Electron Microscopic Observation of the Austenite and the Martensite in High Aluminium Steel

By Tsugio Tadaki*, Ken'ichi Shimizu*
and Toshimitsu Watanabe**

The austenite and martensite phases of Fe-9.70% Al-1.46% C steel have been observed by means of transmission electron microscopy and electron diffraction. It was verified that the austenite γ phase has a superlattice of the Cu_3Au type, and the assumption in a previous paper (Trans. JIM, 11(1970), 152) that the martensite lattice would be formed by the Bain distortion from the Cu_3Au type ordered γ lattice was confirmed unambiguously. Besides, it was found that the ordered γ phase consists of anti-phase domains of about 120 Å size. From the appearance of cross streaks around the fundamental spots as well as superlattice spots in the $\langle 100 \rangle$ directions, the anti-phase domain boundaries were identified to be of the second type.

(Received March 22, 1971)

I. Introduction

A high aluminium steel has been remarked as one of alloys which might give us some information on the Bain distortion and lattice invariant shear in the crystallographic theory of martensitic transformation in ferrous alloys⁽¹⁾⁽²⁾. Nishiyama et al. observed martensites in such a high aluminium steel by means of electron microscopy and diffraction, and obtained some results as reported in the previous paper⁽²⁾.

In that study, they found that the martensite crystal gave the diffraction pattern of a body-centered tetragonal lattice with extra spots. Those extra spots were interpreted as the reflections from the superlattice which was assumed to be formed by the Bain distortion from the parent γ lattice of the Cu_3Au type. Whether the parent γ lattice was of the Cu_3Au type or not, however, could not be confirmed because the γ lattice was not found.

To confirm the above questionable point in the previous paper is the purpose of the present study.

II. Experimental Procedures

The processes of making an alloy ingot are the same as before⁽²⁾, the materials being electrolytic iron, aluminium of 99.99% purity, and graphite of 99.5% purity. The chemical analysis of the alloy gave

9.72 wt% Al and 1.47 wt% C at the top,

9.68 wt% Al and 1.44 wt% C at the bottom

of the ingot of $50 \times 50 \times 150$ mm, which assures the uniformity of composition. The ingot was annealed for 1 hr at 1230°C in an evacuated quartz capsule and then cut into discs of about 0.3 mm in thickness. These discs were again heated for 1 hr at 1230°C in evacuated quartz capsules, followed by quenching into 0°C water. To suppress precipitation, capsules were smashed immediately after quenching into water.

* Institute of Scientific and Industrial Research, Osaka Univ., Yamadakami, Suita, Osaka, Japan.

** Fundamental Research Laboratories, Nippon Steel Corporation, Kawasaki, Japan.

Thin foils of specimens for transmission electron microscopy were made with the same method as before⁽²⁾. The electron microscope used was of a HU-11 D, operated at 100 kV.

III. Experimental Results and Discussion

1. Structure of as-quenched specimen

The previous experiment was not able to retain the parent γ phase at room temperature while the present experiment could successfully do so probably due to the differences in heating atmosphere and quenching method. Photo. 1 (a), which was taken from the specimen as-quenched, is one example of electron micrographs showing the retained γ matrix as well as the martensitic products. (b) is the electron diffraction pattern taken from the martensitic region I in (a), which consists of intense reflections and weak reflections. The intense reflections and a part of the weak reflections can be explained to be the $[311]$ zone pattern of the ordered martensite crystal as proposed previously⁽²⁾, the weak spots being those of the superlattice. The remaining weak reflections are explained to consist of the $[\bar{1}35]$ zone pattern from the twinned crystal on the (112) plane and double reflections. However, they can also be explained to be intersections of the elongated reciprocal lattice points of the untwinned martensite crystal with the Ewald sphere, the elongation being due to the existence of stacking faults and/or twin faults and the direction of the elongation being normal to the (112) plane. In any case, such appearances of the weak reflections are consistent with the result of observations in the previous experiment. For simplicity, let us consider the remaining weak reflections to be the $[\bar{1}35]$ zone pattern of the twinned crystal.

Examining in detail the electron diffraction pattern. (b), the observed d-spacings and angles between reflecting planes are different from those of the ideal $[311]$.

(1) Z. Nishiyama and M. Doi: J. Japan Inst. Metals, 8 (1944), 505.

