The Direct Observation of the $\gamma \rightarrow \alpha'$ Isothermal Martensitic Transformation of Iron Alloys in Electron Microscope*

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Synopsis

Direct electron microscopy of isothermal martensitic transformation in iron-nickel base ternary alloys has been done in order to get positive information on some obscurities involved in the mechanism of $\gamma \rightarrow \alpha'$ martensitic transformation of iron alloys. Although the nucleation process was not observed in the regions free of pre-existing martensites, two levels of growth rate and two types of autocatalysis of martensitic laths were observed. By using the cinephotographic technique, intermittent motions of dislocations were revealed at and in front of the tip of growing laths. The nature of these dislocations were analyzed.

I. Introduction

There remain still controversial concepts on the mechanism of martensitic transformation in iron alloys. Concerning the nucleation of martensite, for example, the origin of nucleation site has been left unestablished. In other words, no clear evidence has been given for the existence of "embryos" which was proposed by Kaufman and Cohen,¹⁾ and lattice imperfections with which nucleation sites may be associated^{2,3)} have not been identified. The growth and propagation mechanisms of the transformation have been quite ambiguously clarified as well. It has not been defined whether so-called lattice-variant and lattice-invariant shears of the martensitic transformation take place simultaneously or successively. Correlation of the real lattice transformation mechanisms with those models as hypothesized in terms of dislocation motion by Jaswan,⁴⁾ Kelly and Nutting,⁵⁾ Bogers and Burgers⁶⁾ and others is also in need of elucidation. In addition, the effect of the formed martensite on occurrence of new defects in the surrounding austenite must be evaluated in connection with the propagation mechanism.⁷⁾

It is thought that dynamical observations of the transformation by thin foils in an electron microscope can provide positive information on these obscurities involved in the mechanism of transformation. However, no investigation has appeared yet as reporting a success in direct observation of the transformation. It seems to be mainly due to that the foils are distorted by martensitic formation during thinning or cooling and no detailed examination can be done.³⁾

This paper is concerned with direct observations of the isothermal transformation to martensite in an electron microscope.

II. Experimentals

Experimental alloys were made from electrolytic iron and other alloying materials of high purity by vacuum induction melting. The ingots were homo-

genized at 1300°C for 3 days and then hot worked and cold rolled into sheets 0.4 mm thick. The chemical analysis of the sheet is found in Table 1.

Samples cut from the sheets were subjected to austenitization in an Ar atmosphere either at 900°C for 30 min or at 1100°C for 1 hr and subsequently cooled down to room temperature. When these samples were held at subzero temperatures, the isothermal transformation was exhibited. The transformation behaviour is shown in Fig. 1 in terms of change in electric resistance as a function of holding time. The obtained values of resistance at various temperatures were normalized to that at 0°C being equated 100. It is clearly seen in the figures that the transformation proceeds isothermally in all alloys NCR, NMO and NMN. The electric resistance was measured by using an amplified recorder with high stability constant current generator and constant voltage generator. Samples were immersed into a specifically designed bath with ethyl alcohol and liquid nitrogen, the temperature of which was controlled within $\pm 1^\circ\text{C}$ utilizing an electric heater, a stirrer and thermostat.

First, the features of bulk transformation products were examined. Foils were prepared from samples austenitized and isothermally transformed as described above. It is now considered on these observations that the substructures of isothermal transformation in bulk specimens includes dislocated martensite laths which are usually observed in the packet type martensite.⁸⁾

Then electron microscopy was applied to *in situ* observation of isothermal martensitic transformation. The nucleation stage of transformation was inspected on samples electrolytically thinned in phosphoric and chromic acid solution directly after austenitization. The foils were cooled in the electron microscope so as to detect the occurrence of martensite phase. The growth and propagation of the martensitic regions were surveyed on the samples partially transformed in the cooling bath after austenitization. Foils were then prepared and re-cooled down to subzero temperatures in the electron microscope to pursue further progress of the transformation.

Table 1. Chemical Composition of alloys used (wt%)

	Fe	Ni	Cr	Mo	Mn	C	N
NCR	bal.	25.7	2.76			0.001	0.0016
NMO	bal.	25.3		4.52		<0.001	0.009
NMN	bal.	25.4			2.17	<0.001	0.0010

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Fig. 1. Isothermal martensitic transformation curves measured by electric resistivity normalized to the 0°C value being equated 100.
(a) Alloy NCR (b) Alloy NMO (c) Alloy NMN

JEM-150 electron microscope equipped with JEM AC-H (conduction type cold stage) was operated at 150 kv.

