

Bainite transformations in hypoeutectoid steels

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SYNOPSIS

Kinetic and structural aspects of bainite formation have been studied by stepped transformation experiments. Growth of lower bainite stops if the temperature is raised to the upper range, and upper bainite either stops growing or grows at a rate substantially below that of lower bainite when the temperature is reduced suddenly to the lower range. It is concluded that the internal structural differences which characterize upper and lower bainite exert a significant influence on the rate at which these products develop.

In the steels studied in this investigation, lower bainite plates thicken from one side only. These plates exhibit a substructural unit oriented at an angle to the growth direction and these units appear to nucleate at the immobile side of the plates. Edgewise growth rates measured on the hot stage thus may reflect primarily the rate of nucleation of these substructural units. A similar situation may prevail in the growth of upper bainite needles.

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INTRODUCTION

THE ACICULAR MODES of austenite decomposition in hypoeutectoid steels encompass Widmanstätten ferrite, upper and lower bainite, and the several forms of martensite. Each of these reactions exhibits the surface relief indicative of a displacive transformation. While growth kinetics provide a convenient criterion for distinguishing the martensitic from the higher temperature decomposition modes, it is less clear whether or on what basis a distinction should be made between Widmanstätten ferrite, and upper and lower bainite.

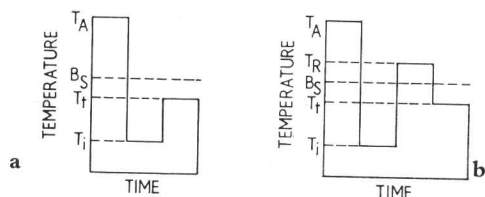
Within the bainite temperature range, crystallographic and morphological features of the transformations point to two basically different forms of bainite depending on the reaction temperature. Upper bainite exhibits a $(111)\gamma$ orientation habit analogous to low-carbon martensite whereas lower bainite develops with the more irrational habit characteristic of high-carbon martensite.¹⁻³ This has led to the suggestion that upper and lower bainite may be structurally closely related to low-

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and high-carbon martensite respectively,^{4,5} and this appears to be confirmed by transmission microscopy studies.⁵⁻⁸ The morphological features of the two forms of bainite⁹ also point to structural relationships between the two forms of bainite and the corresponding forms of martensite, respectively.

The kinetic features of the transformations serve to characterize further the two modifications of bainite. The activation energy determined from overall reaction rates differs for the formation of upper and lower bainite^{10,11} and the time law which describes the overall reaction also differs for the two forms.^{10,12} However, these differences may arise from changes in nucleation rates, in growth rates, or in both. Although studies of growth kinetics¹³ suggest a relatively sharp change in the temperature dependence of the edgewise growth rates of upper and lower bainite, this can be interpreted in terms of a single model which considers that the growth of Widmanstätten ferrite as well as upper and lower bainite is controlled by the diffusion of carbon through austenite away from the advancing edge of a plate.¹⁴⁻¹⁶ Thus, it is not yet clear in what way, if at all, the structural differences between the two forms of bainite influence the reaction kinetics.

The pronounced structural and kinetic differences between upper and lower bainite require further clarification. Stepped quenching experiments provide a method for exploring the kinetics of the transformations and a number of investigations employing these techniques have been reported. Up-quenching experiments conducted with a plain carbon eutectoid steel¹⁷ have demonstrated that lower bainite leads to a retardation of transformation in the upper range, thus indicating that the reactions in the two ranges are not additive. Other investigations on alloy steels^{18,19} point to a marked acceleration of upper bainite by prior transformation at a lower temperature. These investigations have only studied the overall reaction kinetics so that separation of the nucleation and growth contributions to the reaction has not been possible. In the present work, step quenching experiments involving dilatometry to study the overall reaction, and hot-stage metallography to study growth kinetics, have been conducted in an effort to explore further the significance of the kinetic and structural differences between upper and lower bainite.



1 Schematic illustration of heat treating cycles

MATERIALS AND PROCEDURE

The alloys employed in this investigation are listed in Table I and were received from commercial suppliers as 3/4in diameter rounds. The dilatometric and hot stage microscopy techniques have been described previously^{13,20} and will not be repeated here.

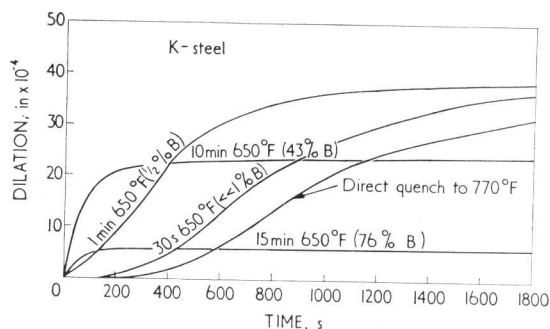
The heat treating procedures employed to study the influence of thermal history on reaction kinetics are illustrated schematically in Figure 1. For the dilatometric studies, austenitizing was conducted for 10 min at 2000°F (TA) in a purified nitrogen atmosphere and transformation was conducted in molten salt baths controlled at the desired temperatures. In the hot-stage microscopy studies all treatments were conducted in the hot stage evacuated to approximately 5×10^{-5} mmHg.

RESULTS

Influence of up-quenching on overall reaction rate

The thermal cycle illustrated in Figure 1a was employed to study the influence of prior transformation at Ti (either in the lower or the upper bainite range) on upper bainite formation at Tt. In each of the steels listed in Table I the rate of the upper bainite reaction was increased significantly by prior transformation to lower bainite, as was shown earlier.^{18,19} Typical results are presented in Figure 2 which compares direct isothermal transformation at 770°F (Tt) with that resulting after prior transformation at 650°F (Ti). The acceleration of the upper bainite reaction produced by the prior treatment is a function of time (extent of decomposition) at Ti. This relationship is illustrated more clearly in Figure 3 which presents the initial rate of transformation at Tt (corrected for the extent of transformation during pretreatment) as a function of the pretreating time. The extent of decomposition during prior treatment is indicated on this figure for each pretreating time.

Several important features of the acceleration phenomenon are shown in Figure 3. The initial rate of transformation to upper bainite attains a maximum after an intermediate time at Ti, and the time for this maximum acceleration corresponds approximately with that for the maximum rate of transformation at the pretreating temperature. It is also apparent that transformation to upper bainite is not induced by the last few



2 Acceleration of transformation at 770°F resulting from prior treatment at 650°F

percent of decomposition at the pretreating temperature. Similar results were observed when Ti and Tt were both within the upper bainite range. Thus, up-quenching will not increase the extent of decomposition to bainite beyond the maximum attainable at the pretreating temperature.

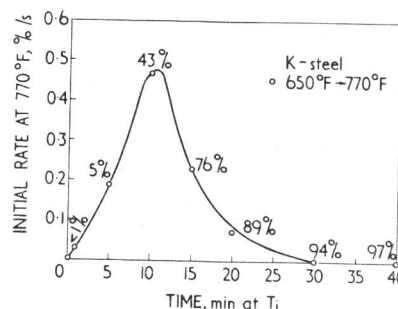
Hot stage metallographic studies

It must be anticipated that prior decomposition accelerates reaction at a higher temperature primarily by influencing nucleation of transformation rather than by modifying the growth rate.¹⁹ Up-quenching experiments were conducted on the hot stage in order to confirm this and to examine the relationship between the initial and final transformation products. Studies were conducted in which Ti and Tt were both in the same temperature range (either that for upper or for lower bainite), and in which lower bainite was initiated at Ti and then decomposition continued in the upper range.

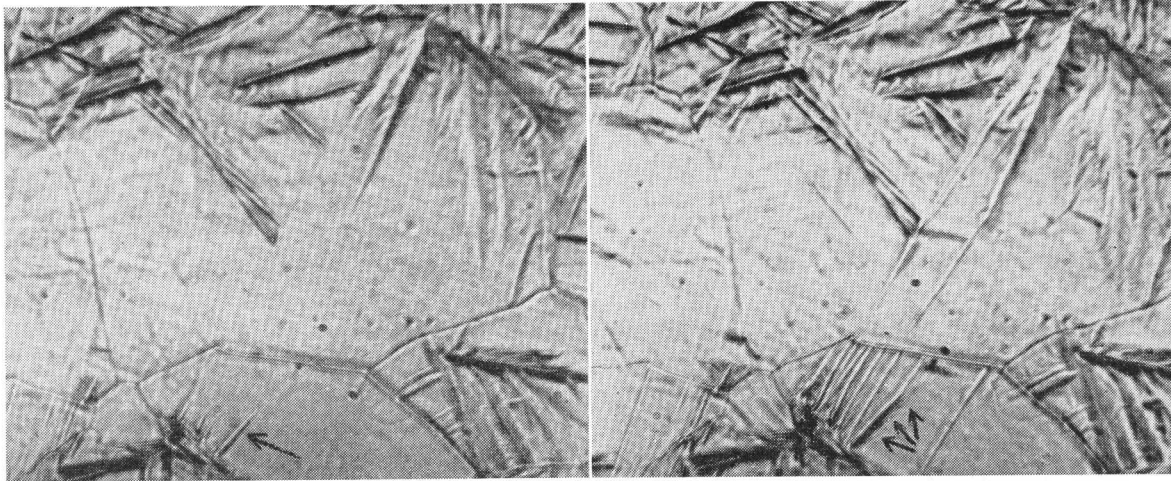
When the initial and final temperatures were both in the same temperature range growth of the transformation product upon change of temperature continued without interruption and without change of direction but at the rate characteristic of direct transformation at the higher temperature. In contrast to this behaviour, edgewise growth of lower bainite generally was stopped completely when the temperature was raised suddenly to the upper range. Occasionally growth continued but with a change of direction and at the rate characteristic of upper bainite. This has been reported previously and a typical example is presented in Figure 4.¹³ Although growth of upper bainite from the edges of lower bainite plates was rare, copious formation of

TABLE I Analysis of steels

Designation	Composition, wt-%					
	C	Si	Mn	Ni	Cr	Mo
K	0.31	0.30	0.76	3.07	1.22	0.49
M	0.69	0.70	0.85	1.80	0.85	0.81
N	0.50	0.36	0.68	8.70	—	—



3 Influence of prior treatment at a lower temperature on the initial rate of transformation at 770°F



a 5 min, 2000°F→11 h, 520°F; steel N × 460 **b** 5 min, 2000°F→11 h, 520°F→21 min, 750°F; steel N × 460

4 Influence of stepped transformation on the growth of bainite

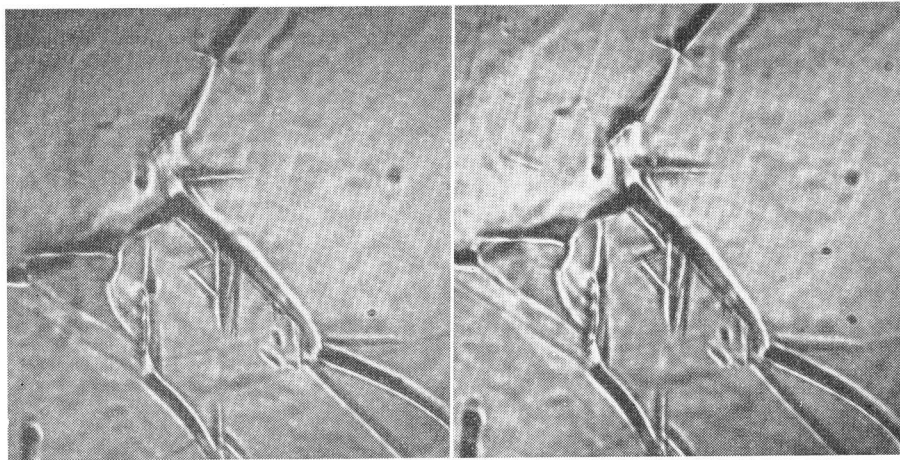
upper bainite needles developed on the sides of virtually all lower bainite plates. This is illustrated most clearly on the plate designated by the arrow in the lower left corner of Figure 4a and b. Since prior transformation has not modified the isothermal growth rate at the higher temperature, the acceleration of the overall reaction associated with up-quenching results from an increased number of growing needles.

The fact that a morphological change is required for transformation to continue when the temperature is raised to the upper range emphasizes the importance of the structural difference between the two forms of bainite. While only incomplete evidence for this structural difference has so far been obtained,⁵⁻⁸ the principal difference between the two forms appears to reside in internal twins in lower bainite and the absence of these twins in upper bainite. On up-quenching, either the twinning mode of transformation becomes inoperative and the interface dis-

sociates into a large number of needles of upper bainite, or independent nucleation of these needles is catalysed by transformation strains in the region of the lower bainite/austenite interface.

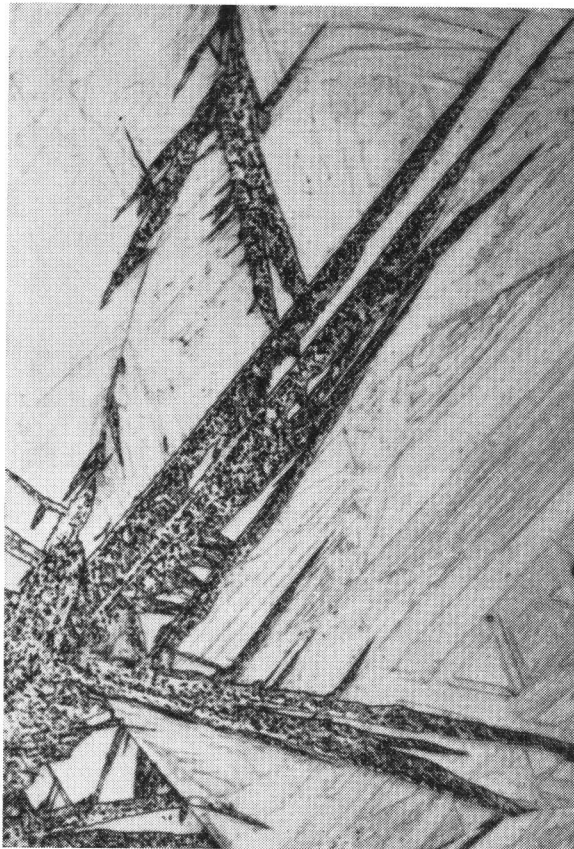
Treatment in the bay

If transformation strain is the primary factor responsible for the acceleration, then an annealing treatment at a higher temperature should relax these strains and reduce or eliminate acceleration. This assumes, however, that transformation can be stopped by heating to some temperature above B_s but below the A_1 temperature in order to allow relaxation to occur. To explore this possibility, transformation to lower bainite was interrupted by raising the temperature above B_s . Figure 5 shows hot stage micrographs for the N steel ($B_s=815^\circ\text{F}$), demonstrating that transformation is stopped completely when the temperature is



a at 500°F, 6½ min before temperature change **b** at 900°F, 55 min after temperature change

5 Interruption of growth of lower bainite by heating to temperature above B_s × 460



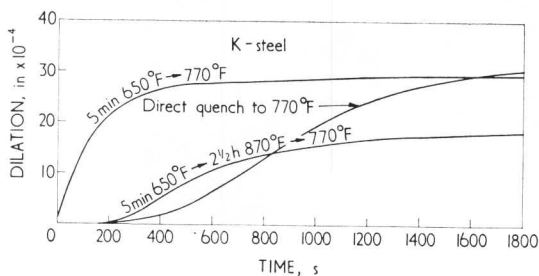
6 Microstructure of hot-stage sample; lower bainite initiated at 500°F and transformation then interrupted by heating to 900°F (above B_s); etched 10% chromic acid $\times 1000$

raised from 500° to 900°F. No growth of these plates was observed on holding for longer than 2 h at 900°F.

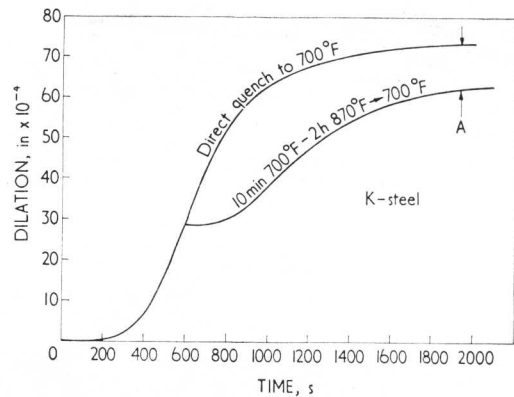
Optical metallography confirms that treatment in the bay has stopped transformation completely (Figure 6). Growth of upper bainite was also stopped when the temperature was raised above B_s . Thus, in these alloy steels, treatment in the bay can be used to relax transformation strains without additional decomposition occurring during the relaxation treatment.

Relaxation of transformation strain

The heat treating cycle illustrated in Figure 1b was employed to substantiate the important role of transformation strain in these



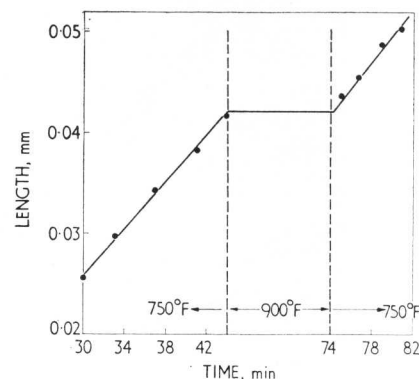
7 Influence of annealing after prior treatment on the course of reaction at 770°F



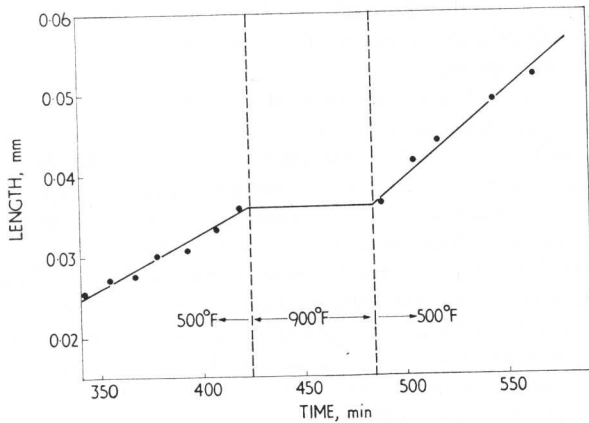
8 Retardation of transformation resulting from an intermediate annealing treatment

up-quenching experiments. As demonstrated in Figure 7, insertion of an annealing treatment in the bay between the initial and final transformations virtually eliminates the acceleration of upper bainite. As a special case of the heat treating cycle represented in Figure 1b, T_i may be chosen equal to T_f , and Figure 8 illustrates the influence on the course of the subsequent transformation of interrupting decomposition by holding for 2 h at a temperature above B_s . In spite of the relatively high reaction rate at the moment of interruption, decomposition after the annealing treatment begins at an extremely low rate.