(2) Z. Nishiyama, K. Shimizu and M. Harada: Trans. JIM, 11 (1970), 152.

and $\langle \bar{1}35 \rangle$ zone patterns. This means that the direction of the incident electron beam is not exactly parallel to the ideal zone axes. Such appearances of reciprocal lattice

Photo. 1 (a) Electron micrograph of Fe-9.70 % Al-1.46 % C steel as-quenched, showing the retained γ matrix (region II) as well as the martensitic product (region I).
 (b) Electron diffraction pattern taken from the martensitic region I in (a). This consists of the $[311]$ zone's of the martensite and the $[135]$ zone's due to twin faults or stacking faults on the (112) plane.
 (c) Electron diffraction pattern taken from the retained γ matrix in (a) (region II), showing the $[211]$ zonal pattern of the Cu_3Au type superlattice.

points off the Ewald sphere may be attributed to the intersections of the elongated one's with the Ewald sphere, as explained above. Numerical analyses based on this consideration showed that the beam direction is deviated by 5.1° from the $[311]$ axis and by 2.4° from the $\langle \bar{1}35 \rangle$ axis of twinned crystal[†]. As the result of such analyses, the martensite crystal was identified to be of the ordered bct with the lattice constant of $a = 2.83 \text{ \AA}$ and the tetragonality of $c/a = 1.11$, this result being consistent with that obtained in the previous experiment⁽¹⁾⁽²⁾. The inclination of the (112) fault plane to the specimen surface was 34° , which is consistent with the indistinctness of striations due to the faults.

(c) is a diffraction pattern taken from the region II in (a). This was analysed to be the $\langle 2\bar{1}1 \rangle$ zone pattern of fcc lattice which shows undoubtedly the retaining of the parent γ phase. In (c), extra spots are seen in addition to the fundamental fcc spots. These extra spots will be discussed in detail in the next section.

2. Crystal structure of the retained γ phase

Photos. 2 (a), (b), (c) and (d) show typical zone patterns of electron diffraction taken from the retained γ phase. From the distributions of fundamental spots, their zone axes were indexed as $\langle 001 \rangle$, $\langle 110 \rangle$, $\langle 111 \rangle$ and $\langle 0\bar{1}2 \rangle$ of the fcc lattice, respectively. Then, the distributions of extra spots are almost identical to those of superlattice spots from the Cu_3Au type ordered lattice. That is, extra spots can be indexed with (hkl) of any combinations of integers with exception of indices for fundamental reflections of fcc lattice. The present alloy, moreover, contains aluminium atoms of about 18% in atomic percentage. Therefore, the γ phase can be concluded to have the Cu_3Au type ordered structure^{††}. Extra spots in Photo. 1 (c) can also be explained consistently with this structure.

Considering that the alloy used contains carbon atoms of about 6% in atomic percentage, there is a possibility of local formation of the Fe_3AlC perovskite type structure⁽³⁾. In fact, the possibility of such an ordering of carbon atoms can be known from the intensity anomalies of superlattice spots in electron diffraction patterns. That is, the intensities of superlattice reflections of $h+k+l=\text{even}$ are slightly weaker than those of superlattice reflections of $h+k+l=\text{odd}$, as seen in Photos. 2 (a), (b) and (d). A strong tendency of the Fe_3AlC perovskite formation has been found by Oshima and Wayman⁽⁴⁾. There is also an experiment which suggests possibility of the formation of the perovskite⁽⁵⁾. These experiments, however, were carried out using aluminium steels of higher carbon ($\approx 2\% \text{ C}$) and lower aluminium concentrations. The perovskite observed in the reference (5) seems to be formed in the domain boundary region, and it

† The $[311]$ direction makes an angle of 6.1° to the $\langle \bar{1}35 \rangle$ direction.

†† The lattice constant was determined to be $a_f = 3.69 \text{ \AA}$ referring the camera length corrected by using TiCl .

(3) L. S. Palatnik, I. A. Tananko and Yu. G. Bobro : Soviet Phys. Cryst., 9 (1964), 163.

(4) R. Oshima and C. M. Wayman : to be published.

(5) T. Watanabe : unpublished work.

Photo. 2 Various electron diffraction patterns taken from the retained γ matrix. (a), (b), (c) and (d) show the $[001]$, $[110]$, $[111]$ and $[012]$ zonal patterns of the Cu_3Au type superlattice, respectively.

may be the decomposition product that was formed together with the γ phase of a lower carbon concentration, the γ phase changing to martensitic products. In that case, the decomposition was observed to have taken place along the cube planes of matrix γ phase, and the size of the decomposition products was nearly 0.1 to 0.3μ . In the present case, however, such a decomposition structure was scarcely observed, but anti-phase domains of about 0.01μ were mainly observed as will be described below.

3. Out-of-step structure of the γ phase

In Photos. 2 (a), (b) and (d), there are recognized short streaks around superlattice spots as well as fundamental spots in the $\langle 100 \rangle$ directions. Those around superlattice spots may be explained by taking account of the existence of ordered domains in the similar way to the case of Cu-Au alloy⁽⁶⁾⁽⁷⁾ though they do not show such clear splitting as in the latter case. However, further considerations will be needed for the appearance of those streaks around fundamental spots. At all events, in order to know the origin, domain structures were observed by the bright field and dark field methods of electron microscopy. Photo. 3 (a) is the bright field

Photo. 3 (a) Electron micrograph of the retained γ matrix, showing the maze pattern due to anti-phase domains.

