retained austenite films at the lath boundaries; the elimination of Mo appears to have little effect on the microstructure.
   b. In the as-quenched condition after austenitizing at 900 °C, the yield strength is ~200 ksi (1379 MPa), and the Charpy impact toughness is ~24 ft-lb (32.4 J). Both these values would indicate that little effect is observed through the elimination of Mo.
   c. After austenitizing at 1100 °C, the impact toughness in the as-quenched condition is substantially lowered by the elimination of Mo.
   d. Upon tempering, the hardness decreases monotonically, whereas the impact toughness experiences a drastic increase at 200 °C and subsequent two troughs due to tempered martensite embrittlement (TME) and temper embrittlement (TE), respectively.
e. TME appears to be caused by the presence of coarse carbides and the absence of retained austenite at lath boundaries. The brittle fracture mode is transgranular quasi-cleavage.

f. The temper resistance above 400 °C is decreased, and the susceptibility to TE is enhanced apparently due to the absence of Mo.

g. As far as the service temperature is limited to ambient temperatures, the present steel is suitable for high strength applications. The optimum heat treatment condition for toughness is austenitizing and quenching from 900 °C and tempering at 200 °C.

2. Characterization of the lower bainitic microstructure

a. The lower bainite is observed as a sheaf-shaped "plate" on the optical microscopic scale; however, when examined on a finer scale (i.e., electron microscopy), the sheaf is found to be divided into lath-like subunits.

b. After 10 min. of isothermal holding at 360 °C, the bainite transformation has barely begun. In this condition, the lower bainitic structure consists of highly dislocated bainitic ferrite laths containing unidirectional intralath carbides with retained austenite films at the lath boundaries.

c. After 1 hour of isothermal holding at 360 °C, the matrix becomes predominately lower bainitic. In this condition, retained austenite has decomposed into interlath carbides. The microstructure consists of
dislocated bainitic ferrite laths with coarse carbides (identified as cementite) both inside the laths and at the lath boundaries, although the number of interlath carbides appear to be much less than intralath ones.

d. Lath boundaries are, in most cases, parallel to \([110]_f\), which is apparently parallel to \([111]_a\) in the parent phase. This suggests the presence of the \([111]_a\) type habit plane which is typical of the transformation of lath martensite. Primary exceptions to this rule are the existence of \([112]_f\) boundaries in the case of twin-related laths and a rare \([103]_f\) type.

e. Both the Kurdjumov-Sachs and the Nishiyama-Wasserman variants are observed in parallel laths within the same bainite region. This implies that the orientation relationship between austenite and bainitic ferrite varies from the K-S through the G-T to the N-W as in the case of lath martensite.

f. Three types of arrangement of adjacent laths are observed, i.e., (i) clustering around a certain pole (little orientation change), (ii) abrupt orientation change between \(<111>_f\) and \(<100>_f\), (iii) twin-related laths. All of these arrangements have also been found in lath martensite and may indicate the strain minimization upon the bainite transformation.

g. The Isaichev orientation relationship:

\[
\begin{align*}
(111)_f & \parallel (010)_c \\
(011)_f & \parallel (103)_c
\end{align*}
\]
is found between bainitic ferrite and cementite instead of the Bagaryatskii orientation relationship which is often observed in tempered martensite.

h. The habit plane of cementite particles is found to be close to one of \( \{112\}_f \) planes as compared to the \( \{110\}_f \) type habit of the Widmanstatten cementite in tempered martensite.

i. The bainite transformation at 360°C is considered to be a shear transformation of austenite, the rate of which is controlled by the formation of cementite at a moving austenite/ferrite interface, followed by the decomposition of austenite trapped along lath boundaries into interlath cementite particles.