Relaxation experiments using heat treating cycles analogous to those reported in Figure 8 were conducted on the hot stage to determine to what extent the annealing treatment interfered with growth of the prior transformation product. Figures 9 and 10 show that annealing is virtually without effect on the growth of either upper or lower bainite. In both cases growth started again immediately upon returning to the original transformation temperature, and the growth rate was basically the same before and after the annealing treatment. Since the pre-existing transformation product begins to grow at its normal rate as soon as the temperature is reduced to its original value, it is concluded that the relaxation treatment lowers the nucleation rate, presumably by removing the self-catalysing influence of transformation strain.



9 Influence of treatment above B_s on growth of upper bainite at 750°F in steel N



10 Influence of treatment above B_s on growth of lower bainite at 500°F in steel N

Elimination of this catalysing influence by the annealing treatment produces significant stabilization of the bainite reaction as shown in Figure 8. Here, distance A represents the amount of stabilized austenite. The relaxation which occurs at temperatures above B_s undoubtedly also accompanies decomposition at temperatures below B_s . This appears to be intimately associated with the incomplete nature of the bainite reaction and with the stabilization of this reaction observed when decomposition at a higher temperature precedes reaction at a lower temperature.²¹

The B_s temperature

The ability to stop transformation to bainite by raising the temperature above B_s emphasizes the significance of this temperature in the formation of bainite. The behaviour described here for the N steel has been observed in a wide variety of steels containing sufficient amounts of alloying elements to produce a bay in the TTT diagram. Nevertheless, Widmanstätten ferrite forms in many of these steels at temperatures in the proeutectoid ferrite range. Thus, B_s in these alloy steels does not represent an upper temperature limit for the formation of coherent ferrite as has been suggested for plain carbon steels.¹⁶ In spite of the experimental difficulty in determining B_s in plain carbon steels, this temperature may have as much significance in these materials as it does in alloy steels.

The inability of upper bainite to grow when the temperature is raised substantially above B_s^* is analogous to the corresponding inability of lower bainite to grow when the temperature is increased above some rather ill-defined temperature near 350°C . In both instances, this may be associated with the nature of the lattice invariant strain (slip or internal twinning) required for the specific mechanism that controls the displacive transformation.

Both B_s and the incomplete nature of the bainite reaction appear to be intimately associated with nucleation of this mode of austenite decomposition. This is illustrated most clearly,

* Growth of upper bainite will continue at temperatures not too far above B_s . This is consistent with the observation that deformation at temperatures just above B_s produces bainite,²² and this appears to parallel the influence of deformation on M_s .

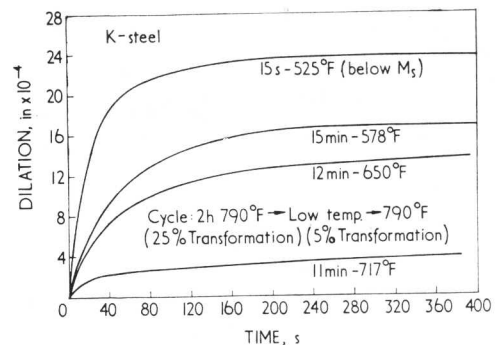
perhaps, by allowing upper bainite to proceed to its maximum possible extent at a temperature between B_s and B_f . As demonstrated in Figure 11, additional transformation at this temperature can then be induced by an inoculation treatment which introduces a few percent of transformation at a lower temperature. The substantial quantities of upper bainite induced in this way at the original temperature indicate clearly that the incomplete nature of the bainite reaction results from a limitation on nucleation of transformation.

DISCUSSION

The fact that bainites in steel are aggregate structures has caused considerable difficulty in deciding upon the reaction mechanism. The extent to which cooperative growth of the ferrite and carbide phases and the way in which diffusion of carbon control the process represent some of the major unresolved questions. It is generally agreed that ferrite is the leading phase in the formation of both upper and lower bainite and current models propose that diffusion of carbon in austenite ahead of an advancing interface controls edgewise growth.^{14-16,23} Upper bainite presumably forms as a stack of such ferrite plates, with carbon accumulating in austenite trapped between the plates in the stack. This austenite subsequently decomposes to ferrite and carbide by a process in which cooperative growth of these phases plays an essential role.¹⁴ This picture appears to agree well with the electron microscope observations on upper bainite, and copper-aluminium alloys may also decompose in basically the same way.²⁴

Growth of lower bainite has been described by an essentially similar two-stage model. Here, edgewise growth of isolated ferrite plates is considered to be controlled by diffusion of carbon through austenite, while thickening of these plates is accomplished by cooperative growth of ferrite and carbide.^{23,25} Replica electron micrographs agree with many aspects of this two-stage model in that the tip of lower bainite plates is often free of precipitate while carbides are in contact with the sides of the plates.²⁵

Alternative views regarding the transformation mechanism emphasize the many similarities between bainites and martensites. In these models lower bainite is considered to form as supersaturated ferrite with a carbon content not basically different from that of the original austenite, and carbides subsequently precipitate from this ferrite.²⁶ In the formation of

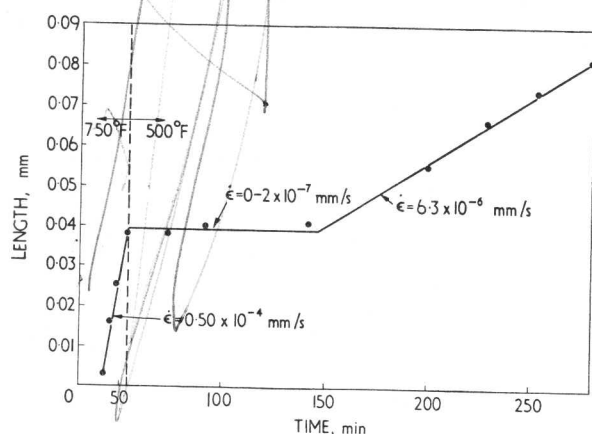


11 Re-initiation of transformation at 790°F by low-temperature treatment

upper bainite, diffusion of carbon in austenite ahead of the interface is presumably required to permit the structure typical of 'low-carbon martensite' to form,⁴ and the transformation is completed by decomposition of carbon-enriched austenite into a ferrite-carbide aggregate. In hypoeutectoid steels, however, it is not clear that upper bainite could not also form initially with substantial supersaturation (perhaps approaching that of the original austenite).²⁰ Clarification of this question awaits additional information on whether the transition from the 'low' to the 'high' carbon martensite structure is dictated by temperature²⁷⁻²⁹ or by composition and on what condition determines the rate at which bainites (and martensites) grow.

Diffusional growth models¹⁴⁻¹⁶ assume that edgewise growth is accomplished by the uniform advance of a coherent or semi-coherent interface at a rate determined by the diffusion of carbon into the surrounding austenite. At constant temperature these models provide a relationship between the growth rate and the radius of curvature at the edge of the plate, and it is assumed that the actual growth rate is determined by the condition that the radius of curvature adopts a value which results in a maximum growth rate. Thus, these models assume that the factors determining the structure and morphology of the transformation product are without influence on the growth rate. This question deserves further evaluation.

Upper and lower bainite sometimes can be identified with separate C curves in TTT diagrams.¹⁷ Since the diffusion model predicts the same growth rate for upper and lower bainite at any particular temperature, the nucleation rate may decide which form arises. The existence of both forms over a narrow temperature range is consistent with this viewpoint. Therefore, a plate of bainite initiated in the lower range should continue to grow when the temperature is raised to the upper range. The stepped quenching experiments demonstrate, however, that this does not occur, in spite of the fact that lower bainite continues to grow when the temperature is increased to a higher temperature within its own range. In the same way, upper bainite essentially stops growing if the temperature is reduced suddenly to the lower range. Figure 12 presents the results of an experiment in which transformation was initiated on the hot stage at a temperature in the upper range and the temperature then changed to the lower range. The needle of upper bainite increased in



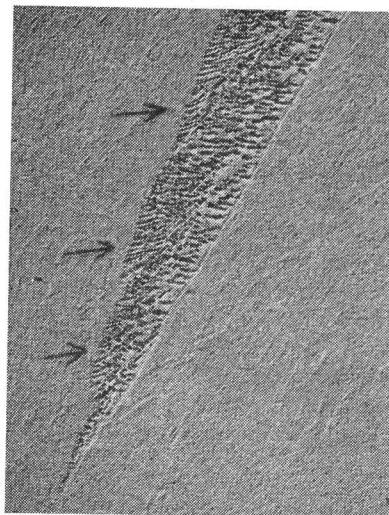
12 Influence of temperature change on growth of bainite in steel N; transformation initiated at 750°F and continued at 500°F

length by less than 2 μm (if at all) during the 1½ h period before growth of a lower bainite plate from the tip of the needle became apparent.

The lower bainite plate grows at a rate at least 30 times faster than that of the upper bainite needle. Conceivably, interface mobility might exert a controlling role in the growth process; however, metallographic studies suggest that edgewise growth is not accomplished by a mechanism involving the simple advance of an interface under diffusion control.

Study of sidewise growth or thickening of lower bainite plates helps to clarify the mechanism by which edgewise growth takes place. It has generally been assumed that lower bainite plates thicken by advancement of the interface on both sides of the plate. This would be expected, for example, if thickening is controlled by the cooperative growth of a ferrite-carbide aggregate.^{14,23,25} This is frequently not the case, however. On the hot stage lower bainite plates exhibit a 'midrib' which appears to separate regions which have transformed with opposite tilts, but this midrib is observed only very rarely in etched microstructures. In addition, measurements on the hot stage suggest that, for the steels examined in this investigation, the plates thicken from one side only.³⁰ The midrib then represents one edge of the plate and the opposite side constitutes austenite which has possibly deformed by a kinking process.⁴ Immobility of one side of the plate can also be recognized in optical and replica micrographs, as illustrated in Figure 13. Here, the straight edge of the plate constitutes the immobile interface and growth has been accomplished by advancement of the curved interface. These straight-sided plates are equally apparent in Figure 6 for lower bainite formed in a different steel.

Lower bainite plates exhibit a substructural unit which appears to constitute the basic growth unit; several are illustrated by arrows in Figure 13. These units appear to nucleate at the immobile side of the plate and advance at an angle to its major growth direction. Thus, lengthening of the plate may be dictated largely by the rate at which these units are generated, while thickening is controlled by the growth rate of the substructural units.



13 Lower bainite formed at 650°F in steel M

× 5000

Carbide particles extend up to the austenite-bainite interface at the end of each of these substructural units (Figure 13). Thus, propagation of the units may occur by cooperative growth of a ferrite-carbide aggregate in the manner proposed previously for the thickening process.²⁵ It is important to note, however, that plates of high-carbon martensite appear to exhibit a substructure similar to, if not identical with, that observed here in lower bainite. While a clear-cut distinction between the cooperative growth and martensitic mechanisms cannot yet be made, the structural similarities between martensite and bainite favour the latter mechanism. Upper bainite also exhibits substructural units analogous to those in low-carbon martensite.⁷ In this case, the substructural units are aligned parallel to the growth direction. It is conceivable that the substructural units in both upper and lower bainite develop at the high velocities characteristic of martensitic transformations, so that edgewise growth rates measured on the hot stage may reflect the rate at which these units nucleate.

Growth of martensite also may be accomplished by repeated nucleation of substructural units, and the piecewise fashion in which the reverse martensite reaction takes place in iron-nickel alloys³¹ seems consistent with this concept. In this case the different growth rates for bainites and martensites arise from factors controlling this nucleation process.

SUMMARY AND CONCLUSIONS

Stepped quenching experiments have been employed to examine the influence of the structural differences between upper and lower bainite on transformation kinetics. Bainite plates initiated at a temperature in the lower range do not continue to grow when the temperature is raised suddenly to the upper range. However, if the new temperature is below B_s , many needles of upper bainite nucleate around the lower bainite plate. Growth of both upper and lower bainite is stopped completely if the temperature is raised above B_s , but growth of either form will start again upon returning to the original temperature. It is concluded that upper bainite will not grow at some temperature just slightly above B_s and that lower bainite will not grow at temperatures above about 350°C. These temperatures appear to be associated with the mode of lattice invariant deformation - twinning in the case of lower bainite and slip in the case of upper bainite - involved in the displacive formation of the ferritic phase.

Growth of lower bainite in these steels appears to be accomplished by the repeated nucleation of a substructural unit which propagates at an angle to the major axis of the plate. As a result,

lower bainite plates were observed to thicken from only one side, and the edgewise growth rate appears to be dictated largely by the rate at which these sub-units nucleate. Since upper bainite exhibits substructural units analogous to those in low-carbon martensite, growth of this product also may be controlled by a repeated nucleation process.

ACKNOWLEDGMENTS

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Discussion 5

Chairman: Dr T. Broom (CEGB)

Dr T. Boniszewski (BWRA) referred to the 0.6% carbon steel which could be transformed to lower bainite, and asked, if the same steel was quenched to martensite and tempered to the same hardness or tensile strength level, what sort of impacts were obtained from the lower bainite as compared with tempered martensite.

Dr K. J. Irvine (The United Steel Cos. Ltd) replied that they were worse.

Dr H. I. Aaronson (Scientific Laboratory, Ford Motor Co., Michigan) said that Professor Hehemann and his co-authors had shown that plates of upper bainite nucleated at the sides of lower bainite plates, following an appropriate increase in reaction temperature after transformation had been initiated in the lower bainite region in an alloy steel (Figure 4). They also described upper bainite as consisting of stacks (sheaves) of ferrite plates, and discussed lower bainite in terms of repeated nucleation of substructural units (Figure 13). Transformation at a constant temperature within the lower bainite range also appeared to have involved nucleation of bainite plates at the interphase boundaries of previously formed bainite plates (Figure 6). This type of phenomenon had been reported some years ago in both the pre-eutectoid ferrite and the bainite reactions in a plain carbon steel and was given the name 'sympathetic nucleation'.¹ It was his aim to point out that this term referred to a specific type of solid-solid nucleation, sufficiently distinctive in character to warrant the general usage of this special designation.

When a bainite plate grew into its parent austenite the carbon content of the austenite abutting the plate was substantially increased. The finding that the rate of lengthening of bainite plates could be adequately accounted for on the assumption that essentially equilibrium partition of carbon took place between austenite and the ferritic component of bainite^{2,3} and the observation frequently made by both optical and replication electron microscopy that the martensite (formed during quenching) at the sides of bainite plates showed clear evidence of substantial carbon enrichment strongly supported this statement. The nucleation of another bainite plate at the interphase boundary of a bainite plate previously formed accordingly must be considered to take place at a site where the supersaturation of the austenite relative to the ferritic component of bainite was greatly reduced relative to positions well removed from this boundary. The high rates of nucleation often observed to occur at such boundaries, despite the small volume free energy change attending these nucleation events,* indicated that some other factor operative at these

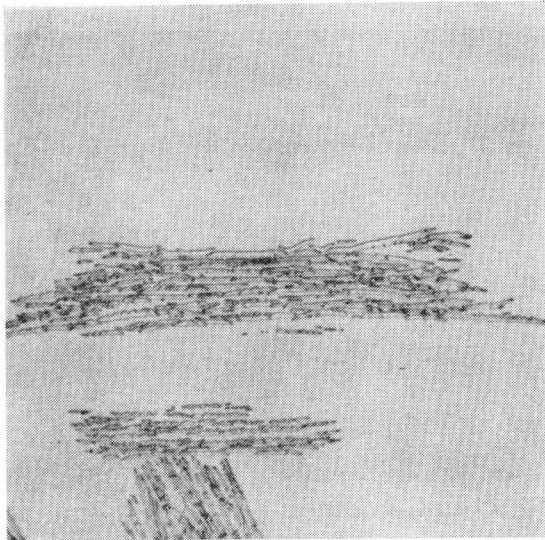
* In the limit, the supersaturation for such nucleation might be represented by no more than the minute departure from the A_{e3} necessary to permit the activity gradient across the boundary required for growth.

boundaries was very strongly encouraging nucleation. The term 'sympathetic nucleation' thus seemed appropriate as recognition of both the compositionally unusual nature of such nucleation and the role which the first-formed bainite plate obviously played in counterbalancing the small supersaturation sufficiently to make nucleation at these sites a kinetically competitive process.