(b) Dark field image of (a) formed by the (010) superlattice reflection.

(6) S. Yamaguchi, D. Watanabe and S. Ogawa : J. Phys. Soc. Japan, 17 (1961), 1030.

(7) R. M. Fisher and M. J. Marcinkowski : J. Appl. Phys., 31 (1960), 1687; Phil. Mag., 6 (1961), 1385.

image of the specimen in (001) orientation. There is seen the maze pattern of ordered domains as observed in many other ordered alloys. (b) is the dark field image formed by the (010) superlattice reflection, which was taken only by displacing an objective lens aperture. In this dark field image, domain boundaries are observed as lines nearly parallel to the (100) and (010) planes and as blocks parallel to the (001) plane.

It has been known that there are two types of anti-phase domain boundaries (APB's) through cube planes in the Cu_3Au type ordered structure. The first type is formed by out-of-step of $1/2\langle 110 \rangle$ vectors lying on the APB planes. On the other hand, the second type is formed by out-of-step of vectors lying on any one of the cube planes perpendicular to the APB planes. An important feature of the first type APB is that Au atoms do not come in contact across the APB's in the case of Cu_3Au alloy, and thus the first type APB is expected to be of lower energy. On the other hand, the second type APB causes a chance for two Au atoms to occupy the nearest neighbor positions across the APB, and is expected to be of somewhat higher energy than the first type APB. Fisher and Marcinkowski⁽⁷⁾ and Yamaguchi et al.⁽⁸⁾ studied in detail the contrast of APB's and established the method to determine which type is the APB's under observations.

Applying their method to the present Photo. 3 (b), it can be concluded that most of the APB's observed are of the second type. Although this result is unfavorable in the energetical viewpoint, it may not be so unreasonable if one consider the situation that the specimen is as-quenched from a high temperature and so in the non-equilibrium state. As described above, the existence of the second type APB's suggests that Al atoms occupy the nearest neighbor positions across the APB's with high proportion and then that there are lattice distortions through each cube plane. These distortions can be considered to cause the cross-streaks around fundamental spots as observed in Photos. 2 (a), (b) and (d). The perovskite formed coherently on {001} planes may also produce such streaks because the lattice parameter difference between their {001} planes is very small ($3.746 \sim 3.786 \text{ \AA}$ ⁽⁸⁾ for perovskite and $3.69 \text{ \AA}$ for ordered γ phase).

4. Habit plane determination of martensite plate

The habit planes were determined for seven martensite plates by the trace analyses using the orientations of parent γ phase. The results are shown stereographically in Fig. 1. In the figure, seven arcs are parts of the great circles of zone planes containing the boundary traces of seven martensites. According to the results,

Fig. 1 Stereographic one-face trace analysis of the habit planes of seven martensite plates in high aluminium steel.

the habit plane of martensites in the Fe-Al-C alloy is located near the $\{3, 15, 10\}_f$ pole though it deviates by a few degree toward the opposite side of the $\{295\}_f$ pole. This result is consistent with that obtained precisely with the two-face method by Watanabe and Wayman⁽⁸⁾.

The orientation relationship between the martensite and austenite was tried to determine. However, it was difficult to obtain a consistent result possibly due to the existence of marked elongations of reciprocal lattice points of martensite crystals.

IV. Conclusions

An electron microscopy study has been made on the retained γ phase in an Fe-9.70 Al-1.46 C alloy water-quenched from 1230°C , with the following results:

(1) The crystal structure of the retained γ phase was of the Cu_3Au type ordered one. This result proves the assumption in the previous paper and also the validity of the Bain distortion in the martensitic transformation in Fe-Al-C alloy.

(2) The size of ordered domains in the retained γ phase was $120 \text{ \AA}$ on the average and the anti-phase domain boundaries were identified to be of the second type for the most part. The domain boundaries were attributed to the formation of cross streaks in electron diffraction patterns as well as the formation of the Fe_3AlC perovskite.

(3) The habit plane of martensite plates was in the vicinity of the $\{3, 15, 10\}_f$ plane although deviated from it toward the opposite side of the $\{295\}_f$ plane.

Acknowledgments

The authors are greatly indebted to Emeritus Prof. Z. Nishiyama for his encouragement and discussion. Critical discussions with Prof. C. M. Wayman and Dr. R. Oshima are also gratefully acknowledged.

(8) M. Watanabe and C. M. Wayman: to be published in Met. Trans.