3. Applicability of the lower bainite for heavy-gauge, high strength uses

a. When compared with the as-quenched martensite, the lower bainitic structure isothermally held at 360°C for 1 hour exhibited substantially lower impact toughness as well as lower strength than the martensitic structure. Toughness is lowered by double austenitizing.

b. The reasons for the low toughness of the lower bainite appear to be the presence of coarse carbides both inside the laths and at lath boundaries and the absence of retained austenite films at lath boundaries. These reasons are eventually the same as those for TME.

c. Upon tempering, the hardness and impact toughness values of the lower bainite are relatively unchanged up
to 400 °C, which suggests that no major microstructural changes have occurred during this range of tempering.

d. Upon tempering above 400 °C, the impact toughness of the lower bainite recovers more rapidly than that of the martensite because of its relatively smaller susceptibility to TE.

e. For high strength uses, the lower bainitic microstructure is not favorable. However, if high strength is not required, i.e., if a structure component is designed to support the load due to its large wall-thickness rather than through its high strength, then the lower bainite tempered at a high temperature may be able to accomplish uniformity in the microstructure and properties better than the martensite.
APPENDIX 1. An Overheating Problem with Homogenization

The homogenization of steels, which is prolonged soaking at high temperatures usually followed by slow cooling, is often conducted to eliminate or alleviate the problem of inhomogeneity in the initial microstructure. Although homogenization is one of the most effective ways to obtain a uniform microstructure (A1) and to improve mechanical properties (A2), it also has disadvantages with respect to steels. Firstly, a soaking at high temperatures causes grain-coarsening. Secondly, most of the inclusions such as carbides, sulfides, oxides and nitrides would be dissolved at high temperatures, which could lead to the segregation of those impurity elements along coarsened austenite grain boundaries during homogenization or subsequent slow cooling. This may result in the re-precipitation of inclusions along the grain boundaries and as consequence in the low-energy, intergranular microdimple fracture on an impact test. This is known as the classical problem of overheating (A3, A4), and it has indeed been found to occur in the present alloy when the steel was homogenized after the rough forging.

One way to avoid the undesirable effect of homogenization would be to reheat the steel up to the homogenization temperature and then to cool it rapidly so that those inclusions along prior austenite grain boundaries could be dissolved upon heating and would not form again afterwards. Another way would be to break up the coarse structure by hot rolling or forging. These two methods have been applied to eliminate the overheating problem, and the results are now described.
After being received in the rough-forged condition, the steel was homogenized at 1200°C for 18 hours followed by furnace cooling before it was taken out of the furnace at around 800°C. Three test blanks were taken from the material after homogenization. The first blank was made into oversized Charpy specimens, and those specimens were subjected to a single austenitizing at 900°C for 1 hour followed by oil quenching. The second blank was also made into oversized Charpy specimens, and they were austenitized at 1200°C for 1 hour, oil-quenched, re-austenitized at 900°C for 1 hour and again oil-quenched. The last test blank was heated up to 1200°C, soaked for 1 hour, then hot-rolled with a finishing temperature above 900°C followed by air-cooling. Oversized Charpy specimens were prepared from the rolled plate and subjected to single austenitizing at 900°C for 1 hour followed by oil-quenching.

Fig. A1.1 shows the optical microstructure after those treatments. Since the final heat treatment in every case is an oil-quenching from 900°C austenitizing, the microstructure is, without exception, fine-grained martensite. However, discontinuous lines which appear to be portions of grain boundaries are revealed by etching in the single-austenitized specimen (indicated by the arrows in Fig. A1.1a). Neither the double-austenitized nor the hot-rolled specimen exhibits such boundaries. Also, it should be noticed that if those lines are prior austenite grain boundaries, the grains are too coarse to be formed by an austenitizing at 900°C.

Table A1.1 shows the results of a hardness measurement and a Charpy impact test. In spite of little difference in hardness,
the impact value in the single-austenitized condition is considerably lower than other conditions.

Fractography has revealed coarse intergranular fracture in the single-austenitized specimen (Fig. A1.2a). However, the grain boundaries are not as flat as those observed in the case of temper embrittlement (e.g. Fig. 42c). They consist of shallow microdimples and rib-like furrows in essentially a fibrous fracture mode. A large number of relatively globular inclusions such as A in Fig. A1.2c were found in the microdimples whereas the rib-like furrows seemed to be accompanied by flaky inclusions of irregular shapes (e.g. B in Fig. A1.2c). An energy dispersive X-ray (EDX) analysis indicates that the globular inclusion A is MnS, whereas the analysis on the flaky particle B has detected only the signals of Fe, Cr and Mn, which are the major alloying elements of the steel. An analysis by auger electron spectroscopy was not successful because of the contamination of the fracture surface although it detected a weak S peak. From the appearance of the intergranular fracture, the grain size is of the order of 100 μm, which is in good agreement with the size of the grains revealed in optical microscopy.