A detailed (though essentially qualitative) analysis of sympathetic nucleation suggested that the interfacial free energy supplied by the pre-existing austenite/bainite (actually austenite/ferrite) boundary was responsible for overcoming the disadvantage with respect to supersaturation.^{1,4} This explanation was principally supported by the circumstance that the net interfacial free energy required for nucleation was raised to the third power in the exponential term of nucleation rate equations, whereas the volume free energy change was taken only to the second power in this term. The tendency of sympathetic nucleation to occur in a repetitive manner, to the point of building up extensive structures, such as sheaves, which were composed of considerable numbers of equivalently oriented individual crystals, was readily explained on this basis.^{1,4}

One might argue that the presence of carbides at the austenite/ferrite boundaries of bainitic structures could drastically change the situation with regard to supersaturation of the austenite in contact with the boundary. This argument could presumably be pursued to the point where the carbides were of submicroscopic dimensions, and was therefore probably most satisfactorily answered by the sheaf structures of Figures A and B. These sheaves were similar to those found in the bainite reaction. They were composed, however, of hcp α -crystals in a bcc β -matrix in Ti-Mo and Ti-Mo-Al alloys. In the temperature-composition region in which these specimens were reacted, the phase diagrams were similar to those of Fe-Ni alloys. Thus, no intermetallic compounds were present; the carbon, oxygen, and nitrogen contents of these alloys were all below the generous solubility limits of these elements in titanium-base alloys. Particularly since the comparatively large scale of these structures rendered a/a boundaries visible to careful inspection in a number of cases, sympathetic nucleation seemed the most probable cause of their formation. Sheaves and other sympathetically nucleated structures appeared with considerable frequency in titanium-base alloys which had undergone the $\beta \rightarrow \alpha$ transformation.⁵

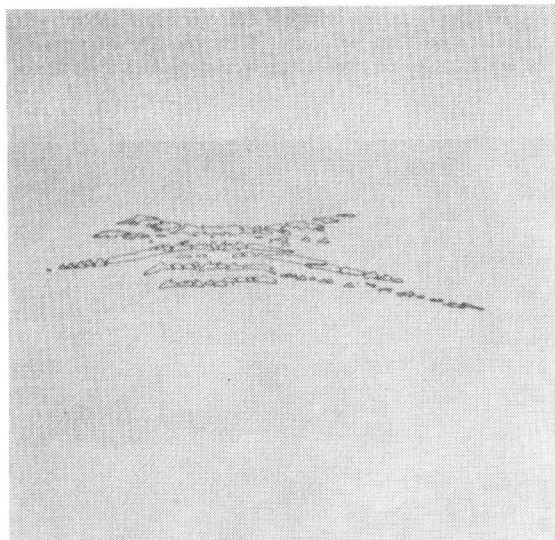
Professor R. W. Hehemann (Case Institute of Technology) replied that he agreed with Dr Aaronson that the term sympathetic nucleation could be used to describe the self-catalysing aspects of nucleation in these reactions. It was not clear, however, that this should be attributed primarily to the interfacial free energy supplied by the pre-existing austenite/bainite boundary. Rather, transformation strains appeared to exert the predominant



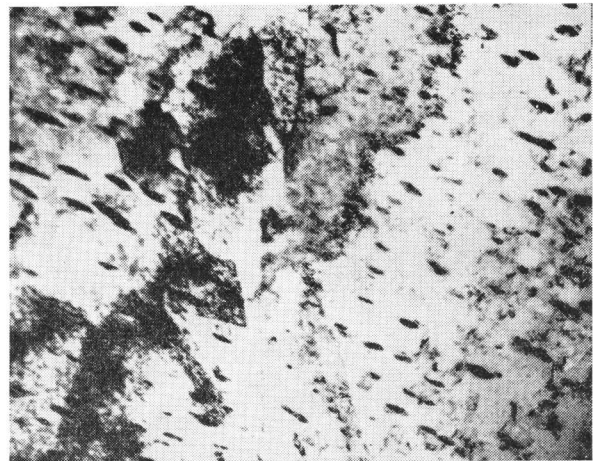
A Ti-10%Mo reacted 3 h at 725°C; solution annealed 40 min at 1000°C, etched in 60% glycerine, 20% HF, 20% HNO₃ × 1000

effect in a manner analogous to that in which plastic deformation of austenite nucleated bainite.

The important role of transformation strains in sympathetic nucleation was revealed most clearly in the relaxation experiments. Figures 7 and 8 demonstrated that annealing at a temperature above B_s reduced substantially the ability of previously existing bainite to catalyse nucleation. This reduction in the rate of nucleation occurred in spite of the fact that the carbon content at the interphase boundary should have been reduced during the annealing treatment, thereby creating a more favourable supersaturation state. The annealing treatment reduced the nucleation rate as would be expected if transformation strain rather than interfacial energy was of primary importance.



B Ti-10%Mo-3.5%Al reacted 3 h at 825°C; solution annealed 40 min at 1000°C, etched in 60% glycerine, 20% HF, 20% HNO₃ × 500



C 0.2% C, transformed at 450°C × 40000

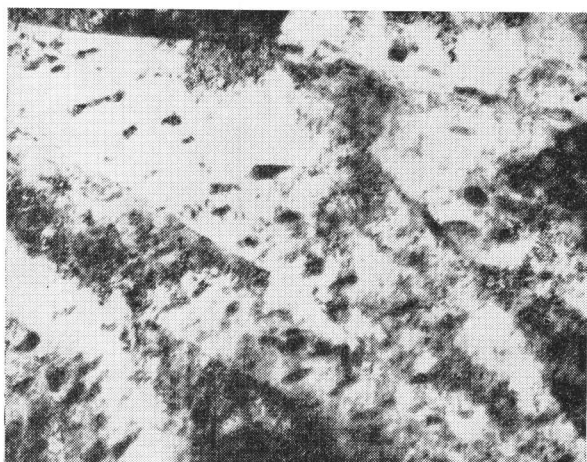
Mr F. B. Pickering and **Mr B. R. Clark** (The United Steel Cos. Ltd) said that Dr Shackleton and Dr Kelly were to be congratulated on an excellent piece of work which had gone far to clarify and, in many respects, to confirm much of the earlier electron microscopical work done on bainite. They were completely in agreement with the argument that the orientation relationship between the ferrite and cementite in bainite largely determined whether the structure was upper or lower bainite.

All the work they had carried out on the electron microscopy of thin foils of bainite in steels containing between 0.10% and 1.0% C had confirmed that, as found by Shackleton and Kelly, in lower bainite irrespective of carbon content, the cementite-ferrite orientation relationship was:

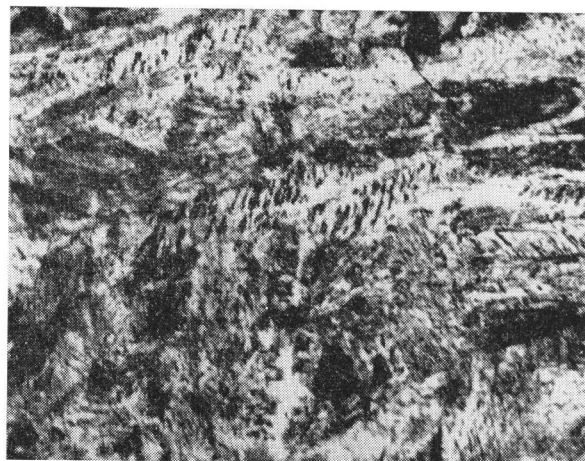
$$\begin{array}{l} (001)_\zeta \parallel (211)_a \\ (100)_\zeta \parallel (0\bar{1}1)_a \\ [010]_\zeta \parallel [1\bar{1}\bar{1}]_a \end{array}$$

This showed that cementite precipitated directly from the bainitic ferrite in lower bainite, and the morphology of the structure indicated that the cementite might be precipitating on twins. They had not, however, been successful in positively identifying such twins, and they agreed with the suggestion of the authors as to why this was the case.

One of the main points they wish to comment upon was that if lower bainite only formed below 350°C, was this criterion obeyed irrespective of carbon content? If so, did this mean that lower bainite could not form in low-carbon steels in which the M_s temperature was above 350°C? The evidence they had obtained led them to believe that lower bainite could in fact be obtained in low-carbon steels, and the existence of lower bainite in these low-carbon steels was also confirmed by the considerably different mechanical property relationships in lower bainite compared with upper bainite. Even in 0.1-0.2% C steels they had observed that lower bainite could form at temperatures just above the M_s temperature, i.e. 450-470°C, with the characteristic orientation relationship between ferrite and cementite. As might be expected the density of carbides was much reduced by the lower carbon content, although they still appeared in the characteristic morphology in the bainitic ferrite plates (Figure C). Due to the very much lower carbon content, the structure was not exactly the same as the high-carbon lower bainite, but it was clear that it had a very similar morphology, and the orientation relationship between the cementite and ferrite indicated that the cementite had precipitated from the ferrite. If this structure was low-carbon lower bainite, and was interpreted as the precipitation of carbide on twins, as Shackleton and Kelly suggested for lower bainite, then the low-carbon lower bainite must be twinned, which hardly seemed likely in view of the fact that low-carbon martensite of the same carbon content was not twinned. Also, the structure could not have originated from the



D 0.4% C, transformed at 350°C × 60000



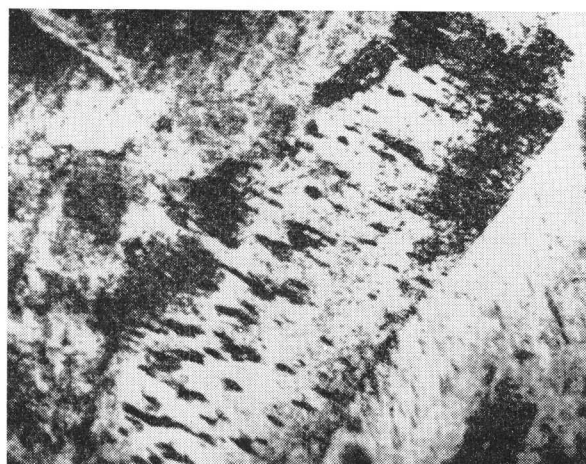
F 0.9% C, transformed at 250°C × 32000

inadvertent formation of martensite followed by tempering, because such martensite would not be twinned and would precipitate cementite in Widmanstätten mode. It was very tentatively suggested that the cementite was precipitated on twins, but that the twins might be developed in the bainitic ferrite due to the transformation stresses. Such twins would tend to be more widely spaced in the lower carbon steels because the transformation stresses were lower than in high-carbon steels. Increasing the carbon content increased the amount of cementite precipitated in the bainitic ferrite until the very typical morphology of the high-carbon lower bainite was observed, Figures D-F, which related to steels containing 0.4, 0.6, and 0.9% C. All the structures in Figures C-F had an orientation relationship confirming that the cementite precipitated directly from the bainitic ferrite, but in the higher carbon steels the transformation temperature had to be decreased to below 350°C to obtain lower bainite.

The structure of upper bainite in low-carbon steels containing 0.1-0.2% C was fairly readily interpreted. It consisted of laths of ferrite with the carbides predominantly occurring between the bainitic ferrite laths (Figure G). Frequently this upper bainite could coexist with lower bainite in the low-carbon steels at temperatures in the range 450-500°C. All their work on the orientation relationship between the cementite and ferrite confirmed the findings of Shackleton and Kelly, in that the cementite and ferrite could only be related through their independent

orientation relationship with the parent austenite. As clearly shown by the authors, this meant that the cementite precipitated from the austenite. As the carbon content of the upper bainite increased, two effects were observed. Firstly, the bainitic ferrite laths did not grow to such wide lateral dimensions, possibly due to the rapid build-up of carbon in the austenite at the growing bainitic ferrite interface. This led to the formation of a pseudo-lamellar structure of cementite between thin ferrite laths. The second feature was that cementite did not always form at the ferrite lath boundaries, but could also occur in parallel striae within a ferrite grain of one orientation, Figures H-J. The many variations in the morphology of upper bainite, particularly of higher carbon content, could usually be attributed to sectioning effects.

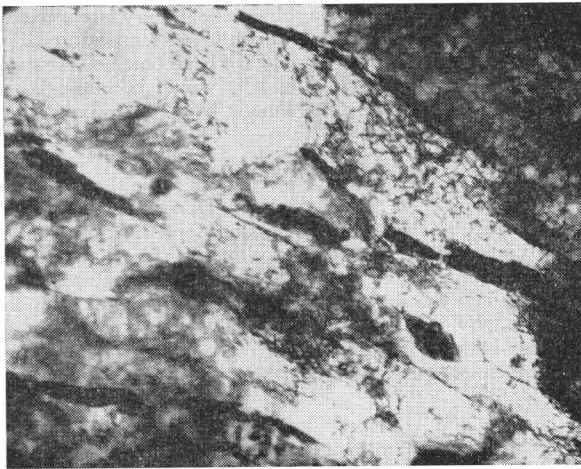
The structures and orientation relationships showed that the cementite precipitated directly from the austenite. In lower carbon upper bainite the cementite nucleated directly from the carbon enriched austenite between the ferrite laths when the extent of local carbon enrichment caused the composition to be hypereutectoid at the particular transformation temperature used. With higher carbon contents, the cementite could nucleate either ahead of the bainitic ferrite, or at or near to the bainitic ferrite/austenite interface where there was more carbon enrichment, and could then be engulfed by the growing ferrite. A feature which required explanation was the lamellar nature of the



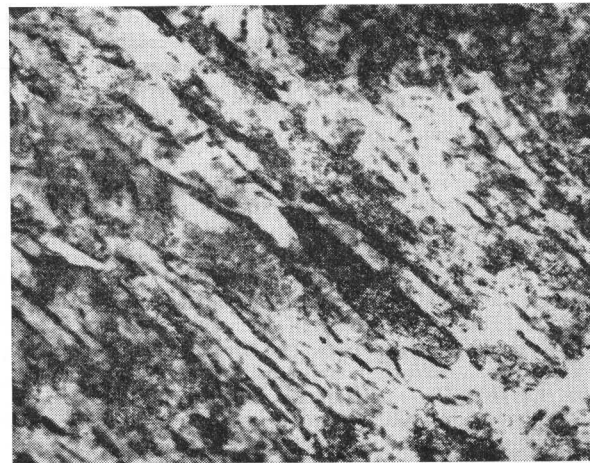
E 0.6% C, transformed at 300°C × 60000



G 0.1% C, upper bainite formed at 450°C × 100000



H 0.4% C, transformed at 550°C × 40000



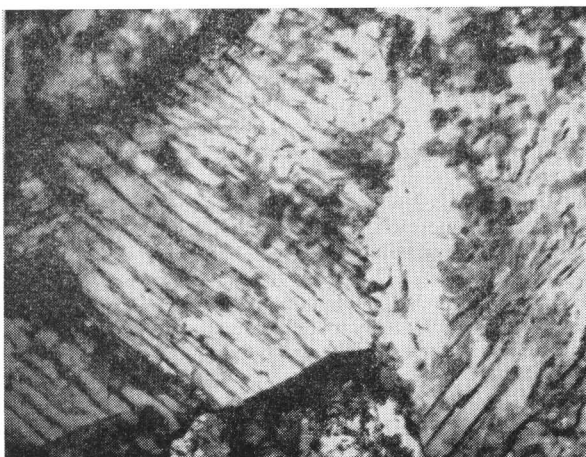
J 0.9% C, transformed at 450°C × 40000

cementite. This could be understood if it nucleated only in the heavily carbon enriched layers at or near to the bainitic ferrite/austenite interface, as it would then tend to form parallel to this interface. If it nucleated well in advance of the ferrite, why did it not form a Widmanstätten structure? In some of the steels, such a Widmanstätten arrangement of cementite could be observed in areas which were quite large and had been enriched in carbon. This was particularly the case in lower carbon steels in which upper bainitic ferrite formed at high temperatures such that carbon could diffuse readily and produce large regions which were enriched to a hypereutectoid composition.

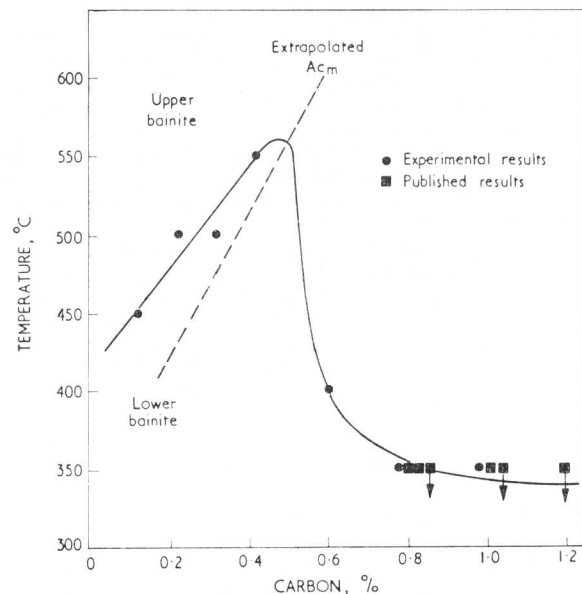
One of the most interesting effects they had observed was that lower bainite could form at about 450°C in low-carbon steels but only below 350°C in steels containing more than about 0.5% C. In fact, the maximum temperature at which lower bainite was replaced by upper bainite appeared to depend very much on the carbon content. The evidence they had obtained tended to indicate that increasing the carbon content from 0.1 to 0.4% increased the upper temperature at which lower bainite was formed. Above about 0.5% C their results agreed with all the published evidence on these high-carbon steels, namely that lower bainite did not form at temperatures above about 350°C. Comparing their own results with those found in the published literature, the variation of the temperature of the change from

upper to lower bainite as a function of carbon content was shown in Figure K.

It was possible to give a tentative explanation of the form of this curve. At low carbon contents, the temperature at which lower bainite replaced upper bainite was that at which the rate of carbon removal from the vicinity of the bainitic ferrite/austenite interface was too slow to allow the growth of the bainitic ferrite, and at the same time the rate of precipitation of cementite in the carbon enriched region was also very slow. Transformation could only proceed at temperatures below this critical temperature by the formation of supersaturated ferrite and the subsequent precipitation of cementite with this ferrite. With increasing carbon content, the carbon concentration at the bainitic ferrite/austenite interface did not alter, being fixed by the extrapolated solubility, but the level of the carbon concentration in the matrix remote from the bainitic ferrite/austenite interface increased with increasing carbon content in the steel, and consequently the concentration gradient down which the carbon



I 0.6% C, transformed at 500°C × 32000



K Effect of carbon content on the temperature of the change from upper to lower bainite

had to diffuse became less steep in the higher carbon steels. Consequently, carbon could not be transported so rapidly away from the interface, and therefore the rate of diffusion had to be increased by increasing the temperature for upper bainite to continue to form. Thus the temperature below which upper bainite could not form and was replaced by lower bainite increased with increasing carbon content.

However, the extrapolation of the A_{c_m} line in the iron-carbon equilibrium diagram showed the carbon content at any given transformation temperature above which cementite would precipitate direct from the austenite. This extrapolated A_{c_m} line was shown in Figure K and it could be seen that it intersected the curve for the change from upper to lower bainite at $\sim 550^\circ\text{C}$ or $\sim 0.5\% \text{C}$. Thus below 550°C all steels containing more than $0.5\% \text{C}$ would precipitate cementite directly from the austenite, and therefore would form upper bainite. All steels containing more than $0.5\% \text{C}$ would form upper bainite at 550°C , and also at all lower temperatures until the rate of precipitation of cementite from austenite became so slow that it was kinetically more favourable for a supersaturated ferrite to form and precipitate cementite within the ferrite. This latter was the criterion for lower bainite, and the temperature at which this occurred was apparently $\sim 350^\circ\text{C}$ and, as might be expected, did not vary very much with carbon content.

This tentative suggestion therefore implied that lower bainite was always nucleated by supersaturated ferrite. Up to $0.4\text{--}0.5\% \text{C}$ upper bainite was nucleated by ferrite, the degree of supersaturation of which depended on the transformation temperature, but above $0.4\text{--}0.5\% \text{C}$ upper bainite was nucleated by cementite.