The transformation occurrence was also monitored by means of cinephotographic technique with a 16 mm cine camera charged with Fuji R250 cinefilm.

In order to obtain correct information valid for the bulk transformation from experiments made on foil specimens, it is necessary to exclude any interference caused by surface martensites and thinning martensites from observations. Thus, a number of foils containing martensites isothermally formed in bulk samples were examined⁸⁾ and martensites with different appearances from those of bulk martensites were omitted from observation. Photograph 1 shows an

Photo. 1. Example of a martensite transformed by thinning effect (Alloy NMN, austenitized at 1000°C for 45 min and cooled in the electron microscope)

example of the discriminating microstructure formed due to thinning effect instantaneously during cooling in the electron microscope. This has a clearly distinguishable feature from that of isothermal martensite formed in bulk samples. In the case of alloy NMN this thinning effect was especially strong and the direct observations of transformation were difficult. So the direct observations were carried on only in alloys NCR and NMO. Formation of the thinning martensite did not initiate during preparation of foils at room temperature, that was confirmed by examining foils without subzero-cooling after austenitization.

III. Results

1. Nucleation

Through this investigation the nucleation process was not observed inside the field of view under the electron microscope despite a number of careful examinations on the foils. Thus the direct observation of the nucleation in an electron microscope has not been accomplished yet in the area of foils where no martensite laths pre-exist.

2. Growth and Propagation

Photograph 2 shows growth of laths in alloy NCR. The foil was cooled in the electron microscope after austenitization at 1100°C for 1 hr and subsequent subzero cooling at -78°C for 10 min. The lapse of time is indicated for each micrograph, Photo. 2 (a) being the starting-point. It was difficult to keep the temperature of foils accurately constant, and the temperature during taking a photograph is shown in parenthesis. At the center of these micrographs two laths are recognized to grow with gradual twisting. The average growth rate of the laths can be estimated using point contrasts indicated by arrows as a mark. The upper lying lath exhibited an average growth rate of about 0.1 $\mu\text{m}/\text{min}$ between Photos. 2 (a) and (d), whereas the lower one grew at a mean speed of some 0.03 $\mu\text{m}/\text{min}$.

Photograph 3 presents another example of growth propagation sequence of martensite laths in alloy NCR. The growth rate in general was slow as in the

Photo. 2. Electron micrographs showing growth of martensite laths in the microscope (Alloy NCR, austenitized at 1100°C for 1 hr and transformed at -78°C for 30 min)

Photo. 3. Electron micrographs showing growth of martensite laths in the microscope. Rapid growth of laths happened in portions A, B and C (Alloy NCR, austenitized at 900°C for 30 min and transformed at -78°C for 10 min)

cases of Photo. 2. However, a rapid growth of laths also happened in some portions like A, B and C centered on the micrographs. The growth rate of these portions was only roughly estimated to be in the order of 1 to 10 $\mu\text{m}/\text{sec}$.

Photograph 4 shows still another growth mode of martensite laths observed in alloy NMO. The foil surface is almost parallel to $\{001\}_\gamma$ and the lath direction to $\langle 110 \rangle_\gamma$. Laths seem to have grown perpendicular to the interface of a pre-existing lath. Inter-

Photo. 4. Electron micrographs showing growth of martensite laths in the microscope. Laths seem to have grown perpendicular to the interface of a pre-existing laths. Intermittent motions of dislocations were recognized inside laths and in the austenite near the tip of laths. (Alloy NMO, austenitized at 900°C for 30 min and transformed at -78°C for 10 min)

mittent motions of dislocations were recognized inside martensite laths and in the austenite near the tip of laths. Photograph 5 shows a sequence of such dislocation motions recorded on a 16 mm cinefilm at 12 frames per second, and the micrographs being picked out from every 20 frames. Parallel dislocations are seen moving forwards simultaneously with the advance of a lath front M. The Burgers vector of these dislocations was decided as following.