From these results, it is certain that these coarse grains were formed during the homogenization process, and not during the final austenitizing. The inclusions at these grain boundaries are apparently the source of the decohesion, making these boundaries preferential sites for crack propagation upon impact testing. They are also prone to the preferential attack during etching for optical metallography.
As shown in Fig. A1.3, both the double-austenitized specimen and the hot-rolled specimen do not present an intergranular fracture mode consistent with the results of impact testing. Therefore, the segregation and precipitation which are observed in the single-austenitized specimens must have taken place during slow cooling after homogenization, and thus can be avoided by either rapid cooling or subsequent hot working.

The cause of overheating has been attributed to the dissolution, segregation and re-precipitation of sulfides, especially MnS (A3, A4) and CrS (A4), or carbides (A6). Although the present steel contains both Mn and Cr in substantial amounts, it appears from the EDX analysis that only MnS plays an important role in the problem of overheating. This is consistent with the fact that the standard free energy of formation of MnS is lower than that of CrS (A4) and also with the observation by McLead and Nutting (A7). Yet, the reason for the unusual rib-like fibrous fracture on the grain boundaries is unknown and is to be clarified.

It may be significant to point out that the detrimental effect of overheating on toughness remains even if the material undergoes another austenitizing afterwards unless those intergranular inclusions are once again dissolved. If austenitizing at low temperatures is required as a final heat treatment, the preceding heat cycle must be carefully designed. Since the austenitizing at 900°C is preferred for the present steel, it is not adequate to supply the steel in the as-homogenized condition to the final heat treatment.
It is also worth noting that overheating is a reversible phenomenon. Therefore, even though an overheating problem has already occurred, it can be eliminated either by re-austenitizing and rapid cooling or by hot working. If the material is thick, such rapid cooling from a high austenitizing temperature is associated with a risk of quench cracking. Hot working such as rolling or forging, however, can be applied to steels of large dimensions. The present steel was subjected to another forging treatment after overheating was observed. The second forging followed by no homogenization removed the overheating problem as was reflected in the results shown in a previous chapter.

In industry the addition of rare earth metals (REM) (AB) has successfully ceased overheating through the formation of REM sulfides which are stable even at high temperatures in the austenite region.
APPENDIX 2. Direct Quenching Experiment on a 0.3%C-3%Cr-2%Mn Steel

In previous chapters it was shown that the mechanical properties of martensite after the final quenching, in particular toughness, are in some cases apparently affected by the products of the preceding heat treatments such as undissolved carbides and/or overheated grain boundaries. In Appendix 1, hot rolling was proved to be an effective way to eliminate these influences from the microstructures prior to the final heat treatment. Moreover, hot rolling could be an economical process of heat treatment, i.e., a steel plate would not have to be reheated for austenitizing if it was directly quenched immediately after its final dimension is reached. For this reason, a hot rolling-direct quenching process was attempted with a 0.3%C-3%Cr-2%Mn steel.

A test blank which was initially 1.25 inches (32 mm) thick was soaked at 1150 °C for 1 hour then rolled down to 0.5 inches (12.7 mm) thickness in two passes. The finish-rolling temperature was controlled at 900 °C so that austenite could be recrystallized before quenching. After the finish-rolling, the steel plate was plunged into an oil bath. The whole process is illustrated in Fig. A2.1.

As also shown in Fig. A2.1, the resulting impact value is remarkably high as compared to the reheated and quenched material (e.g. Table 2) while hardness remains at the same level. According to the optical micrographs in Fig. A2.2, prior austenite grains have not been recrystallized and remained coarse. Never-
theless, the directly quenched material exhibited a high impact value possibly because other toughening processes were taking place. Transmission electron micrographs revealed extensive precipitation of auto-tempered Widmanstatten carbides together with the presence of retained austenite films at lath boundaries.