A critical test of this suggestion would be the observation of primary cementite needles precipitated from the austenite at the start of the upper bainite transformation in high-carbon steels.

Dr D. N. Shackleton (BWRA) referred first to the question of twins in lower bainite. He said that he had been unable to see any twins in the microstructure observed, but this would be expected since twins would disappear when cementite formed on them. One diffraction pattern had been obtained containing twin reflections from a lower bainite region, but no twins were observed in the corresponding area of the micrograph.

With regard to the transition temperature between upper and lower bainite and the formation of lower bainite at temperatures greater than 350°C in low-carbon steels, he considered that the interpretation of the data was extremely difficult. Mr Pickering had observed structures in the low-carbon steels formed above 350°C which had the lower bainite orientation relationship and were similar in morphology to lower bainite, but, as he pointed out, the structures were not exactly the same as that in the higher carbon steels although they were interpreted as such. From his own results, he had experienced the same difficulty in interpreting these structures but from the crystallographic data had labelled them as upper bainite. Full crystallographic data of the cementite-ferrite orientation relationships and cementite habits had only been determined in the $0.4\% \text{C}$ steel in the lower carbon steels. The transformation made at 420°C in this steel had produced the characteristic orientation relationships associated with the upper bainite range. Furthermore the habit plane of the cementite particles had been determined as $\{101\}$ in the cementite lattice and this was again consistent with upper bainite rather than the lower bainite structure. In view of this he considered that even in the lower carbon steels the structures formed above 350°C were upper bainite in nature, although the morphology was difficult to rationalize.

Mr Clark referred to the $0.4\% \text{C}$ steel that Dr Shackleton found to have an upper bainitic structure at 420°C . He wondered how many diffraction patterns Dr Shackleton had examined, since, in the case of his own work, the structure was found to be a mixture of upper and lower bainite at this temperature. The graph shown by Mr Pickering related to the highest temperature

at which any lower bainite was observed, and it was possible to observe areas of upper bainite at much lower temperatures.

Most of the kinetic data available had been obtained for steels containing more than $0.6\% \text{C}$, and he would be interested to know if anyone had done any kinetic work on lower carbon steels, particularly around 450°C .

Professor C. M. Wayman (University of Illinois) said that, in their conclusions, Dr Shackleton and Dr Kelly appeared to propose that the mechanism operating for the formation of lower bainite was identical to that for the formation and tempering of martensite. Was it to be concluded that the habit plane of the bainite and the shape strain would be the same? Was the crystallography of bainite, in their view, identical to that of martensite in the same material? He was not so much concerned about cementite as about where the ferrite was in relation to the austenite.

Dr P. M. Kelly (University of Leeds) said that, with some reservations, the reply to Professor Wayman's question was 'yes', but he asked Professor Wayman to specify which of the four martensite habit planes he was referring to.

Professor Wayman said that he would qualify his query and ask about martensite and bainite in the same material. Perhaps a hypothetical case could be taken of an alloy containing $1\% \text{C}$ in which the crystallography of martensite was in reasonable shape.

Dr Kelly pointed out that the comparison between bainite and martensite in the same material was not necessarily a good one as the region of austenite forming bainite was probably depleted in carbon and so might have a different habit plane from martensite formed in a region with no carbon depletion.

Professor Wayman said that the magnitude of the strains that generated bcc lattice or bct lattice from the fcc lattice was by and large independent of carbon content. The principal strains of the Bain strain really did not depend largely on the carbon content. The point he wanted to make concerned an alloy containing $1\% \text{C}$ and $8\% \text{Cr}$. This was a material on which one could obtain a small amount of lower bainite and make a two-surface analysis of the bainite plates, so it was rather an easy system to work with. However, one had to wait to get the bainite. This work was being done by Mr Srinivasan in his laboratory. The martensite habit plane in this particular material was approximately described as 449 relative to the austenite. This had to be compared with the usual 259 or 3, 10, 15 habit. The bainite in this particular material formed at 310°C with a habit plane quite distinct from martensite. The bainite did not appear to be twinned.

Dr J. W. Christian (University of Oxford) asked if the bainite had taken much carbon out of solution before transformation to martensite.

Dr Kelly said that this was not yet known.

Dr Christian asked whether the habit planes were in material of the same composition, not necessarily the same specimen, but material of the same composition.

Dr Kelly replied that it was nominally the same material. He asked Professor Wayman to be more specific. A habit plane result for martensite or bainite was only significant if the variant of the orientation relationship was specified. Were the two habit planes which Professor Wayman had shown in this stereographic triangle really in the same variant of the orientation relationship? If they were not they could not be compared directly.

Professor Wayman replied that they were, that and the

orientation relationship in each case was close to the standard Kurdjumov–Sachs variant.

Dr L. Kaufman (ManLabs, Inc.) asked how long the transformation took in the 8%Cr–1%C alloy at 320°C to get a lower bainite plate in this way.

Professor Wayman said that the incubation time was approximately five and a half days; at a total reaction time of 17 days one was still retaining about 70% austenite.

The Chairman said he was glad that Mr Pickering had used the term ‘Widmanstätten structures’. He would be glad if Mr Pickering would interpret them! He and others had had discussions as to whether, in fact, all the Widmanstätten structures they had ever seen were bainite!

Mr Pickering stated that whether in fact all Widmanstätten structures were bainite depended upon the definitions given to both these terms. If one took the classical Widmanstätten structure in carbon and lower alloy steels, which consisted of laths of ferrite across a prior austenite grain formed by the transformation of the austenite to ferrite, whether this was bainite depended upon the mechanism of transformation. Recent work in Germany had apparently shown that the formation of Widmanstätten ferrite was a shear reaction, and also it was well known that the formation of Widmanstätten ferrite was accompanied by the diffusion of carbon away from the interface between the ferrite and the austenite. Under these conditions it would seem that Widmanstätten ferrite fulfilled the definition of upper bainite with one major exception, namely that the carbon enriched austenite frequently transformed to pearlite rather than to carbide films between the bainitic ferrite laths. In low-carbon steels therefore Widmanstätten ferrite probably was a form of upper bainite. However, many precipitation structures, such as those found in over-aged alloys, had been called Widmanstätten structures but these could hardly be termed bainite.

Professor J. Nutting (University of Leeds) said that Dr Irvine and Mr Pickering had pointed out that the ductility of bainite steels was inferior to that of the same steel transformed to martensite and then tempered back to the bainitic hardness. On the other hand, Dr Kelly and Dr Shackleton in their paper, and Professor Wayman in his comments, had pointed out the similarity between the bainitic and martensitic transformations. Was it possible from the structural studies on bainite to account for their low ductility when compared with those of tempered martensites?

The Chairman asked to what extent the scale of the lower bainite structure could be altered if, in fact, it was being produced isothermally. Could the needle size or the plate size be altered?

Mr Pickering replied that Professor Nutting might well be correct in his suggestion that the form of the carbides, particularly those carbides which occurred in films at the bainite ferrite grain boundaries, could be detrimental to ductility. These were particularly observed in upper bainite, which in low-carbon steels was not such a good structure from the point of view of room-temperature ductility. Some carbides, apparently at lower bainite grain boundaries, could also be observed, but they were much less pronounced, and generally he had observed that lower bainite was a better crack-resisting structure than upper bainite. The whole problem of the ductility–strength relationships mentioned by Professor Nutting in fact emphasized the general lack of systematic information on the mechanisms controlling ductility in the martensite–bainite structures. There was now a considerable amount of information and understanding of the strengthening mechanisms in steels, but until the effect of each of these strengthening mechanisms on ductility was clearly estab-

lished it would be very difficult to decide which of them, and in what sort of intensity, could be applied to obtain a given strength with the maximum ductility. At present, the one strengthening mechanism which also increased the ductility seemed to be a refinement of the grain size, but many of the other strengthening mechanisms decreased the ductility. It was suggested that as much effort should go into understanding the structural basis of ductility as went into the work on strengthening mechanisms.

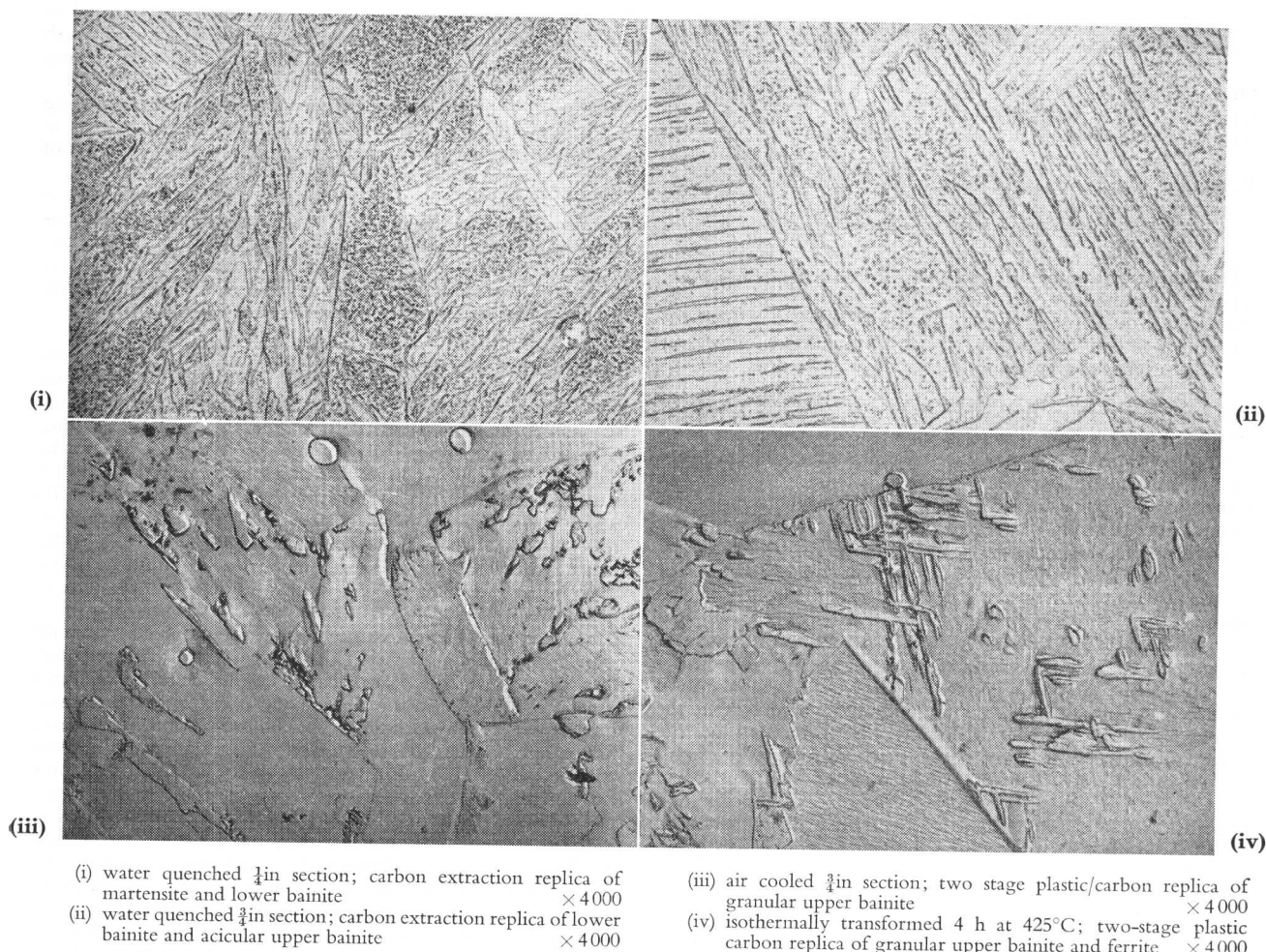
With regard to the point raised by the Chairman, it was possible to decrease the length of lower bainite plate by decreasing considerably the prior austenite grain size, but it was not certain just how effective this would be in decreasing the width. At present it was not clear which of these, the length or width, was the more important in controlling the ductility.

Dr Boniszewski said that he wished to add a few more comments concerning the different resistance to fracture of bainites and martensites, particularly when the fracture was accompanied by small amounts of plastic deformation, e.g. in impact testing or hydrogen embrittlement. Bainites appeared to be inferior to martensite, particularly in low-carbon structural steels of higher strength than mild steel. This difference had practical significance, particularly in welded structures. The American T-1 steel was a notable example. To obtain good resistance to brittle fracture, the weld heat-affected zone of this steel must be quenched with a rate high enough to ensure the transformation to martensite.

It might be thought that the answer to the problem would come from the consideration of the crystallographic relationships between all the three phases, i.e. austenite, ferrite, and cementite, involved in the bainitic and martensitic transformations. However, this need not be so. High resolution, i.e. electron microscopic studies of the bainitic and martensitic microstructures also might not reveal why these two microstructures had different fracture resistance. It was possible to have two steels of the same strength, but one bainitic and another martensitic. The strength was determined by the difficulty with which the dislocations moved through the metal matrix. In both the microstructures the resistance to the dislocation motion might be determined, to a first approximation, by the carbide dispersion. However, the toughness of the two microstructures would be determined, to a first approximation, by their resistance to cleavage cracking. Crack formation was to a great extent a geometrical phenomenon. A dislocation source must operate on a slip system large enough so that a sufficient number of dislocations were piled-up to produce a cavity. Therefore, it was necessary to consider the geometrical interrelation between the individual martensitic and ferritic bainite crystals within the prior austenite grains.

It would be a good thing to return to the good old light microscope, to see how the prior austenite grain was subdivided after the transformation into the various acicular microstructures. Thus, again he had come to the concept of what he called earlier the ‘intragranular texture’ within the prior austenite grain.⁶ In low-carbon alloy steels, bainite tended to form colonies or sheaves of similarly aligned ferrite crystals, while martensite crystals tended to form ‘multidirectionally’, taking a greater advantage of the 24 Kurdjumov–Sachs orientation relationships. Examination of fracture surface replicas in the electron microscope⁶ revealed that cracking could occur through a colony of bainite, as if it was one larger grain. When acicular crystals were aligned side by side and their crystallographic misorientation was low, cracking could occur more easily in such a microstructure than when the acicular crystals formed a multidirectional texture.

Dr K. A. Ridal and **Mr J. McCann** (English Steel Corporation Ltd) said that the terminology used by the authors of the papers in this session on bainite transformations would lead one to conclude that only two forms of bainite existed, and that these could be classified according to the morphology and crystallography of the cementite precipitate. However, it was their experience that



L Electron micrographs of 1%Cr- $\frac{1}{2}$ %Mo steel solution treated at 925°C and cooled at different rates

at least one other form of bainite was commonly found in low-alloy ferritic steels, the features of which were sufficiently distinctive to warrant classification. This structure occurred in both isothermal and anisothermally cooled samples, in the upper part of the bainite range.⁷ It was of special importance in commercial molybdenum- and vanadium-bearing steels, because on tempering a uniform dispersion of alloy carbides could be obtained, which conferred mechanical and creep properties superior to those attainable from the common (acicular) bainites.^{8,9}

To illustrate the structures found in alloy ferritic steels, samples of a 1%Cr- $\frac{1}{2}$ %Mo steel, of similar hardenability to the 1%Cr-Mo-B steels examined by Shackleton and Kelly, were isothermally transformed and cooled at different rates. The steel contained 0.12% C, 0.19% Si, 0.61% Mn, 0.30% Ni, 0.80% Cr, and 0.50% Mo; typical electron micrographs were shown in Figure L.

After water quenching $\frac{1}{4}$ in and $\frac{3}{4}$ in diameter bars from 925°C, the classical lower and upper bainite structures were obtained (Figure L (i) and (ii)). However, an air cooled $\frac{3}{4}$ in diameter bar exhibited a granular bainitic structure, similar to that described by Habraken.⁷ This structure consisted of cementite and a light etching polygonal phase in a ferritic matrix (Figure L (iii)). The ferritic grain size was much larger than in the acicular bainites, which suggested that a massive transformation had occurred. X-ray diffraction proved that the structure contained 10% austenite, and it was deduced that this corresponded to the polygonal regions observed in the micrograph. This was confirmed

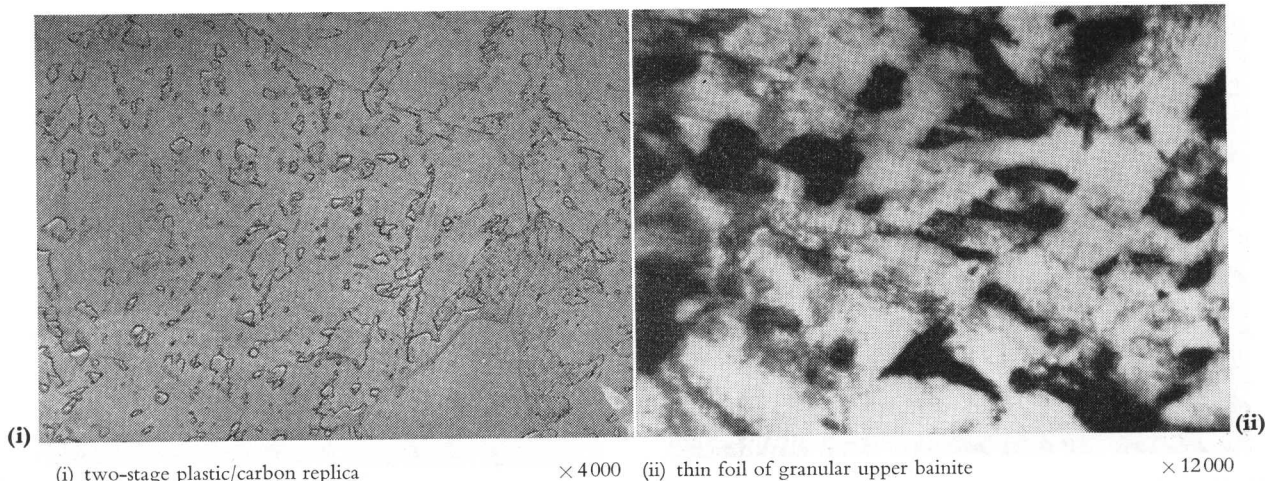
when after 2 h tempering at 550°C, the austenite had completely decomposed to ferrite and cementite.