3. Decision of the Burgers Vector of the Parallel Dislocations

Photograph 6 is an image of alloy NCR in the condition of $\langle 200 \rangle_\gamma$ Bragg two beam orientation, the main beam direction being nearly normal to the $\{001\}_\gamma$ plane. Photograph 7 is an image of the same foil in the condition of $\langle 020 \rangle_\gamma$ Bragg two beam orientation, which is normal to that in the case of Photo. 6. The parallel dislocations are vanishing.

Photograph 8 is a bright field image of alloy NMO, the main beam direction is nearly normal to the $\{001\}_\gamma$ plane. Photograph 9 is a dark field image from the $\langle 200 \rangle_\gamma$ spot. The parallel dislocations which are being analyzed are vanishing.

Photograph 10 is an image of alloy NMN in the condition of the $\langle \bar{1}11 \rangle_\gamma$ Bragg two beam orientation. The main beam direction is nearly normal to the $\{110\}_\gamma$ plane. Photograph 11 is an image of the same foil in the condition of $\langle 1\bar{1}1 \rangle_\gamma$ Bragg two beam orientation. The parallel dislocations are vanishing.

From Photos. 6 to 11 we can draw schematically the interrelation between the lath and the these parallel dislocations. Supposing the habit planes of lath

being $\{1\bar{1}1\}_\gamma$ and $\{1\bar{1}2\}_\gamma$ as shown, Figs. 2 to 4 are gotten. The indices of habit planes were not decided by the precise orientation analysis but by coarse trace analyses of Photos. 6 to 11. These figures are revealing the parallel dislocations being $a/2\langle 011 \rangle_\gamma$ screw dislocations in the case of Photos. 6 to 9 and $a/2\langle 101 \rangle_\gamma$ mixed dislocations in the case of Photos. 10 and 11.

IV. Discussion

1. Nucleation

According to Raghavan⁹⁾ and Pati and Cohen,¹⁰⁾ the number of nucleation sites is of an order of $10^7/\text{cm}^3$. If this is true, one nucleation site would be encountered by examining about 3 000 fields of view in the electron microscope. Magee⁷⁾ and Shih *et al.*¹¹⁾ estimated this number to be about $10^5/\text{cm}^3$, which would lead to less probability of finding one nucleation site. The number of fields of view inspected in this study amounted to only 100 or thereabouts, so it seems quite likely that no nucleation event was observed in the lath-free area being examined.

The formation of martensite outside of the field of view may involve autocatalytic nucleations due to strain induced martensite generated by mounting a foil in the sample holder. Distortion contours associated with the formation of martensite outside the monitoring area or with thermal inhomogeneity in the foils were observed on the screen during cooling in the electron microscope.

On the basis of the present results it appears possible to achieve direct observation of the nucleation

Photo. 5. Cinemicrographs recorded at 12 frames per second revealing a sequence of dislocation motions, micrographs being picking out from every 20 frames

process of martensite by a futural investigation, taking into account the effects of incidental formation of martensite mentioned above.

2. Growth Rate

Bunshah and Mehl,¹²⁾ Beisswenger and Scheil,¹³⁾ and Mukherjee¹⁴⁾ measured the growth rate of twinned

Photo. 6. Electron microscopic image of an isothermal martensite lath and the surrounding austenite in the condition of $\mathbf{g}=\langle 200 \rangle_{\gamma}$ Bragg two beam orientation. The main beam is nearly normal to the $\{001\}_{\gamma}$ plane. (Alloy NCR, austenitized at 900°C for 30 min and transformed at -78°C for 10 min)

Photo. 7. Image of the same foil as Photo. 6 in the condition of $\mathbf{g}=\langle 020 \rangle_{\gamma}$ Bragg two beam orientation. The parallel dislocations are vanishing.

martensite to be 10^5 cm/sec. Marder and others reported the growth rate of dislocated martensite (M_s temperature was higher than room temperature) to be 800 $\mu\text{m}/\text{sec}$ in Fe-C alloys¹⁵⁾ and 72 to 400 $\mu\text{m}/\text{sec}$ in Fe-Ni alloys.¹⁶⁾ In the present experiments two levels of growth rate were revealed. One is much lower than those informed in all of the earlier investi-

Photo. 8.
Bright field image of isothermal martensite laths and the surrounding austenite. The main beam is nearly normal to the $\{001\}_\gamma$ plane. (Alloy NMO, austenitized at 900°C for 30 min and transformed at -78°C for 10 min)

Photo. 9.
Dark field image of the same foil as Photo. 8 from the $\mathbf{g}=\langle 200 \rangle_\gamma$ spot. The parallel dislocations which are being analyzed are vanishing.

gations. The other is comparable to those reported in dislocated packet martensite transformation. It should be noted, however, that the growth rate estimated in the present study is concerned with an experiment on foils and not on bulk samples.