In order to apply direct quenching to real steel production processes, the effects of rolling schedule, finish rolling temperature and cooling rate still have to be clarified. However, these present results indicate that direct quenching is a promising method by which the desirable microstructure of lath martensite can be obtained economically. Since the hardenability of this steel is high, not only oil-quenching, but also forced-air cooling or even still-air cooling can produce this structure depending upon the plate thickness. In other words, the steel can possess a combination of high strength and high toughness even in the as-rolled condition, if its thickness is very small.
REFERENCES


55. G. V. Kurdjumov and G. Sachs, Zeit. fur Phys. 64, 325 (1930).


### Table 1 Chemical composition of the steel.

<table>
<thead>
<tr>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>P</th>
<th>S</th>
<th>Cr</th>
<th>Mo</th>
<th>Al</th>
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<td>0.29</td>
<td>0.01</td>
<td>2.10</td>
<td>0.003</td>
<td>0.007</td>
<td>2.97</td>
<td>0.01</td>
<td>0.027</td>
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(in wt. %)
Table 2 Mechanical properties of martensite and lower bainite.

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<th></th>
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<th></th>
<th></th>
<th></th>
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<td>Martensite</td>
<td>1100</td>
<td>205.3 (1415.5)</td>
<td>274.4 (1892.0)</td>
<td>8.1</td>
<td>15.4</td>
<td>49.2</td>
<td>16.1 (21.8)</td>
<td></td>
</tr>
<tr>
<td>(As-quenched)</td>
<td>900</td>
<td>197.8 (1363.8)</td>
<td>270.8 (1867.2)</td>
<td>13.9</td>
<td>38.7</td>
<td>49.6</td>
<td>23.9 (32.4)</td>
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</tr>
<tr>
<td>Lower Bainite</td>
<td>1100</td>
<td>160.5 (1106.6)</td>
<td>213.5 (1472.0)</td>
<td>9.9</td>
<td>19.8</td>
<td>43.7</td>
<td>11.3 (15.3)</td>
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<tr>
<td>(As- transformed)</td>
<td>900</td>
<td>157.7 (1087.3)</td>
<td>211.8 (1460.4)</td>
<td>12.4</td>
<td>36.0</td>
<td>45.0</td>
<td>11.9 (16.1)</td>
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<tr>
<td>Tempered**</td>
<td>900</td>
<td>178.6 (1231.4)</td>
<td>216.1 (1489.9)</td>
<td>15.3</td>
<td>58.6</td>
<td>44.4</td>
<td>24.2 (32.8)</td>
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<td>Martensite (8)</td>
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<td>200.0 (1378.9)</td>
<td>253.0 (1744.3)</td>
<td>6.3</td>
<td>-</td>
<td>48.5</td>
<td>28.7 (39.0)</td>
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<tr>
<td>(with 0.5%Mo)</td>
<td>900</td>
<td>199.0 (1372.0)</td>
<td>259.0 (1785.7)</td>
<td>8.9</td>
<td>-</td>
<td>45.0</td>
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</tr>
</tbody>
</table>

*Isothermally transformed at 360°C for 1 hour.

**Tempered at 300°C for 1 hour.
Table 3 Effect of double austenitizing on hardness and impact toughness in the untempered conditions.

<table>
<thead>
<tr>
<th>Microstructure</th>
<th>Austenitizing</th>
<th>Hardness $H_{RC}$</th>
<th>Charpy Impact Value at R. T. ft-lb [J]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Single</td>
<td>49.6</td>
<td>23.9 (32.4)</td>
<td></td>
</tr>
<tr>
<td>Martensite</td>
<td>Double*</td>
<td>49.7</td>
<td>28.3 (38.4)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Single</td>
<td>45.0</td>
<td>11.9 (16.1)</td>
<td></td>
</tr>
<tr>
<td>Lower Bainite</td>
<td>Double*</td>
<td>42.2</td>
<td>6.6 (8.9)</td>
</tr>
</tbody>
</table>

*1st austenitizing: 1200°C for 1 hour followed by oil quenching.

2nd austenitizing: 900°C for 1 hour followed by oil quenching or isothermal holding.
Table 4 Change in hardness and impact toughness upon tempering.