A sample which was isothermally transformed at 425°C for 4 h consisted of 20% ferrite and 80% granular upper bainite. However, the bainite in this sample consisted of dendritic aggregates of cementite in a ferrite matrix (Figure L (iv)); a structure similar to that obtained on tempering the previous sample. It seemed that the two forms of granular bainite were similar, but that on very slow cooling or isothermal transformation the austenite regions were able to decompose to ferrite and carbide.

A sample of 1%Cr-Mo-V steel was also examined after austenitizing at 975°C and air cooling as a 1 in bar. A granular upper bainite was formed which consisted of small regions of austenite within large ferrite grains (Figure M). It was confirmed that the second phase was austenite, by both selected area electron diffraction on thin foils and by X-ray diffraction analysis, which indicated 15% austenite.

Professor L. Habraken (CNRM, University of Liège) said that he completely agreed with the views expressed by Dr Ridal. Very often, in low-carbon alloyed steels the intermediate transformation did not show a microstructure with ferrite and cementite, but ferrite and carbon-rich austenite. It was what he had called some years ago granular bainite. 10-12, 7, 13.

It was possible, to a first approximation, to connect this aspect of the bainitic transformation with the massive structures described in the Professor Owen's paper. In his opinion, these could



M Electron micrographs of 1%Cr-Mo-V steel air cooled as 1in section

be looked on as an example of the 'short range diffusion' transformation across an incoherent, or better semi-coherent, interface. But, he thought that the transformation appeared in an dehomogenized austenite.

The dehomogenization took place during the stay of the austenite in the metastable zone between the critical points A_3 or A_1 and the beginning of the transformation in the TTT or CCT diagram. The path of the cooling or the isothermal 'path' had a large influence on the degree of the dehomogenization.

During the 'travel' in the metastable austenite, he thought that regions enriched in carbon appeared, in relation to the accumulation of dislocations on $\{111\}$ planes or ordered zones. This effect finally gave, like a check pattern, regions alternatively rich and poor in carbon. The size of this pattern was related to the path that the austenite had followed in the metastable region.

With this hypothesis, he explained that the granular bainite (or the massive microstructure) was directed by ferritic nuclei appearing in low-carbon regions of the austenite; the driving force being the $\gamma \rightarrow \alpha$ transformation. The $\gamma \rightarrow \alpha$ boundaries were semi-coherent with the two lattices and were able to engulf carbon-rich austenitic regions. These regions could give stabilized austenite or could transform in ferrite and cementite during the cooling.

WRITTEN CONTRIBUTIONS

Dr H. Warlimont (Max-Planck-Institut für Metallforschung) wrote that metallographic evidence obtained from numerous steels and experimental Fe-C binary and ternary alloys indicated a pronounced variation of the upper bainite morphology with carbon concentration and the occurrence of cementite precipitation within the upper bainitic ferrites.¹⁴⁻¹⁶ The findings by Shackleton and Kelly that 'the effect of carbon content on the microstructure of the (upper) bainite was slight' and 'in upper bainite the cementite and the ferrite are in fact both formed separately in the austenite' were thus at variance with previous observations on very similar alloys.

This might be due to difficulties in interpreting transmission electron micrographs of upper bainite microstructures arising from their complex contrast effects. Also, since 'the specimens were held at temperature to produce a fully bainitic structure' the observations might not have given a clear indication of the differences in morphology and carbide distribution during the growth stage and might, in particular, have led to misinterpretation of over-aged states of bainite plates formed early during isothermal transformation as representing the immediate transformation product.

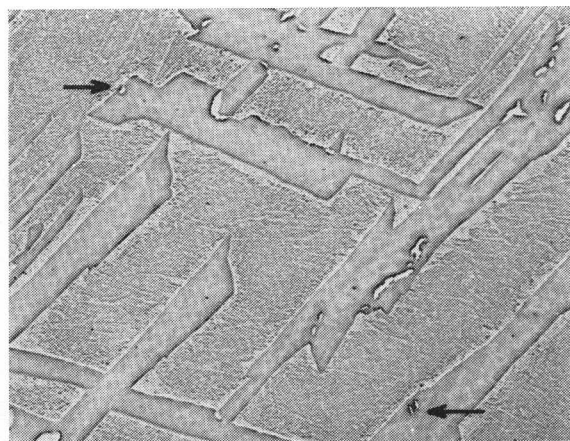
As an example of upper bainite morphologies produced at various carbon concentrations some replica electron micrographs

of partially transformed specimens of Fe-C-5wt-%Ni alloys were presented. The replica technique and partially transformed specimens were found to give more conclusive evidence in this case than the thin-foil technique and fully transformed specimens.

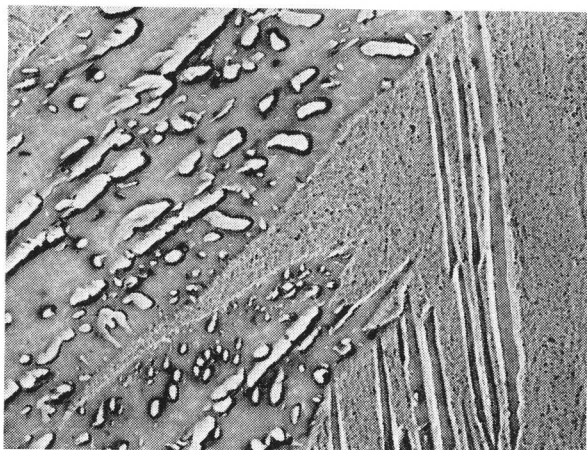
Figure N showed upper bainite laths in an intersecting array. The alloy contained 0.17wt-%C and was transformed at 500°C. The cementite particles pointed out by arrows had clearly not been nucleated from carbon enriched austenite entrapped between two ferrite laths and were, consequently, most probably nucleated within the ferrite phase.

Figures O and P, which showed upper bainite formed at 450°C in alloys containing 0.38 and 0.66 wt-%C, respectively, exhibited that upper bainite formation at these carbon concentrations began by the nucleation of ferrite plates and that there were two distinctly discernible modes of carbide precipitation: nucleation within the ferrite plates at their growth fronts with some common angle of the precipitate long axes to the plate interfaces, and nucleation of cementite in the austenite entrapped between neighbouring bainite plates, these latter cementite particles being extended parallel to the direction of lengthwise growth of the ferrite.

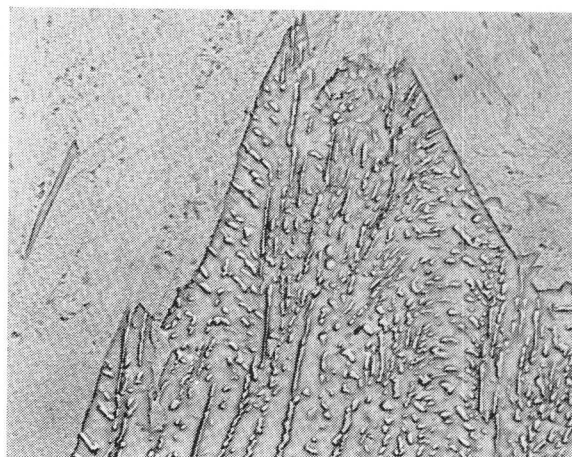
At 1.17 wt-%C and 450°C, as in Figure Q (i) and (ii), the development of an additional bainitic transformation product



N Upper bainite and martensite; the arrows indicate cementite particles formed within the ferrite; Fe-0.17wt-%C-5.1 wt-%Ni, 1 min 500°C $\times 8000$



O Upper bainite and martensite; Fe-0.38 wt-%C-5.0 wt-%Ni, 2 min 450°C $\times 8000$



P Upper bainite and martensite; Fe-0.66 wt-%C-5.0 wt-%Ni, 4 min 450°C $\times 8000$

could be seen: it consisted of one extended cementite plate with a layer of ferrite on either side. This 'inverse bainite'¹⁷ was very probably carbide nucleated. The initial carbide plate was surrounded by a ferrite layer, which would, at a later growth stage, break up into a coarse ferrite-carbide aggregate when carbon diffusion through the ferrite layer became unfavourable. It was interesting to note that a bainitic aggregate containing smaller carbide precipitates was formed concurrently, which was more closely related to ordinary upper bainite.

With a stronger carbide former, in an Fe-1.12 wt-%C-3.55 wt-%-Cr alloy at 450°C as in Figure R*, it was found that the formation of Widmannstätten carbide plates gave rise to a bainitic reaction lagging behind the edgewise growth of the cementite plates. This was an extreme case of bainitic transformation at high supersaturation of austenite with respect to cementite.

On the reasonable assumption that the upper bainite morphology in the series of steels investigated by Shackleton and Kelly was essentially the same as in the Fe-C-Ni alloys of the accompanying Figures N-P (and, possibly, Q (i) and (ii)) the crystallographic results might be reinterpreted such that the 23 cementite-ferrite orientation relationships which were of type

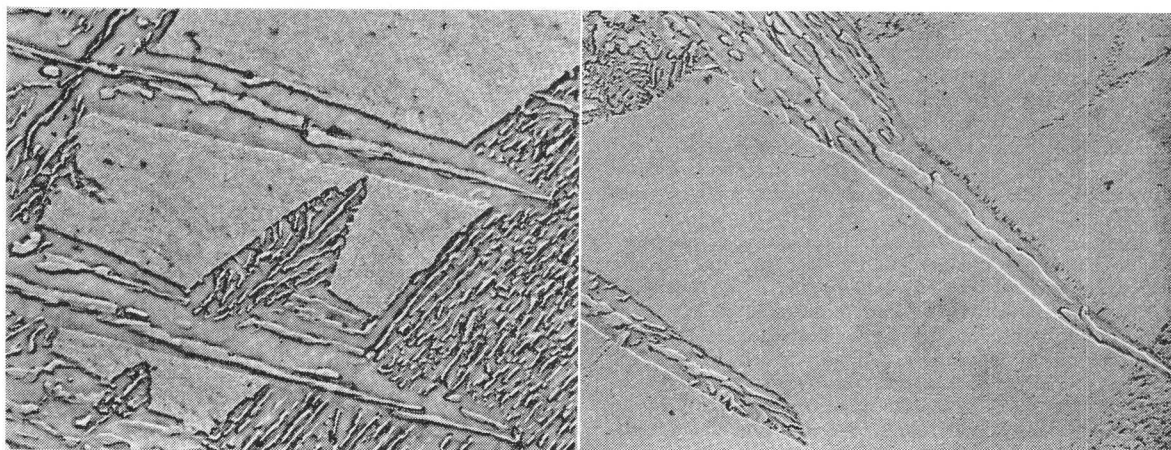
* This micrograph is due to Dr G. R. Speich of the US Steel Corporation, Research Center, Monroeville, Pennsylvania, USA.

(1) had actually resulted from cementite precipitation from ferrite. The remaining 13 alternative relationships only would then remain to be interpreted as resulting from cementite precipitation from austenite.

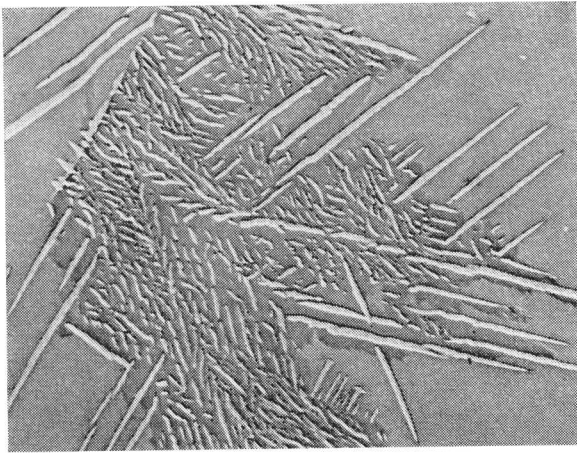
The work quoted in this discussion had been done by the author while employed with the US Steel Corporation, Research Center, Monroeville, Pennsylvania, USA. The experimental assistance by Mr R. C. Glenn and Mr C. E. Groff was gratefully acknowledged.

Dr Shackleton and Dr Kelly replied that Dr Warlimont had raised two interesting points concerning the effect of carbon upon both the microstructure and formation of upper bainite.

Dealing first with the effect on microstructure, they disagreed with Dr Warlimont's reference to the wide variations in the morphologies of upper bainite with carbon content, as reported in the literature. If the micrographs presented in these references were carefully examined it would be found that the main features of the structures were essentially the same at all carbon levels. It might well be that in their paper they had oversimplified this similarity, but in the steels examined in the present investigation the strongest effect on the upper bainite microstructure resulted from change in transformation temperature, not carbon content. Differences in morphology, which could



(i) and **(ii)** Upper bainite, inverse bainite, martensite, and residual austenite; Fe-1.17 wt-%C-4.9 wt-%Ni, 90 s 450°C; (i) $\times 16000$, (ii) $\times 8000$



R *Widmanstätten cementite with bainite formation lagging behind and residual austenite; Fe-1.12 wt-%C-3.3 wt-%Cr, 5 min 450°C*
× 8000

be related to changes in carbon content, did appear and could considerably affect the appearance of the upper bainite (e.g. the amount of carbide present), but these did not alter the basic type of microstructure.

Before turning to the second point raised by Dr Warlimont they would like to comment on the micrographs which he had presented. In Figure *N* he showed ferrite laths formed during isothermal transformation at 500°C in a low-carbon 5%Ni steel. Published transformation diagrams,¹⁸ although available for higher carbon steels only, indicated that proeutectoid ferrite should form first at this temperature. It was thus not inconceivable that Dr Warlimont's Figure *N* was in fact proeutectoid ferrite. If, however, this was a true upper bainite structure, they could see no reason why he maintained that cementite was most probably nucleated within the ferrite. From his micrographs it seemed just as likely that the cementite could have been engulfed by the ferrite after having formed in the austenite. In a similar manner, it could not be concluded from Figures *O* and *P* that the cementite formed within the ferrite rather than in the austenite. Although cementite was not observed ahead of the bainite/austenite interface this was no proof that the carbide did not form in the austenite, since bainitic ferrite would grow into depleted carbon regions of the austenite during the quench.

In the 1.17% carbon steel transformed at 450°C, the 'inverse structure' (Figure *Q*) observed could well be proeutectoid cementite, which, according to the transformation diagrams, preceded any transformation to bainite in high-carbon steels.

In view of these doubts it was suggested that more information would be gained by carrying out the transformations at lower temperatures where these effects did not occur and the structures could be defined with greater ease.

Returning to the second point raised by Dr Warlimont they were unable to agree with the interpretation he made concerning the formation of upper bainite. He discarded much of the crystallographic evidence supporting the idea of cementite precipitation in austenite on the somewhat dubious interpretation of replica electron micrographs. It should be remembered that as well as the determination of the ferrite/cementite orientation relationship made in this investigation the habit plane of the cementite was also determined. This was found to be $(101)_{Fe_3C}$ in the cementite lattice irrespective of the orientation relationship obeyed, and $(211)_\alpha$ in the ferrite lattice when relationship (1) was obeyed.

All the published information on the precipitation of cementite in ferrite or martensite indicated that when cementite formed in untwinned ferrite the habit plane of cementite was $(100)_{Fe_3C}$ which was usually parallel to $(110)_\alpha$. The only time that cement-

ite was observed to precipitate in martensite with a $(211)_\alpha$ habit was when precipitation occurred on twins, and in this case it was likely that the cementite habit plane was $(101)_{Fe_3C}$. As there was no evidence for twins in upper bainite then the shape and habit of the cementite was not a result of precipitation in ferrite. Independent nucleation of ferrite and cementite in austenite so that relationship (1) was obeyed was, however, consistent with this $(101)_{Fe_3C}$ - $(112)_\alpha$ habit. It was on this evidence that it was proposed that both cementite and ferrite formed in the austenite during transformation to upper bainite, irrespective of the orientation relationship developed between the cementite and ferrite.

Mr B. J. Waterhouse (Anderton Springs Ltd) wrote that, during the discussion to the paper by Dr Irvine and Mr Pickering, Dr Boniszewski asked: 'would, in a 0.6% C steel, a martensitic structure have better ductility than a bainitic structure of the same tensile strength?' Dr Irvine's answer to this was 'yes'.

To himself, and he was sure to others concerned in the commercial isothermal heat treatment of plain carbon steels, this statement caused astonishment. They were producing over 10⁷ components a day in 0.8% C steel with bainitic structures and they, and their customers, knew that these were superior in all respects to similar components with martensitic structures.

They had done considerable investigation within their resources to compare the mechanical properties of identical materials in the bainitic and martensitic conditions. In every aspect relevant to ductility and toughness the bainitic structure showed marked superiority – this was particularly so with notched impact testpieces. At 100 tons/in² UTS the bainitic structure had three times the impact value of the martensitic structure. At 115 tons/in² UTS the martensitic structure no longer had a measurable impact value while the bainitic structure still had an impact value twice that of the martensitic at 100 tons/in² UTS. These tests were on commercial steels.

Dr Irvine qualified his previous statement by stating that he was considering ideal conditions, and that in practice the reverse might be true due to formation of micro-cracks in martensitic structures. He would like to know what Dr Irvine would consider ideal conditions, and how these might be achieved particularly in commercial practice.

Dr K. J. Irvine (The United Steel Cos. Ltd) replied that to make a comparison between the properties of bainitic and martensitic structures, it was necessary to be precise about the conditions. In the first place, a distinction must be made between upper bainite and lower bainite. Upper bainite had worse impact resistance than lower bainite, so the comparison sought was between lower bainite and tempered martensite. Their experience was that, providing the hardenability was such as to give effective quenching, then at an equivalent tensile strength level a tempered martensite had superior impact properties to lower bainite. However, as the carbon content increased to the high levels of 0.6–0.8% mentioned, there were greater problems with the martensitic transformation. There was appreciable internal stress associated with the transformation and the possibility of micro-cracking. These features limited the tensile strength which could be obtained at low tempering temperatures and precluded the use of 0.8% C tempered martensitic steels for structural applications. However, because of the greater strength possibilities from martensite, it was possible to get a comparable level of tensile strength to a 0.8% C bainite at a much lower carbon level. Consequently, very good impact resistance could be obtained from a tempered martensite at the tensile level of 100–115 tons/in² quoted. The question should perhaps therefore have been whether it was correct that at a tensile strength level of 115 tons/in² the best impact properties could be obtained from a 0.8% C bainite. His own view was that this was not correct and that a lower carbon tempered martensite would give superior properties. There had been suggestions that the fatigue strength of the bainitic structure might be superior but the question was restricted to impact resistance.

