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Effects of high austenitizing temperature and austenite deformation on formation of martensite in Fe-Ni-C alloys

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The effects of high austenitizing temperature and the deformation of austenite matrix below the range of strain-induced martensite formation on the morphology, substructure and crystallography of martensite formed in different Fe-Ni-C alloys have been studied by means of transmission electron microscopy. The formation behaviours of both thermal and stress-assisted martensites were examined under various physical conditions and martensite morphology was found to be closely dependent on the high austenitizing temperatures besides the influence of austenite deformation. Although the orientation relationship between austenite and thermally induced martensite was found as the Kurdjumov-Sachs type, it was also observed to change to Nishiyama-Wasserman type in the samples transformed under the stress-assisted conditions.

1. Introduction

Early studies on the formation of martensite in different ferrous alloys revealed that the formation mechanism and substructure of martensite are considerably altered by the transformation temperature, alloy composition, strengths of the material in the austenite and martensite phases, cooling rate of the matrix structure during the transformation and the austenite stacking fault energy in addition to the well known effects of the austenite grain size and the formation sequence of the martensitic structures [1–5]. Previous results on the morphology of ferrous martensites were summarized in a review by Mc Dougall and Wayman [6] and the observed morphologic and structural properties of martensite were discussed in conjuction with the predictions of the present crystallographic theories, these authors examined the influence of all possible effects in detail by using the available data and concluded that the attempts to identify the particular effects of these variables on morphology are rather complicated since they may produce interrelated changes in the various thermodynamic quantities. A similar result was also reached by Visvesvaran [7] after the observation of martensite formations in some Fe-Ni and Fe-Ni-C alloys and it was found difficult to make a general comment concerning the possible effects of the transformation temperature and the composition especially on the martensite substructures of the examined alloys. In a recent review James and Hane [8] also examined the dependence of martensite microstructure on the lattice parameters of the parent and product structures in shape memory alloys and pointed out the importance of these parameters on the general morphology of the martensite structures. Zeng *et al*. [9] investigated the effect of austenitizing temperature on the martensite morphologies of medium and high carbon steels and attempted to explain the dependence of martensite morphology to this temperature. According to their conclusions chemical composition becomes more homogeneous at higher austenitizing temperatures and observable changes occur in the essential characteristics of the martensite formation. Furthermore high temperature heating of the austenite matrix was also reported to influence the strain energy of the martensitic transformation by changing the density of lattice imperfections. Although a different interpretation was offered, their results still agree well with those of the early studies on various ferrous alloys in which the austenitizing temperature was found to alter the grain sizes and defect structures of the austenite and also the martensite start temperatures [9, 10].

On the other hand, it is well established that the deformation of austenite matrix structure prior or during the martensitic transformation create big changes on almost all essential features of the martensitic transformation due to the formation of crystal lattice defects. Studies have been carried out with a view to estimate the influence of deformation since late 1950's [11] and revealed important results in different ferrous alloys concerning the morphology, substructure and crystallography of the martensite formed in deformed austenite structures.

It is evident from the published experimental data that the austenitizing temperature and applied stresses greatly influence the martensitic transformation behaviour in ferrous alloys. Although numerous work have been reported to clarify the subject, it is hardly possible to give a generalized model for a fair understanding to describe the effects of these two factors in the same alloy groups. It seems therefore necessary to perform more experimental studies with various alloys and compositions to observe the role of the both effects on the morphology, substructure and crystallography of the martensite formation. Therefore it was thought worthwhile in the present study to examine the possible effects of austenitizing temperature and externally applied plastic deformation on the formation of thermally induced martensite for different Fe-Ni-C alloys whose several transformation characteristics are well defined in the literature.

2. Experimental

Three Fe-Ni-C alloys which all had sub-zero martensite start temperatures (M_s) were prepared by vacuum induction melting: Fe-21.3%Ni-0.13%C ($M_s \approx -40°C$), Fe-40.2%Ni-0.36%C ($M_s \approx -55°C$) and Fe-17.1%Ni-0.81%C ($M_s \approx -65°C$). Samples were austenized at various temperatures between 900–1350°C for 12 hours in a vacuum of 10^{-5} torr and this procedure yielded austenite grain sizes changing in about 200 μm and 4.5 mm ranges. The M_s values given above were determined for the samples austenized at 1200°C temperatures. Specimens were cooled at the same rate by slow withdrawal from the furnace to maintain a constant vacancy content. Transformation to martensite was observed by quenching austenite specimens into liquid nitrogen-methanol baths and the M_s temperatures were measured by dilatometry in an Adomel-Lhomargy LX-02 dilatometer. Compression specimens were prepared by spark-cutting and electrolytic polishing and examined with a compression test machine. A crosshead speed of 100 μmin^{-1} was used during the all experiments and the load cell of the machine was calibrated to 0.5% prior to each test to measure the applied stresses precisely. Thin foil samples used in transmission electron microscope (TEM) observations were prepared from 3 mm discs electropolished by using a double-jet polishing technique with a solution of methanol-15% perchloric acid at −5°C and the specimens were examined in Jeol-200CX and Jeol-400FX TEM's operating at 200 kV and 400 kV, respectively.