<table>
<thead>
<tr>
<th>Microstructure</th>
<th>Tempering* Temperature °C</th>
<th>Hardness HRC</th>
<th>Charpy Impact Value at R.T. ft-lb [J]</th>
</tr>
</thead>
<tbody>
<tr>
<td>As-quenched</td>
<td></td>
<td>49.6</td>
<td>23.9 (32.4)</td>
</tr>
<tr>
<td></td>
<td>200</td>
<td>47.4</td>
<td>48.0 (65.1)</td>
</tr>
<tr>
<td></td>
<td>300</td>
<td>44.4</td>
<td>24.2 (32.8)</td>
</tr>
<tr>
<td>Martensite</td>
<td>400</td>
<td>42.1</td>
<td>20.8 (28.2)</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>37.2</td>
<td>13.9 (18.8)</td>
</tr>
<tr>
<td></td>
<td>650</td>
<td>23.6</td>
<td>29.8 (40.4)</td>
</tr>
<tr>
<td>As-transformed</td>
<td></td>
<td>45.0</td>
<td>11.9 (16.1)</td>
</tr>
<tr>
<td></td>
<td>200</td>
<td>43.1</td>
<td>23.4 (31.7)</td>
</tr>
<tr>
<td>Lower Bainite</td>
<td>300</td>
<td>43.0</td>
<td>16.8 (22.8)</td>
</tr>
<tr>
<td></td>
<td>400</td>
<td>42.4</td>
<td>16.4 (22.2)</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>38.9</td>
<td>19.4 (26.3)</td>
</tr>
<tr>
<td></td>
<td>650</td>
<td>20.6</td>
<td>116.0 (157.3)</td>
</tr>
</tbody>
</table>

*Austenitizing: 900 °C for 1 hour followed by oil quenching or isothermal holding.

Holding time of tempering is 1 hour for all the temperatures.
Table A1.1  Effect of thermal and mechanical treatments after homogenization on hardness and impact toughness.

<table>
<thead>
<tr>
<th>Heat Treatment after Homogenization</th>
<th>Hardness $H_{RC}$</th>
<th>Charpy Impact Value at R.T. ft-lb [J]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single Austenitizing</td>
<td>51.0</td>
<td>10.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(13.6)</td>
</tr>
<tr>
<td>Double Austenitizing*</td>
<td>49.7</td>
<td>27.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(37.4)</td>
</tr>
<tr>
<td>Hot Rolling and Single Austenitizing</td>
<td>50.5</td>
<td>25.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(35.0)</td>
</tr>
</tbody>
</table>

*Same as in Table 3.
FIGURE CAPTIONS

Fig. 1. Specimen configurations. (a) Dilatometer specimen (b) tensile specimen (c) Charpy impact specimen.

Fig. 2. Typical dilatation curves showing a considerable decrease in the amount of expansion with the increase in temperature.

Fig. 3. Incomplete TTT diagram of the steel.

Fig. 4. Change in the optical microstructure during the isothermal holding at 360°C. (a) 10 min. (b) 20 min. (c) 40 min. (d) 1 hour (e) 4 hours (f) 24 hours. Austenitized at 900°C. Etchant: 5% Nital.

Fig. 5. Change in the volume fraction of bainite during the isothermal holding at 360°C. Austenitized at 900°C.

Fig. 6. Microstructure of the steel in the as-quenched condition. (a) Optical micrograph (b,c) SEM micrographs (d) BF image by TEM. Etchant: 5% Nital.

Fig. 7. Transmission electron micrographs of the steel in the as-quenched condition. (a) BF (b) DF from (002) reflection (c) SAD pattern (d) analyzed pattern.

Fig. 8. Microstructure of the steel after isothermal holding at 360°C for 10 min. (a) Optical micrograph (b) SEM micrograph showing lower bainite sheaves. Etchant: 5% Nital.

Fig. 9. Transmission electron micrographs of the steel after isothermal holding at 360°C for 10 min. (a) BF. A bainite region growing from a prior austenite grain boundary is shown by dashed lines. (b) DF from an
austenite reflection.

Fig. 10. Transmission electron micrographs of the steel after isothermal holding at 360 °C for 10 min. (a) BF showing another bainite region (b) DF from \((311)_{a}\) reflection (c) DF from \((121)_{f}\) in \([\overline{1}13]_{f}\) zone (d) DF from \((200)_{f}\) in \([011]_{f}\) zone (e) SAD pattern (f) indexed pattern.