The present results indicated that the growth of individual laths was considerably discontinuous with a variety of growth rates. In this regard the growth mechanism will be discussed in a later paragraph.

Determination of the growth rate of martensite in the nucleation stage must offer some keys to the interpretation of the transformation mechanism and is a serious subject still in the present study.

3. Propagation Mechanism

To interpret the nucleation mechanism of marten-

sitic transformation and consequently the interrelation among isothermal, packet-type (massive), and lenticular (burst-like) martensite transformations, it is quite important to know how a martensite crystal formed causes the initiation of new crystals in the surrounding matrix.⁶⁾ Marder and Krauss,¹⁵⁾ Marder and Marder,¹⁶⁾ and Pubrov¹⁷⁾ reported the type of propagation of laths side by side with one after another in the packet martensite transformation. They observed other types of propagation at the same time. In the present study some other types of propagation of lath than the "side by side" type (Photos. 2 to 4) were observed. As an example of these types of propagation, Photo. 4 exhibits the formation of new laths normal to the pre-existing lath. Photograph 6 reveals the parallel dislocations B. These are analyzed on

Photo. 10.

Electron microscopic image of isothermal martensite laths and the surrounding austenite in the condition of $\mathbf{g}=\langle\bar{1}11\rangle_{\gamma}$ Bragg two beam orientation. The main beam direction is nearly normal to the $\{110\}_{\gamma}$ plane. (Alloy NMN, austenitized at 900°C for 30 min and transformed at -78°C for 10 min)

Photo. 11.

Image of the same foil as Photo. 10 in the condition of $\mathbf{g}=\langle\bar{1}11\rangle_{\gamma}$ Bragg two beam orientation. The parallel dislocations are vanishing.

the $\{111\}_{\gamma}$ planes being conjugate with planes on which the parallel dislocations A are lying. These parallel dislocations B might interrelate with the formation of new laths normal to the pre-existing lath.

4. Growth Mechanism

Electron metallography of the bulk transformed isothermal martensite together with that of the well established packet martensite suggests that the lath martensite grows discontinuously.⁸⁾ A direct observation of the transformation in the present study verified this discontinuous growth of laths. Photograph 3 exhibits a variety in growth rate of laths. Photograph 2 involves different levels of slow growth rate. Photograph 5 reveals the intermittent advance of a lath front in a more microscopic scale.

The necking portion of laths transformed in foils

and variation in their image contrast, as seen in Photos. 2 and 3, were also recognized in relation to isothermal and packet martensite formed in bulk samples, which would suggest occurrence of a favorable accommodation of transformation stresses during growth process through small variations in the fcc $\rightarrow$ bcc orientation relationship.⁸⁾

It is thought that the dislocation motion detected in the areas adjacent to the front of growing laths can be closely related to the transformation mechanism. Jaswon,³⁾ Kelly and Nutting,⁴⁾ Venables⁸⁾ and Bogers and Burgers⁵⁾ made attempts to explain the fcc $\rightarrow$ bcc lattice transformation in terms of partial dislocation motions on $\{111\}_{\gamma}$. Though no verification of their models has yet been performed, it is quite likely that some information related to the mechanism will be provided by investigating the nature of dislocation

Fig. 2. Schematic illustration showing the interrelation between the lath and the parallel dislocations in the austenite, corresponding to Photos. 6 and 7. Upper picture is a section by a plane of thin foil.

Fig. 3. Schematic illustration showing the interrelation between the lath and the parallel dislocations in the austenite, corresponding to Photos. 8 and 9. Upper picture is a section by a plane of thin foil.

Fig. 4. Schematic illustration showing the interrelation between the lath and the parallel dislocations in the austenite, corresponding to Photos. 10 and 11. Upper picture is a section by a plane of thin foil.

Fig. 5. Schematic illustration showing the interrelation between the Burgers vectors and the elongated dislocations in the austenite adjacent to the tip of the martensite lath

motion near the tip of growing laths.