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Ferrous martensites

C. M. Wayman

SYNOPSIS

This paper is concerned with the morphology and fine structure (internal dislocations and twins) of martensite in iron-based alloys. The relationships between the fine structure and theories on the crystallography and strength of martensite are considered. The two-stage growth of martensite plates in certain cases is pointed out, and attention is called to the formation of surface martensite and needle-like martensite in some alloys.

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INTRODUCTION

MARTENSITE has long been a subject of interest to metallurgists. This is scarcely surprising since the hardening of steel depends solely on the diffusionless martensitic transformation of austenite. New techniques have continually been employed to study martensitic transformations. The last few years particularly have witnessed numerous investigations which have applied transmission electron microscopy to the study of both ferrous and non-ferrous martensitic transformations. As is frequently the case, new techniques provide some answers to old questions, but raise as many new ones.

This paper considers some recent observations, many of which are due to transmission microscopy, having to do with the morphology and fine structure (internal dislocations and transformation twins) of ferrous martensites. In particular, the origin and consequences of this fine structure are explored. The discussion is concerned with:

- (i) the origin of the transformation substructure (from the theoretical viewpoint)
- (ii) the effect of the internal transformation twins and substructure on the mechanical properties of Fe-Ni, Fe-Ni-C, and Fe-C martensites
- (iii) some recent observations on the transformation twins and substructure, and the implications of these observations where the crystallography and mechanical properties of ferrous martensites are concerned.

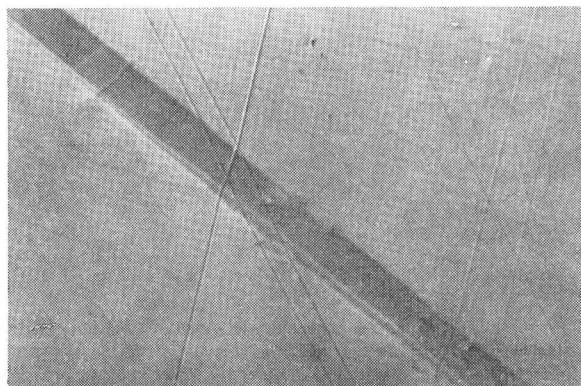
The author is Associate Professor of Metallurgy, Department of Mining, Metallurgy, and Petroleum Engineering and Materials Research Laboratory, University of Illinois, Urbana, Illinois, USA. (MG/Conf/81/65). UDC No. 669.112.227.342

It seems that the crystallography of ferrous martensites is more complex than originally thought, and that the effect of transformation twins on the strength of iron-based martensites is not entirely clear. Some additional observations which suggest that some martensite plates thicken in two distinct stages are presented.

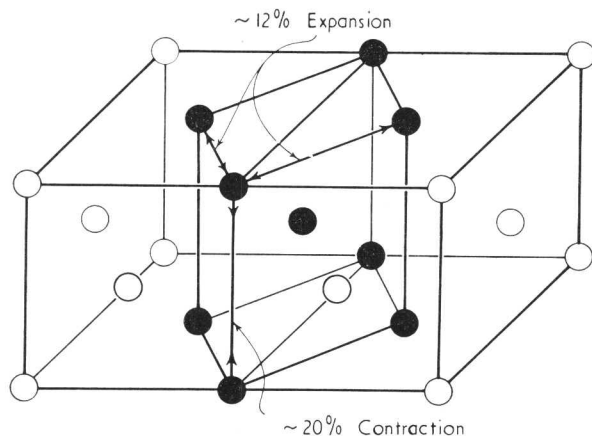
ORIGIN OF THE FINE STRUCTURE IN MARTENSITE

The well-known surface relief or *shape deformation* produced by the formation of a martensite plate infers that the parent-to-martensite transition involves a homogeneous distortion, as in mechanical twinning. From the observed displacement of fiducial reference scratches inscribed in the parent phase before transformation (Figure 1) the components of the shape deformation can be deduced. Straight scratches in the austenite are transformed into straight scratches in the martensite. The martensite plates appear to be tilted about their junction plane with the austenite, i.e. the habit or interface plane, and this plane is thus unrotated as a result of the shape deformation. The continuity of the scratches at the interface further suggests that the habit plane is not significantly distorted, if at all.

It was shown by Greninger and Troiano¹ in 1949 that the



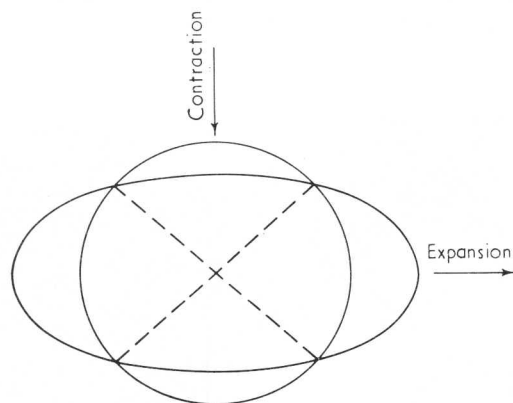
1 Displacement of scratches due to the shape change caused by the formation of a martensite plate in Fe-7.90%Cr-1.11%C



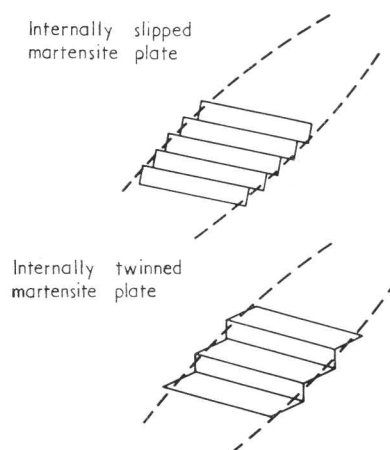
2 The homogeneous $fcc \rightarrow bct$ lattice distortion proposed by Bain in 1924³ involves a contraction of $\sim 20\%$ in the direction which becomes the martensite c axis and a uniform expansion of $\sim 12\%$ in the plane containing the directions which become the martensite a axes

shape change distortion when applied to fcc austenite will not generate the known bct martensite lattice. To rationalize this apparent discrepancy they proposed a phenomenological 'two shear' theory in an attempt to predict the observed crystallographic features of the transformation: the habit plane, parent-product orientation relationship, and shape deformation. However, this theory, the forerunner of present crystallographic theories, was not entirely successful because the shape deformation was assumed to be purely a simple shear on the habit plane. The normal component of the shape strain due to the volume change accompanying the $fcc \rightarrow bct$ transformation was not taken into consideration in their analysis. The shape deformation is now known to be (approximately) an *invariant plane strain*.²

In 1924 Bain³ made a simple but elegant proposal for the $fcc \rightarrow bct$ austenite-martensite lattice transformation, or *Bain deformation*⁴ as it is called (Figure 2). Of the many ways to



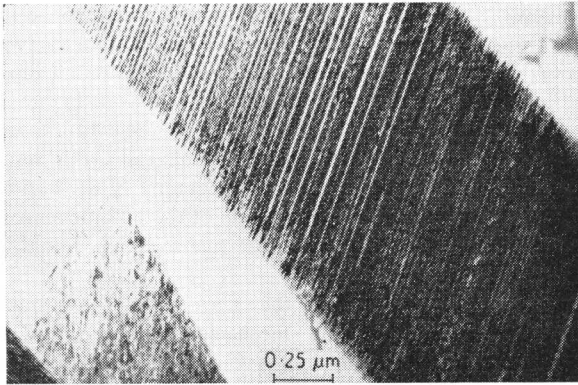
3 A sphere (parent phase), homogeneously distorted into an ellipsoid (martensite) as shown, will meet the ellipsoid along the undistorted planes shown as broken lines only if the principal axis which is perpendicular to the page is unchanged in length (i.e. principal strain in unity); this condition does not generally exist for a homogeneous deformation which distorts one crystal structure into another



4 Schematic representation of martensite plates which are internally slipped and internally twinned^{4b}

generate a bct lattice from an fcc lattice, the Bain distortion involves minimum atomic displacements, as pointed out by Jaswon and Wheeler.⁵ However, the magnitudes of the principal strains of the Bain distortion are inconsistent with the surface relief observations which suggest that the austenite-martensite habit plane is essentially undistorted. A simple geometrical analogue wherein the austenite, a unit sphere, is transformed into martensite, an ellipsoid, shows that for the case of 'partial transformation', if the sphere and ellipsoid are to fit together along an undistorted contact plane, one of the principal strains must be unity and the other two must be greater and less than unity.⁴ This is shown in Figure 3 where the principal strain perpendicular to the page of print has unit value. According to Bain's proposal two of the principal strains are greater than unity (along the expansion axes) and the third is less than unity (along the contraction axis). Hence, the values of the principal strains of the Bain distortion are not such that the martensite-austenite interface can remain undistorted after the Bain deformation occurs.

The difficulties that the most obvious lattice distortion is inconsistent with an observed undistorted habit plane and that the apparently homogeneous shape strain does not generate the martensite lattice from fcc austenite were soon resolved. In 1953 Bowles and Mackenzie^{2,6,7,8} in Australia and Wechsler *et al.*⁹ in the USA proposed independently a 'two shear' theory which successfully predicts the observed habit plane, orientation relationship, and shape deformation for a number of martensitic transformations. This theory is phenomenological and involves the Bain distortion 'occurring' simultaneously with an inhomogeneous shear. The end result is schematically depicted in Figure 4 which shows internally slipped and internally twinned plates of martensite. Because of the theoretical fine scale inhomogeneity the local interface distortions are alternated, and when considered over many interatomic distances these strains average out to zero so that the interface is macroscopically undistorted. With this theory internal slip and twinning are (mathematically) equivalent although these are of course physically different modes of deformation. According to the theory for the case of twinned plates alternate regions undergo



5 Transmission electron micrograph of martensite plates in Fe-29.7%Ni-0.42%C showing $\{112\}_m$ internal transformation twins (courtesy Dr I. Tamura)

the Bain distortion along different crystallographically equivalent contraction axes (the martensite c axis). In the slip analysis the martensite plate becomes a slipped single crystal.

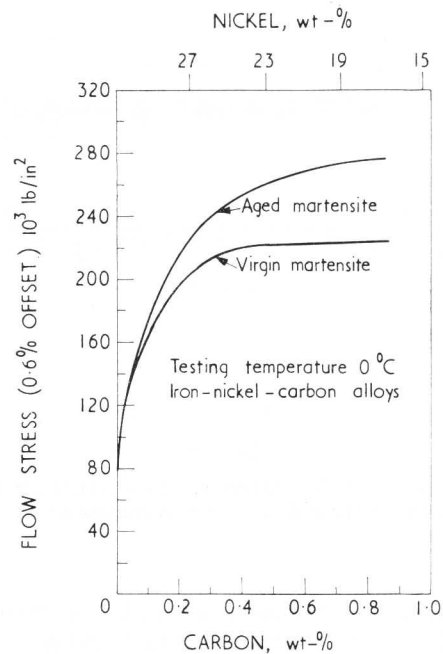
The phenomenological theory thus predicts a fine structure within the martensite. Within the framework of this theory the plane of shear inhomogeneity is a variable. For Fe-C, Fe-Ni, and Fe-Ni-C with a $\{259\}_a$ habit plane the observed crystallographic features are quite well predicted by supposing that slip or twinning occurs on $\{112\}_m$ (subscripts a and m refer to austenite and martensite respectively).

Kelly and Nutting¹⁰ and Nishiyama and Shimizu¹¹ found thin internal $\{112\}$ twins in several Fe-Ni and Fe-Ni-C martensites which exhibit a $\{259\}_a$ habit plane, thus suggesting the essential correctness of the Bowles-Mackenzie and Wechsler-Lieberman-Read theory. Figure 5 is a transmission electron micrograph of a martensite plate in Fe-29.7%Ni-0.43%C.* The parallel striations are $\{112\}_m$ transformation twins, and this photograph is to be compared with the ideal case for twinned plates, as shown in Figure 4; the correspondence is excellent. However, the agreement between theory and observation is not always so good. For low-carbon steels Kelly and Nutting¹⁰ found only a few plates of martensite which were internally twinned (on $\{112\}_m$). Most of the martensite was reported to be in the form of untwinned needles rather than plates. According to their work the relative percentage of internally twinned plates increased with increasing carbon content (decreasing M_s temperature). Needle-like martensite has also been observed in other iron alloys and is discussed later.

MECHANICAL PROPERTIES OF FERROUS MARTENSITES

The unusual strength of hardened steels is well known. The question naturally arises concerning the role of internal twinning or transformation substructure on the mechanical behaviour of martensitic materials such as hardened steels. The thin transformation twins would be expected to exert a strengthening effect since the twin boundaries would be barriers to dislocation movement. According to Kelly and Nutting¹⁰ internally twinned plates should be stronger than un-

*Compositions are given in weight percentages.



6 Variation of flow stress at 0°C of martensitic Fe-Ni-C alloys ($M_s \sim -35^\circ\text{C}$) as a function of carbon content (Winchell and Cohen)¹²

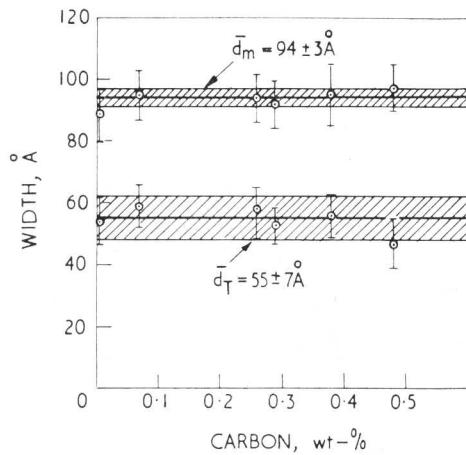
twinning plates because the number of possible slip systems common to both twin orientations in a twinned plate is only a quarter of that of an untwinned plate.

For carbon steels, since the volume fraction of internally twinned plates (remainder needles) increased in the range 0.3–0.8% C (paralleling the well-known increase in hardness with carbon content in the same range), a strong correlation between hardness increase (with carbon content) and the relative proportion of internally twinned plates seemed likely.¹⁰ Kelly and Nutting concluded that the hardness of quenched steel is due to three main factors:

- (i) solid solution strengthening by carbon
- (ii) work hardening due to the transformation itself
- (iii) hardening due to numerous twin interfaces.

A systematic study of the strength of martensite in Fe-Ni-C alloys (0.02–0.96% C; 15.1–30.8% Ni) was made by Winchell and Cohen¹² who adjusted the nickel and carbon contents of their alloys so that the M_s temperature in all cases was $\sim -35^\circ\text{C}$. Both tensile and compressive tests were made to determine the flow stress of the martensitic alloys, and the results of their work are shown in Figure 6. As can be seen the flow stress increases with carbon content from an extrapolated value of 42 000 lb/in² for carbonless Fe-Ni martensite. When their freshly quenched specimens were aged above -60°C tempering occurred, which imparted an additional strengthening effect, i.e. martensite aged for 3 h at 0°C was stronger than virgin martensite (Figure 6). It was concluded that carbon strengthening was not a diffusion effect (i.e. segregation), that the strength of martensite is mainly due to solution strengthening by carbon, and that the transformation twins in the martensite plates contribute at most only a base strength.

A transmission microscopy study of the fine structure of



7 Transformation twin thickness *v.* carbon content for Fe-Ni-C alloys. The widths of the two twin orientations are shown (Richman)¹³

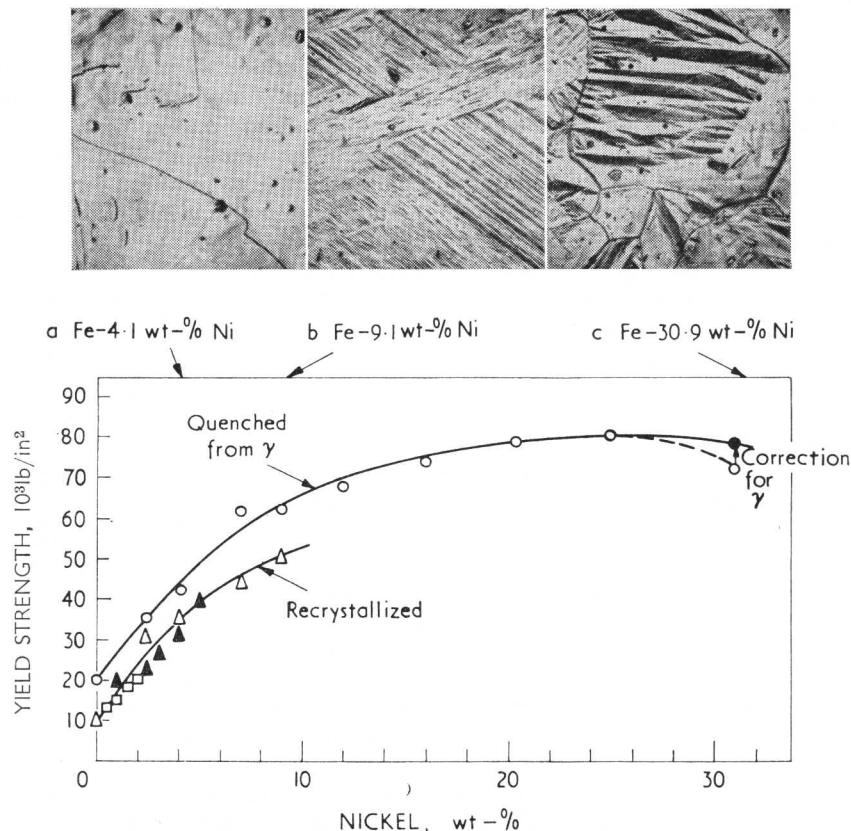
Fe-Ni and Fe-Ni-C martensites was made by Richman¹³ who was motivated by the suggestion of Kelly and Nutting¹⁰ that the transformation twins make a significant contribution to the strength of martensite. He contended that if the martensite fine structure was largely responsible for the observed variation of flow stress with carbon content it would be necessary for the twin spacing to decrease with increasing carbon content of the

martensite. The observed twin spacing was found to be independent of carbon content and M_s temperature, and the twin thickness for the two martensite twin orientations for all alloys studied was roughly 60\AA and 100\AA , as shown in Figure 7. Since the habit plane of the martensite in these Fe-Ni and Fe-Ni-C alloys is evidently $\{259\}_a$ in all cases, this result is predicted. That is, since the crystallography of the various alloys is the same, so is the magnitude of the inhomogeneous shear, and hence no variation in the fine structure would be expected.