Fig. 11. The same area as Fig. 10 at a higher magnification. (a) BF (b) DF from an austenite reflection.

Fig. 12. Transmission electron micrographs of the same bainite region as Fig. 10 near the tip of the region. (a) BF (b) DF from an austenite reflection (c) DF from a carbide reflection.

Fig. 13. Microstructure of the steel after isothermal holding at 360 °C for 1 hour. (a) Optical micrograph (b, c) SEM micrographs (d) BF image by TEM. Etchant: 5% Nital.

Fig. 14. Transmission electron micrographs of the steel after isothermal holding at 360 °C for 1 hour. (a) BF (b) DF from a carbide reflection.

Fig. 15. BF image showing martensitic regions formed from untransformed austenite during quenching from the isothermal holding.

Fig. 16. Change in the amount of retained austenite with holding time at 360 °C.

Fig. 17. Change in the amount of retained austenite in bainite with the holding time at 360 °C.
Fig. 18. Transmission electron micrographs of the steel after isothermal holding at 360°C for 1 hour. (a) BF (b) DF from (113)_c reflection (c) DF from (220)_a austenite reflection (d) indexed SAD pattern.

Fig. 19. Stereographic projection showing the Isaichev orientation relationship between ferrite and cementite.

Fig. 20. Microdiffraction patterns from the laths near the area shown in Fig. 18.

Fig. 21. Microdiffraction patterns from the area in Fig. 22.

Fig. 22. Transmission electron micrographs of the steel after isothermal holding at 360°C for 1 hour. (a) BF (b) SAD pattern (c) indexed pattern.

Fig. 23. Transmission electron micrographs of the steel after isothermal holding at 360°C for 1 hour. (a) BF (b) DF from a cementite reflection (c) microdiffraction from the matrix (d) microdiffraction from the matrix and cementite.

Fig. 24. Transmission electron micrographs of the steel after isothermal holding at 360°C for 1 hour. (a) BF (b) SAD pattern (c) indexed pattern (d - i) DF images from respective reflections.

Fig. 25. Microdiffraction patterns from the area in Fig. 24 showing that laths are twin-related. M: matrix T: twin.

Fig. 26. Transmission electron micrographs of the steel after isothermal holding at 360°C for 1 hour. (a) BF (b) DF from both (011)_T and (122)_c reflections (c) SAD pattern
(d) analysis of the pattern.

Fig. 27. Transmission electron micrographs of the steel after isothermal holding at 360 °C for 1 hour. (a) BF (b) DF from (122)\textsubscript{c} reflection. (c) SAD pattern (d) analysis of the pattern.

Fig. 28. Stereographic projection showing the result of trace analysis to determine the habit plane of cementite in the ferrite matrix.

Fig. 29. Microstructure of the steel quenched and tempered at 300 °C (in the TME condition). (a) Optical micrograph (b,c) SEM micrographs (d) BF by TEM. Etchant: 5% Nital.

Fig. 30. Charpy impact toughness transition curves of martensite and lower bainite.

Fig. 31. Change in hardness of martensite and lower bainite with tempering temperature.

Fig. 32. Change in Charpy impact toughness at room temperature with tempering temperature.

Fig. 33. Optical micrographs of martensite in the as-quenched condition (a and b) and lower bainite (c and d). (a,c) austenitized at 1100 °C (b,d) austenitized at 900 °C. Etchant: 5% Nital.

Fig. 34. Fractographs of Charpy specimens broken at room temperature. (a) As-quenched (b) quenched and tempered at 300 °C (c) isothermally transformed at 360 °C.

Fig. 35. Fractographs of Charpy specimens broken near the lower shelf. (a) As-quenched (b) quenched and tempered at 300 °C (c) isothermally transformed at 360 °C.
Fig. 36. SEM micrograph of a Ni plated broken Charpy specimen in the as-quenched condition. The principal crack direction is from the top to the bottom of the photograph.

Fig. 37. SEM micrographs of a Ni plated broken Charpy specimen after quenched and tempered at 300 °C. Arrows indicate interlath portions of the crack paths.