Some transient faults on $\{111\}_\gamma$ in front of growing laths during $\gamma \rightarrow \alpha'$ transformation were observed in an Fe-32.3%Ni alloy by Brown¹⁹⁾ and in Fe-33.1%Ni, Fe-27%Ni-0.4%C, and Fe-24%Ni-3%Mn alloys by Magee.⁷⁾ Shimizu *et al.*²⁰⁾ reported $\{111\}_\gamma$ associated faults in $\{225\}_\gamma$ martensite of an Fe-8%Cr-1.1%C alloy. These faults were inferred to act as nuclei stimulating further martensite formation.

In the present study we observed the parallel dislocations in the austenite adjacent to the tip of laths and these dislocation moved intermittently with the growth of laths. It is reasonable to think that these dislocations are relieving the stress of the transformation. As for the mechanism of the lattice invariant deformation of the martensite transformation there is Suzuki's model.²¹⁾ In this model most of transformation stress in the tip of martensite plate can be relieved by $a/2\langle 101 \rangle_\gamma$ screw dislocations moving on the austenite-martensite interface in the growing direction of the plate. The results of the present study are re-

vealing some part of these stress relief occurring in the austenite.

On the other hand, Photo. 5 shows that the dislocation movement is complex in the very adjacent region of the tip of laths. In such region we can observe the dislocations elongating into the growing direction of the lath in Photo. 8. From Photo. 8 we can decide these dislocation D locating on the $\{1\bar{1}1\}_\gamma$ plane P as showing in Fig. 5. Photograph 12 is an image of the same foil of Photo. 8 in the condition of $\langle 200 \rangle_\gamma$ Bragg two beam orientation being normal to these dislocations. So the Burgers vector of these dislocations are not $\mathbf{b}_a$ (screw) in Fig. 5. Photograph 13 is an image of the same foil in the condition of $\langle 002 \rangle_\gamma$ Bragg two beam orientation. Photographs 9 and 13 reveal that the Burgers vector is not $\mathbf{b}_b$ and $\mathbf{b}_c$ in Fig. 5, respectively. Photograph 14 is a dark field image of $\mathbf{g} = \langle 202 \rangle_\gamma$, showing that the Burgers vector

Photo. 12.

Image of the same foil as Photo. 8 in the condition of $\mathbf{g} = \langle 202 \rangle_\gamma$ Bragg two beam orientation being normal to the elongated dislocations

Photo. 13.

Image of the same foil as Photo. 8 in the condition of $\mathbf{g} = \langle 002 \rangle_\gamma$ Bragg two beam orientation

Photo. 14. Dark field image of the same foil as Photo. 8 from the $\mathbf{g}=\langle 202 \rangle_r$ spot

of these elongated dislocations is not $\mathbf{b}_d$ (edge) either. These dislocations are thought to be movable, and they are not Frank dislocations. As a result these elongated dislocations might be partial dislocations.

The motion of dislocations in the martensite can be seen too by the direct observation of the transformation in the electron microscope. Some of these dislocations are elongated to the lath direction (Photo. 4).

More detail observations of these dislocations at and in front of the tip of the growing laths might give further information about the mechanism of the martensite transformation.²²⁾

V. Summary

The present study deals with the direct electron microscopy of isothermal transformation to martensite in iron alloys. The results obtained may be summarized as follows.

(1) Nucleation of the martensite in virgin austenite was not detected even by a number of observations tried.

(2) Two levels of growth rate of isothermal martensite were observed. The higher rate was of the order of $1 \sim 10 \mu\text{m}/\text{sec}$ and the other one was slower than $0.1 \mu\text{m}/\text{min}$.

(3) The propagation of martensite was found to proceed in the two ways, *i.e.*, the successive formation of laths by the so called side-by-side mechanism and the initiation of new laths oblique to the pre-existing ones.

(4) Intermittent motions of dislocations at and in front of the tip of the growing laths were revealed by using the cinephotographic technique.

(5) Of these dislocations the parallel dislocations in the austenite near the tip of laths were analyzed to be $a/2\langle 110 \rangle_r$ screw and mixed dislocations.

(6) Elongated dislocations in the austenite very adjacent to the tip of the lath might be partial dis-

locations.

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