For carbonless Fe-Ni alloys the effects of the $\gamma \rightarrow \alpha$ transformation substructure on mechanical properties were evaluated by Speich and Swann,¹⁴ who attempted to separate the solid solution strengthening effect of nickel in the bcc lattice from that of the transformation substructure. By recrystallizing low-nickel alloys ($<9\%$ Ni) in the single-phase α -region, they were able to determine the solution strengthening contribution of nickel in well annealed bcc Fe-Ni alloys. The transformed Fe-Ni alloys were quenched from 1000°C into iced brine and tested in tension.

The findings of Speich and Swann (Figure 8) are not obviously straightforward to interpret because the transformation morphology varies with nickel content.* For alloys containing 0-4% Ni the transformation product exhibited no

*The variation in transformation morphology with nickel content as reported by Speich and Swann is not in complete agreement with other work, as is pointed out later.



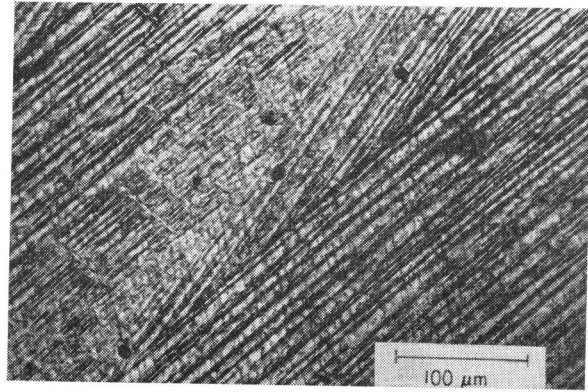
8 Yield strength of quenched and annealed Fe-Ni alloys as a function of nickel content; the inserts show the variation in surface relief with composition (Speich and Swann)¹⁴

surface relief effects and consisted of grains with irregular boundaries and random dislocations (transmission microscopy). For alloys containing 6–25%Ni polished and etched specimens showed that the transformation resulted in a blocky structure, but surface relief effects characteristic of a bundle of plates placed side by side were seen. Transmission micrographs of these specimens revealed ‘cells’ (blocks) containing many irregular dislocations. For alloys with more than 25%Ni lenticular, internally twinned martensite plates were reported. Figure 8 also shows the surface relief for these different cases, and the solution-strengthening contribution of nickel in the recrystallized bcc Fe–Ni alloys. As can be seen a rather smooth variation of yield strength (0.2% offset) with nickel content was found despite the variation in product morphology. It was suggested¹⁴ that the results for transformed Fe–Ni alloys led to the conclusion of Winchell and Cohen¹² that the transformation twins (and transformation substructure) are of secondary importance and that the prime hardening factor in transformed Fe–Ni alloys is solid solution strengthening by nickel.

MORPHOLOGY AND SUBSTRUCTURE OF FERROUS MARTENSITES

It appears that some ferrous martensites exhibit a plate-to-needle transition as the solute content (either interstitial or substitutional) is decreased. The morphological difference between low- and high-carbon martensites in carbon steels has been reported in numerous instances. Bowles¹⁵ suggested that the martensite transformation in low-carbon steels is characterized by the degeneration of $\{225\}_a$ plates into needles; as already mentioned the transmission microscopy of Kelly and Nutting¹⁰ led to the conclusion that the frequency of needles increased with decreasing carbon content. A somewhat parallel situation seems to exist for Fe–Ni alloys. The variation in transformation morphology with nickel content in Fe–Ni alloys has been well documented by Honma^{16,17,18,19} whose extensive work is apparently not yet widely recognized. Honma extended the work of Förster and Scheil²⁰ who reported two different martensite morphologies in Fe–Ni alloys. The martensite which formed rapidly with an audible click and which resembled mechanical twinning was termed ‘umklapp’ while a different martensite not accompanied by audible noises and resembling slip deformation (on the surface) was termed ‘schiebung’. The optical microstructures of these were different.²⁰

Honma established a rather sharp morphological boundary at ~ 30 wt-%Ni, above which the familiar plate-like (umklapp) martensite was observed in surface relief, and below which the relief effects were different and composition-dependent as well. The composition-dependent low-nickel relief markings fall into two categories: a series of side-by-side parallel markings (20–29.5%Ni, shown in Figure 9), and a Widmanstätten-like relief (25–29.5%Ni, shown in Figure 10). Both of these relief structures were termed *schiebung* by Honma. However, upon etching it was found that neither of these relief structures corresponded well to the underlying transformation structure revealed by polishing and etching away 0.01–0.1 mm. On the other hand, a good 1 : 1 correspondence between the umklapp, plate-like martensite and the structure obtained by removing the surface layer was found. In further contrast, the umklapp

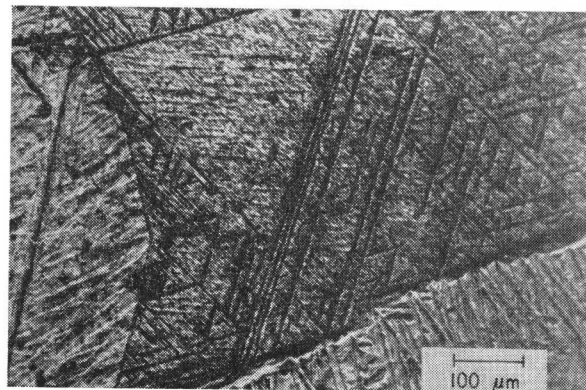


9 Typical ‘side-by-side’ surface relief in Fe–25%Ni (courtesy Professor T. Honma)

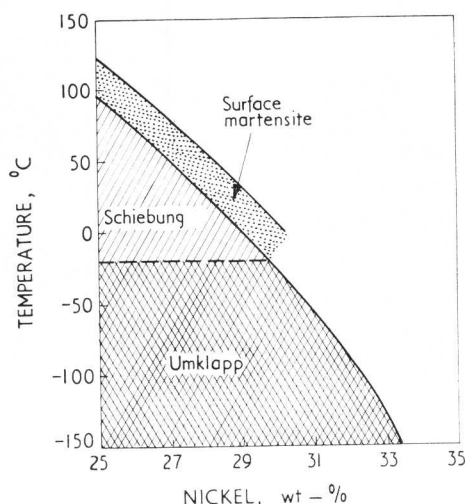
transformation was an instantaneous burst effect while the parallel *schiebung* relief markings (Figure 9) formed sequentially, side by side.

In alloys containing 20–29.5%Ni Honma found that, after the surface relief structure was removed by electropolishing and etching, the inner structure was revealed to be brick-like or blocky in nature, with the long dimension not parallel to the surface relief markings; in some cases the difference was as much as 30° . By trace analysis, the long dimension of the blocks was found to be near $\{111\}_a$. However, trace analysis of the parallel side-by-side relief marking did not reveal any crystallographic consistency. The Widmanstätten-like surface relief (Figure 10) was also consistent with a $\{111\}_a$ habit, but the needle-like nature of this transformation product makes any habit *plane* concept rather meaningless and in addition obscures a direct connexion with the blocky structure, although trace analyses of both indicate a $\{111\}_a$ habit.

In alloys containing 20–30%Ni substantial surface relief was observed in specimens which were cooled to 20–30 deg C above the usual M_s temperature as determined by dilatometer measurements. For Fe–29.5%Ni the extent of surface relief markings suggested that $\sim 20\%$ transformation had occurred when room temperature had been reached. But length change measurements revealed that only $\sim 1\%$ transformation had actually



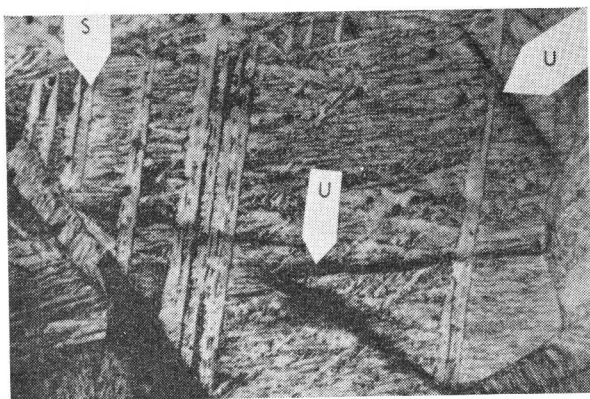
10 Widmanstätten type surface relief observed in Fe–27.5%Ni (courtesy Professor T. Honma)



11 Variation in transformation morphology of Fe-Ni alloys with nickel content, according to Honma¹⁸

occurred. Metallographic examination showed that transformation occurred only near the surface in these alloys with an average depth of $30\mu\text{m}$ and with an M_s temperature $20\text{--}30\text{degC}$ higher than that (the abrupt expansion) revealed by dilatometer measurements. When this surface martensite was electropolished away at room temperature new surface martensite would subsequently form.

Honma's results are summarized in Figure 11, which shows transformation morphology as a function of nickel content. Apparently four different transformation morphologies may be found in Fe-Ni alloys containing more than 20%Ni. The presence of surface martensite in alloys containing between 20 and 30%Ni, which can form above the usual M_s temperature, makes it difficult to determine the nature of the 'interior' transformation product in this composition range, especially since no particular correspondence between the surface markings and the interior etched structure exists. Indeed, the preferential nucleation and pronounced transformation at the surface in



12 Displacement of surface schiebung martensite (S) by plate-like umklapp martensite (U) which formed at a lower temperature in an Fe-29.0%Ni alloy; after the formation of schiebung martensite, the specimen was etched at room temperature and then cooled (courtesy Professor T. Honma) $\times 160$

these alloys does not permit one to assert at all that the interior transformation structure exhibits any relief effects or shape change. For Fe-Ni alloys containing $\sim 20\%$ Ni or less and with an M_f temperature above room temperature the situation is obviously complicated because complete transformation occurs before room temperature is reached. Honma^{18,19} also observed that the parallel side-by-side relief markings in Fe-29.5%Ni (not the Widmanstätten-type relief, which can also form) were the same as those observed in Fe-20%Ni, and can be preferentially nucleated at grain boundaries and twin boundaries.

It is of interest to point out that alloys containing $\sim 28\%$ Ni, when cooled to sufficiently low temperatures, may consist of four different transformation morphologies: surface martensite in the form of Widmanstätten-like needles, and/or numerous side-by-side parallel needles; an interior transformation product, the crystallography of which is not clear; and the well-known umklapp or plate-like martensite which forms in bursts at lower temperatures. An interesting case is shown in Figure 12 in which the initially formed surface martensite is displaced by the shape deformation of the underlying plates of martensite which formed at a lower temperature. In cases such as this it is possible to distinguish between the umklapp and schiebung martensite by means of an acoustical device. The umklapp martensite produces audible clicks and exhibits the familiar midrib; the schiebung martensite (surface and interior) exhibits neither.¹⁹

The side-by-side surface martensite needles observed by Honma for Fe-Ni alloys containing 20-30%Ni appear similar to the isothermal martensite in low-carbon Fe-Ni alloys observed by Yeo²¹ and to the surface martensite in Fe-30.2%Ni-0.04%C described by Klostermann and Burgers.²² Klostermann and Burgers observed surface martensite on various flat surfaces of single crystal specimens; this martensite formed spontaneously after electropolishing at $5\text{--}30\text{degC}$ above the plate-like martensite M_s temperature. By controlled heating of their specimens with the electropolishing current the surface martensite, which extended to a depth of $5\text{--}30\mu\text{m}$, could be removed without the formation of additional martensite. The length of these surface martensite needles was often 1 000 times their breadth and thickness, and trace analysis showed their axial orientation to be random, although the needles themselves were confined to $\{112\}_a$ (or perhaps $\{225\}_a$) planes.

The relationship between the inner block structure observed by Honma and the 'dislocation cell structure' reported by Speich and Swann is not known. The width of their cells, $\sim 1\mu\text{m}$, corresponds well with the surface relief (see Figure 8, insert for Fe-9.1%Ni). Whatever the case, the presence of surface martensite in the range 20-30%Ni (and perhaps lower) greatly complicates interpretations, especially when thin sheet specimens are thinned down to electron transparency. In fact, specimens $\sim 100\mu\text{m}$ thick may consist entirely of surface martensite.

The needle-like martensite which can form above the usual M_s temperature has been explained on the basis of the absence of constraints which oppose the transformation volume change.^{18,22} Although the surface needles orient in no particular direction, the fact that they are confined to $\{112\}_a$ planes and exhibit a well defined shape change suggests that their

nature is controlled by some crystallographic boundary conditions.

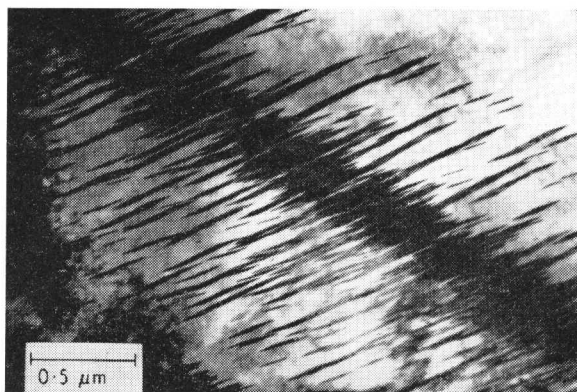
The previous commentary suggests the importance of examining in more detail the 'inner martensite' in Fe-Ni alloys. The well known distinction between the low- and high-carbon martensites in steels may be related to the observed morphological differences in Fe-Ni alloys. It is also desirable to reactivate concern over the morphology of martensite in low-carbon steels, especially since the $\{111\}$ needle-like martensite^{10,15} may be only a surface effect. The presence of surface martensite and the blocky interior structure in Fe-Ni alloys seem relevant to the thermodynamic analyses²³ of martensitic transformations which suppose that martensite is a plate-like product, and on such a basis estimates are made of surface and strain energies. Bibby and Parr²⁴ have pointed out that the present thermodynamic approach to martensitic transformations is not consistent with their recently measured M_s value of $\sim 750^\circ\text{C}$ for high-purity iron.

Experiments concerned with quenching rate v. transformation mechanism have established that very dilute iron alloys will transform martensitically if quenched rapidly enough. The most recent of such investigations are due to Bibby and Parr²⁴ and Swanson and Parr²⁵ who studied pure iron and Fe-Ni alloys respectively. The criterion of the shape change was used to assess whether or not the $\gamma \rightarrow \alpha$ transformation occurred martensitically. Fe-0.0017% C would transform martensitically ($M_s \sim 750^\circ\text{C}$) at cooling rates in excess of 35 000 degC/s. This is to be contrasted with an M_s temperature of $\sim 520^\circ\text{C}$ obtained by extrapolating available M_s v. %C data to 0% C. To obtain such quenching rates very small specimens must be used. Surface relief in quenched zone-refined iron was observed by Wayman and Altstetter.²⁶ The relief markings were similar to those obtained by Bibby and Parr²⁴ for pure iron, by Swanson and Parr²⁵ for Fe-7%Ni (30 000 degC/s), and by Honma^{16,17,18} for Fe-20-29.5%Ni (parallel surface needles). Attempts to relate the surface relief in quenched zone-refined iron²⁶ to the underlying etched structure were not successful. It may be that the surface relief in rapidly quenched iron and dilute Fe-Ni alloys is due to surface martensite, which, as pointed out previously, can form at temperatures significantly above the usually accepted M_s temperature. This may be one reason for the unusually high M_s temperature for pure iron.

Further work on martensite in Fe-Ni and Fe-Ni-C alloys^{27,28} shows that the situation for plate-like martensite is not as simple as originally assumed. The recent investigations dealing with the strength of martensite^{12,13,14} have implicitly assumed that martensite plates are completely twinned as in the ideal case (Figures 4 and 5). This is not in general true. For Fe-Ni and Fe-Ni-C alloys in which plate-like martensite is formed the extent of internal twinning depends on the M_s temperature.^{27,28} That is, only those plates which form at quite low temperatures are completely twinned.

Variation in twin density in Fe-Ni and Fe-Ni-C martensites

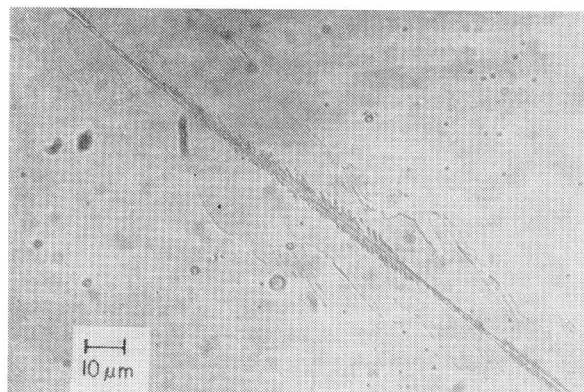
For Fe-Ni alloys containing near 30%Ni ($M_s \sim -20^\circ\text{C}$) transmission micrographs show a high twin density near the midrib region, but this density drops off sharply as the austenite-martensite interface is approached (Figure 13). This effect is more clearly seen in the optical microscope after the specimens



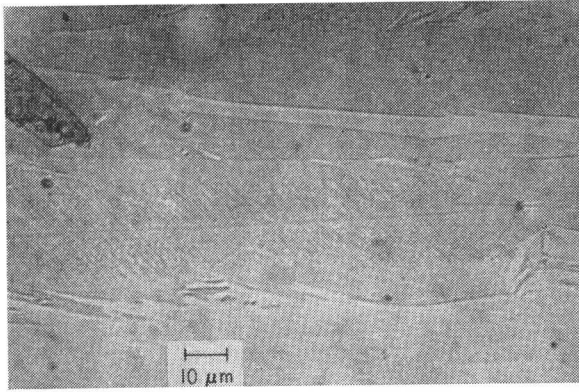
13 Transmission electron micrograph of martensite plate in Fe-29.8%Ni alloy showing high density of twins at midrib region and absence of twins in other regions of the plate²⁷

are etched with certain etchants (Figure 14). There is a 1 : 1 correspondence between the transmission micrographs and the optical micrographs. The untwinned volume of the plates consists of a regular pattern of dislocations (not in contrast in Figure 13), as was first pointed out by Warlimont.³⁰ These will be discussed later. As the nickel content is increased by about 2% (and the M_s temperature lowered to $\sim -90^\circ\text{C}$) the twins are no longer confined to the immediate vicinity of the midrib region (Figure 15) but extend over a considerable distance. It is also to be noted that the twins terminate rather abruptly and define an interface which is more plane-like than the austenite-martensite interface itself. As the nickel content is increased to 33.2% ($M_s \sim -150^\circ\text{C}$) the twins extend all the way to the austenite-martensite boundary (Figure 16) and these plates exhibit decidedly more regular interfaces. It has also been noted (for the lower nickel alloys) that in cases where the internal twinning is not complete in the initially formed plates (larger ones), the smaller plates which evidently formed at lower temperatures were completely twinned (and exhibited more regular interfaces).