Fig. 38. Two-face fractographs of a broken Charpy specimen after quenched and tempered at 300 °C.

Fig. 39. SEM micrograph of a secondary crack in a broken Charpy specimen after isothermally transformed at 360 °C.

Fig. 40. Two-face fractographs of a broken Charpy specimen after isothermally transformed at 360 °C.

Fig. 41. SEM micrograph of a Ni plated broken Charpy specimen after isothermally transformed at 360 °C. M: martensite island.

Fig. 42. Fractographs after tempering. [a,c,e] Martensite tempered at (a) 300 °C (c) 500 °C (e) 650 °C. (b,d,f,g) Lower bainite tempered at (b) 300 °C (d) 500 °C (f,g) 650 °C. All fractured at room temperature.

Fig. 43. Stereographic projection showing three orientation relationships, i.e., Kurdjumov-Sachs between austenite and ferrite, Isaichev between ferrite and cementite and Pitsch between austenite and cementite.

Fig. 44. Stereographic projection showing an orientation relationship between ferrite and cementite which would arise if the Pitsch orientation relationship between austenite and cementite and the Nishiyama-Wasserman orientation
relationship between austenite and ferrite were assumed.

Fig. 45. Superimposed TTT diagrams for the bulk steel and hypothetical steels having the compositions of retained austenite phases in lower bainite and martensite, respectively.

Fig. 46. Schematic illustrations showing the effect of microstructure on the plastic zone at a crack tip. (a) As-quenched martensite (b) tempered martensite (TME) (c) lower bainite.

Fig. A1. 1. Optical micrographs of the steel. (a) Single austenitized (b) double austenitized (c) hot rolled and single austenitized after homogenization. As-quenched. Etchant: 5% Nital.

Fig. A1. 2. Fractographs of a broken Charpy specimen single austenitized after homogenization. (a–c) SEM images (d) EDX spectrum of inclusion A in (c) (e) EDX spectrum of inclusion 8 in (c).

Fig. A1. 3. Fractographs of broken Charpy specimens. (a) Double austenitized after homogenization (b) hot-rolled and single austenitized after homogenization.

Fig. A2. 1. Heat diagram of the hot-rolling - direct quenching process and mechanical properties in the as-quenched condition.

Fig. A2. 2. Optical micrographs of the directly quenched steel at different magnifications.

Fig. A2. 3. Transmission electron micrographs of the directly quenched steel. (a) BF (b) DF from an austenite reflection (c) DF from a carbide reflection.
Fig. 1
Austenitizing: 1100°C

\[ \frac{\Delta L}{L_0} \times 10^3 \]

Time (sec)

370°C

402°C

490°C

Fig. 2
Austenitizing: 1100°C

Fig. 3
Fig. 6
Fig. 16

Time (min)

Volume % Austenite

Oil
Water
As Quenched
Fig. 19

• hkl Ferrite
• hkl Cementite

XBL831-5159
Fig. 23
Fig. 24
Fig. 26
Double diffraction

Fig. 27
Fig. 28
Martensite
- 0.3 C/3 Cr/2 Mn Fine grained
- 0.3 C/3 Cr/2 Mn/0.5 Mo Coarse grained

Lower Bainite
- 0.3 C/3 Cr/2 Mn Fine grained

Hardness, $R_c$

Transformed
Tempering Temperature ($^\circ$C)

Fig. 31
Fig. 32

Martensite
- 0.3C/3Cr/2Mn Fine grained
- 0.3C/3Cr/2Mn/0.5Mo Coarse grained

Lower Bainite
- 0.3C/3Cr/2Mn Fine grained

V Charpy Impact Value at Room Temperature (ft-lb) vs. Tempering Temperature (°C)
Fig. 33
Fig. 34
Fig. 42
Fig. 43
Fig. 44
a) As-quenched Martensite

b) Tempered Martensite (TME)

c) Lower Bainite

Fig. 46
Fig. A1.2
1150 °C

1st pass: 36% reduction

2nd pass: 38% reduction at 900 °C

Charpy impact value at R.T: 31.6 ft-lb (42.8 J)

Hardness: 49.0 H<sub>Rc</sub>

Fig. A2.1
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