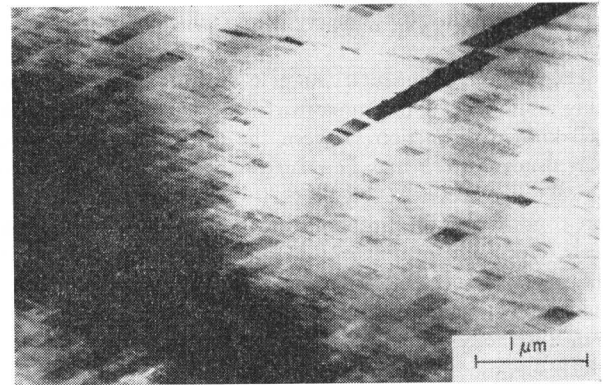
A similar behaviour pattern is found for Fe-Ni-C alloys. For example Figure 17 shows a partially twinned plate in Fe-



14 Optical micrograph corresponding to Figure 13 showing localized twinning at the midrib²⁷



15 Optical micrograph of martensite plate in Fe-32.0%Ni showing extent of internal twinning and regular plane-like interface defined by terminus of twins²⁷



17 Transmission electron micrograph of martensite plate in Fe-23.8%Ni-0.57%C alloy showing incomplete internal twinning and high twin density at centre of plate (Wells²⁹)

23.8%Ni-0.57%C ($M_s \sim -80^\circ\text{C}$), while Figures 18 and 19 are optical and transmission electron micrographs of martensite in Fe-21.7%Ni-1.0%C with a much lower M_s temperature. The optical micrographs (Figure 18) reveal no midribs, but the transmission micrographs (Figure 19) do show relatively diffuse midribs. The straight nature of the completely twinned Fe-Ni-C plates is evident.

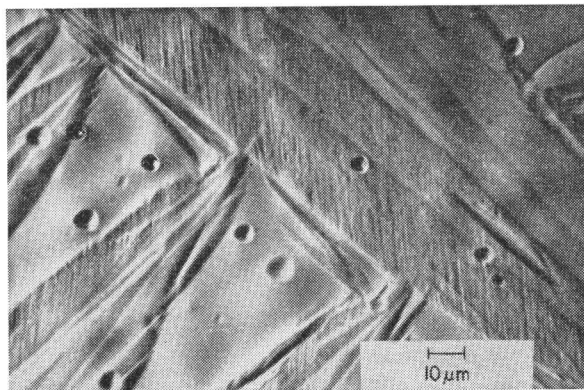
An attempt was made to compare the previous results for Fe-Ni and Fe-Ni-C martensites with those of Kelly and Nutting¹⁰ for plain carbon steels, particularly Fe-1.0%C (which incidentally has a $\{225\}_a$ habit plane and not $\{259\}_a$ as with the Fe-Ni and Fe-Ni-C martensites). However, it is not possible to conclude from their published photographs whether these plates, which formed at well above room temperature, are completely twinned. However, an observation at variance with Richman's findings¹³ for Fe-Ni and Fe-Ni-C martensites was reported by Kelly and Nutting¹⁰: the twinned to untwinned ratio varied considerably for Fe-1.0%C martensite. On the other hand, Nishiyama and Shimizu³¹ found that martensite plates in Fe-0.7%C were not completely twinned. Some preliminary work in the author's laboratory on martensite in Fe-7.90%Cr-1.11%C (with a $\{225\}_a$ habit plane and $M_s \sim -34^\circ\text{C}$) suggests that these plates may not be completely

internally twinned and exhibit a higher twin density near the midrib region. It seems that more detailed studies of the extent of internal twinning in ferrous martensites are required.

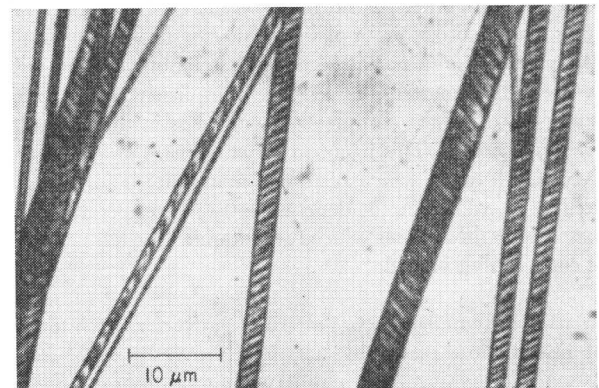
Figure 20 is a transmission micrograph showing typically the appearance of dislocations in the untwinned portion of a martensite plate (Fe-32.0%Ni). The well-defined nature of the dislocations is apparent, and there is no evidence of transformation twins in the dislocated regions.

For Fe-Ni and Fe-Ni-C martensites, where only the plates which form at low temperature are completely twinned, several questions arise:

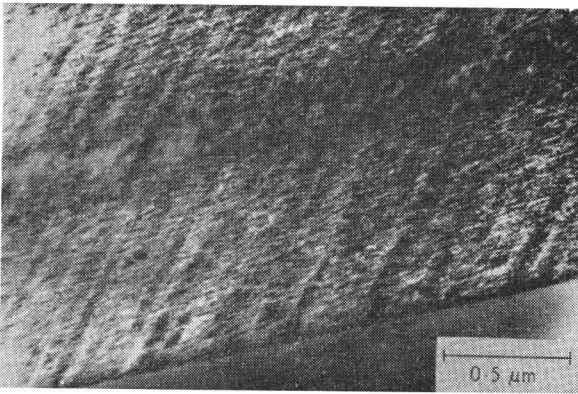
- (i) in the work concerned with the contribution of the transformation twins to the strength of martensite, to what extent were the plates twinned?
- (ii) since there appears to be a definite crystallographic character to the dislocations in the partially twinned plates, is it implied that the plates thicken in two stages, the first in which the inhomogeneous shear is twinning and the second in which slip is involved?
- (iii) why are the plates which form at lower temperatures completely twinned whereas those which form at a higher temperature are not? This point seems all the more curious since some non-ferrous martensites with



16 Optical micrograph (oblique illumination) of martensite plates in Fe-33.2%Ni; the straight nature of the plates and complete internal twinning may be noted²⁸



18 Optical micrograph of etched martensite in Fe-21.7%Ni-1.0%C showing twins extending completely across plates and apparent absence of midrib region²⁷

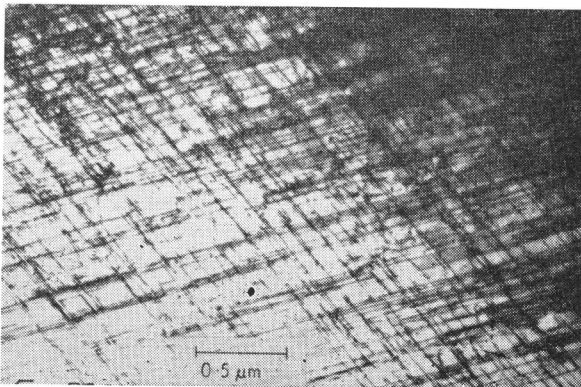


19 Transmission electron micrograph corresponding to Figure 18 showing high twin density complete to interface²⁷

high M_s temperatures, which have been studied in detail^{32,33,34} reveal that the inhomogeneity (stacking faults) extends completely throughout the volume of the plates.

Concerning the effect of twins on the mechanical properties of martensite, it is interesting to note that the Fe–Ni–C martensites studied by Winchell and Cohen¹² had M_s temperatures near -35°C , indicating that these plates were not completely internally twinned. Similarly the Fe–Ni alloys studied by Speich and Swann¹⁴ extended up to Fe–30.9%Ni ($M_s \sim -35^\circ\text{C}$), at which composition the plates are only partially twinned. It would be instructive to have more mechanical property information for ferrous martensites with low M_s temperatures and which exhibit complete internal twinning. To obtain such information, however, is not easy because the low M_s temperature alloys also contain substantial amounts of retained austenite. A correction must be made for the retained austenite, as done by Speich and Swann¹⁴ who found 9% retained austenite in Fe–30.9%Ni cooled to -196°C .

No attempt is made here to suggest that the transformation twins in martensite plates result in an extraordinary contribution to the strength of martensite, but it should be pointed out that in the cases examined the martensite plates appear to have contained a substantial untwinned volume. The observation of Speich and Swann¹⁴ that no sharp increase in strength occurs as the (Fe–Ni) martensite morphology changes from untwinned



20 Dislocations in untwinned region of martensite plate in Fe–32.0%Ni; plane of foil $(112)_m$

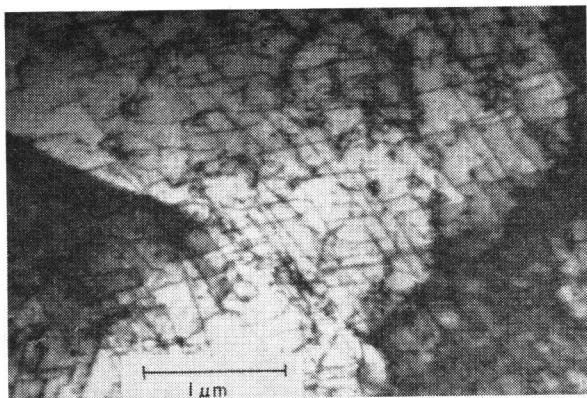
(cellular) to twinned (plates) is readily understood since the percentage of twinning in their martensite plates was low. Since the untwinned fraction of the plates decreases with increasing nickel content, a gradual rather than abrupt increase in yield strength with nickel content would be expected, as was observed.

Dislocations in martensite

The duplex fine structure in Fe–Ni martensite plates, consisting of twins near the central region (midrib) of the plates and dislocations near the interface, led Warlimont³⁰ to suggest a two-stage process for the formation of a martensite plate. The internal twins were identified as $\{112\}_m$ twins and according to him the dislocations were distributed along $\langle 111 \rangle_m$ directions in $\{110\}_m$ planes. It was suggested that initially a thin plate of martensite forms in which the inhomogeneous shear is twinning, to be followed by thickening according to a process in which the inhomogeneous shear involves slip on $\{110\}_m$. This proposal was criticized³⁴ because, if the inhomogeneous shear is $\{110\}\langle 111 \rangle$ slip, a $\{259\}_a$ habit plane would not be predicted. Ideally, if the inhomogeneous shear is slip, 'residual' dislocations should not be observed in the interior of the plates because the slip dislocations must move to the interface to accomplish their necessary act of producing a macroscopically undistorted interface. In spite of this the absence of transformation twins near the interface implies that either detwinning has occurred or that the plate has thickened by a non-twinning mechanism. Hence Warlimont's proposal is an attractive one.

In the light of additional analysis²⁸ of the dislocations in the untwinned regions of plates, the observed dislocations probably do not lie in $\{110\}_m$ planes as provisionally concluded by Warlimont.³⁰ Referring to Figure 20 again, the normal to the foil (photograph) in this case is $[110]_m$. The dislocations form a diamond-like pattern consisting of two prominent directions about 70° apart. Because of the long length of the dislocation lines it is clear that the dislocations themselves lie in the plane of the foil, but it is not clear that they glide in the plane of the foil, i.e. $(110)_m$. Trace analysis²⁸ shows that the dislocations define $\langle 111 \rangle_m$ directions. Since the martensite (Fe–32.0%Ni) is bcc the dislocations evidently have a $\langle 111 \rangle$ Burgers vector.

The diffraction pattern²⁸ corresponding to Figure 20 is essentially two-beam with the observed dislocation contrast caused by a 110 reflection. Although there are four possible $\langle 111 \rangle$ directions, for 110 contrast only dislocations having $\bar{1}\bar{1}1$ and 111 Burgers vectors are visible³⁶ since $\mathbf{g}\cdot\mathbf{b}=0$ for $\bar{1}\bar{1}1$ and $1\bar{1}1$. Little more can be concluded by observing foils in a $[110]$ orientation. However, when the zone axis is near $\langle 111 \rangle$ the specimen can be tilted to operated six 110 reflections, i.e. $\bar{1}01$, $0\bar{1}1$, $1\bar{1}0$, $10\bar{1}$, $01\bar{1}$, and $\bar{1}10$, and other dislocations can be seen. For example when the contrast is due to $\bar{1}01$, dislocations with Burgers vectors $\bar{1}\bar{1}1$ and $11\bar{1}$ are seen ($\mathbf{g}\cdot\mathbf{b}=0$ for $1\bar{1}1$ and 111). If the specimen is oriented so that the source of contrast is the $0\bar{1}1$ reflection, only dislocations with Burgers vectors $1\bar{1}1$ and $11\bar{1}$ are seen. However, the dislocation with Burgers vector $1\bar{1}1$ is uniquely determined because it remains in contrast for both the $\bar{1}01$ and $0\bar{1}1$ reflections. Furthermore the direction defined by this non-vanishing dislocation with $1\bar{1}1$ Burgers vector is



21 Dislocation configuration in deformed (10%) Fe-9%Ni alloy recrystallized at 500°C before deformation²⁸

verified by trace analysis to be $[1\bar{1}1]$, proving its pure screw nature. It was found* that all four possible $\langle 111 \rangle$ screws are present in the untwinned regions of the plates.

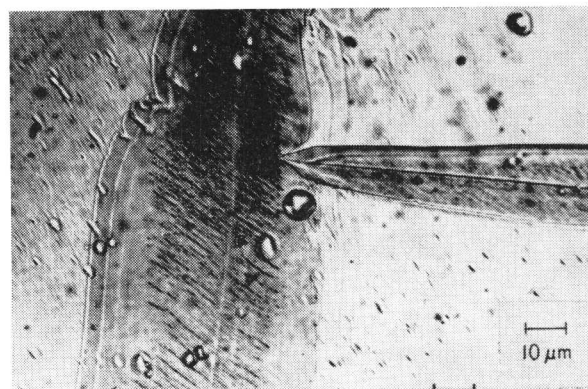
These $\langle 111 \rangle$ screw dislocations can glide in $\{112\}$, $\{110\}$, $\{123\}$ planes, etc., and thus the glide plane is not unique. Indeed the straightness of the $[\bar{1}11]$ and $[1\bar{1}1]$ screw dislocations in Figure 20 (zone axis $[110]$) suggests that they do not glide in the plane of the foil.

One naturally inquires about the origin of four sets of $\langle 111 \rangle$ dislocations in the martensite plates. If the inhomogeneous shear for the thickening stage of the plate is slip, one set of 'residual' transformation dislocations might be observed, but hardly four sets. However, if the growing plate is plastically deformed as it expands into the matrix, several variants of dislocations might be expected. To examine such a possibility the deformation structure of Fe-9%Ni which can be recrystallized in the all- α condition at 500°C¹⁴ was observed. Sheet tensile specimens 0.015 in thick were plastically strained in tension at room temperature for various amounts after the recrystallization treatment. A diamond-like array of dislocations (Figure 21) was also found†, and by analogy it seems reasonable to conclude that the 'extra' dislocations in the untwinned portions of martensite plates are a result of plastic deformation. The dislocation density in the untwinned regions is similar to that in annealed specimens plastically strained about 10%.

Interference microscope studies of the partially twinned Fe-Ni plates have been made,²⁸ and it was deduced that the shape deformation in the twinned and untwinned regions of the plates is identical. The possibility was considered that the plates may form entirely by an internal twinning mechanism, to be followed by detwinning (due to constraints) of those regions of the plate which showed only dislocations when examined by transmission electron microscopy. This was ruled unlikely because, if detwinning did occur, i.e. one of the two twin orientations growing at the expense of the other until only a single crystal remains, a noticeable shape change in the detwinned region (relative to the twinned region) would result.

*By measuring the angle between the $\langle 111 \rangle$ dislocations and the known habit plane, it can be deduced that all four variants of the $\langle 111 \rangle$ dislocations are present.

†Although these dislocations are not as straight as those in the martensite plates, their mean direction was also determined to be $\langle 111 \rangle$ and they are essentially screws.



22 Two intersecting martensite plates in Fe-32.0%Ni; optical micrograph, specimen polished and etched after transformation²⁸

By observing the interference fringes, this could not be detected.

It thus appears that the inhomogeneous shear involved for the thickening of the partially twinned plates is slip, and it seems that the plane of inhomogeneity, i.e. the initial twinning plane and the subsequent plane of slip, is a common $\{112\}_m$ plane, as suggested by the same shape deformation in the two regions. If the plane of inhomogeneity changes during the thickening, i.e. from $\{112\}$ to $\{110\}$ as suggested,³⁰ the shape deformation according to theory would be different for the different parts of the plate.

Two-stage thickening of martensite plates

Recent evidence²⁸ suggests that some Fe-Ni martensite plates do in fact thicken in two kinetically different stages (perhaps separated by a time lapse). Figure 22 is an optical micrograph (etched) showing two intersecting martensite plates in Fe-32.0%Ni. The vertical plate is only partially twinned (as revealed by the etching). The plate which intersects this plate from the right penetrates to the end point of the transformation twins in the vertical plate. The midrib in the vertical plate is displaced in the vicinity of the intersection, and it therefore seems clear that the twinned portion of the vertical plate was the first to form, to be followed by the formation of the horizontal plate whose growth caused a displacement of the original plate (jagged region of midrib near intersection). This is then followed by the thickening of the initial plate which of course could not grow in the region of the indentation. The absence of the characteristic twin etchings in the thickened portion of the initial plate suggests that the mode of inhomogeneous shear for the thickening of this plate is slip rather than twinning. Numerous cases similar to Figure 22 were observed. It was also noted²⁸ that the formation of subsequent plates (i.e. another burst at a lower temperature) caused further thickening of previously formed plates by the slip mechanism. Figure 22 also shows that, since the midrib of the horizontal plate penetrates furthest into the vertical plate, this region must represent the first part of the plate to form. The transformation twins in the horizontal plate (Figure 22) are not clearly seen because the etching effect which reveals them is orientation dependent, i.e. a plate of habit variant (hkl) will show the twins while a neighbouring plate of variant $(h'lk)$ will not.