3. Results and discussion

It is well known that in ferrous alloys thermally induced martensite plates are generally formed with lenticular shapes at transformation temperatures close to the M_s temperature. As the transformation temperature is lowered martensite plates tend to form with long, thin shapes which can be better described as needle-like. However despite this familiar appearance of the martensite plates observed in the austenite grains grown at up to ∼1200°C, different martensite morphologies were also observed during the present work beside these plates in the samples austenitized at higher temperatures. Fig. 1 is a TEM micrograph of such martensite structures formed in Fe-21.3%Ni-0.13%C alloy sample austenized at 1350°C prior to the martensitic transformation at the M_s temperature. The austenite grain size was found around 4 mm and as shown in the figure martensite plates with different morphologies were formed as adjoined to a lenticular shaped martensitic structure. Although it is rather difficult to attain a definite geometrical shape to the observed coarse martensite morphology it may be described as similar to the butterfly-shaped martensite couples which contain two wings with a straight interface between them. It was alos observed that the wings have twinned substructure crossing their volumes. The overall morphology of the observed wings were found quite similar to those of the individually formed and dislocated coarse strain-induced martensites observed earlier in some ferrous alloys [11]. However the martensite structures shown in the figure were formed without any

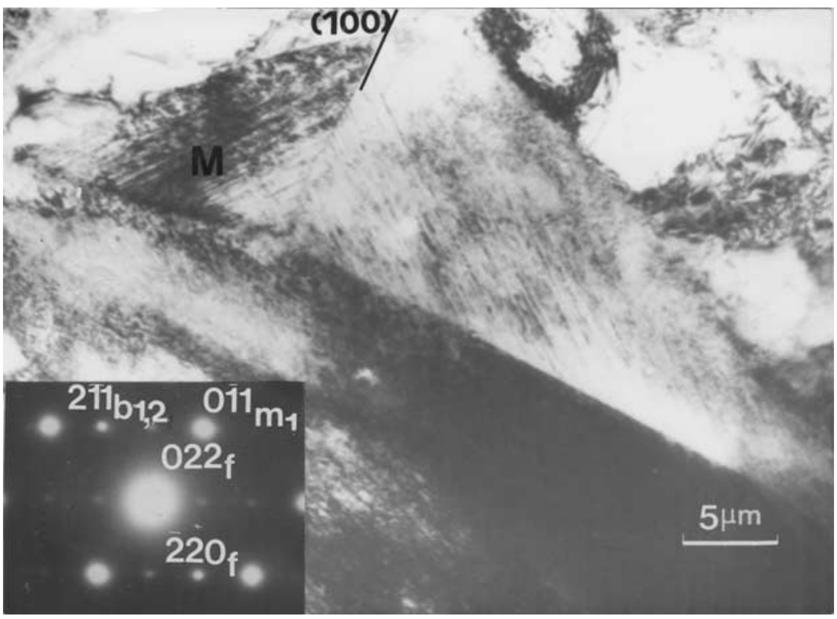

Figure 1 TEM micrograph of the coarse martensites formed with butterfly-like configurations in Fe-21.3%Ni-0.13%C alloy and superimposed electron diffraction pattern of austenite and a twinned martensite marked M (b_1, b_2 refers to martensite twins; f refers to austenite).

external deformation and contains twins. The superimposed selected-area electron diffraction pattern of the austenite matrix and a martensite (marked M) which contains superimposed [111] austenite and twin related [011] martensite zones is also given in the figure. It was revealed by the single surface analysis that the twins are formed on the {112}⟨111⟩ system of the martensite. The interface plane of the martensite wings was found as the {100} plane of austenite structure and the both wings were determined to have a Kurdjumov-Sachs type orientation relationship with the austenite matrix. The habit planes of the observed martensite structures could not be found due to the lack of any particular direction on the plates which indicates a geometrical reference direction for the habit plane determinations. Fig. 2 shows a TEM micrograph of another unfamiliar martensite morphology which was observed in Fe-40.2%Ni-0.36%C alloy sample austenized at 1350°C and transformed at the M_s temperature. The selected area difraction pattern obtained from a twinned martensite plate (marked M) which contains twin related [113] martensite zones is also given in the figure. As shown in the figure martensite plates were formed in nearly rectangular shapes with fully twinned substructures beside the other well known plate morphologies. This different morphologies were observed again only in the larger austenite volumes obtained at the austenitizing temperatures higher than 1200°C. Crystallographic analysis revealed that the martensite twins shown in Fig. 2 are formed on the {112}⟨111⟩ systems and again there is a Kurdjumov-Sachs type orientation relationship between the matrix and product crystal structures. Although there was no particular interface plane between these martensites, the {100} austenite plane was found as the plane of interface in some cases.

The previous work on the formation of strain-induced martensite in Fe-Ni-C alloys revealed that although the minimum deformation ranges necessary to start this type of martensite formation depend on the austenite grain sizes, still the strain-induced martensite formation can only be initiated after the 3–5% deformation of the matrix crystal structure [12]. During the present study the austenitic samples were deformed at different sizes below the strain-induced martensite formation range and cooled simultaneously to the M_s temperatures to observe the combined effects of deformation and cooling on the martensite formation. Fig. 3 shows a TEM micrograph of the butterfly-shaped martensite plates formed in an Fe-40.2%Ni-0.36%C alloy sample at the M_s temperature under 2% deformation. The sample was austenized at 1150°C prior to the transformation and the M_s temperature was measured as ∼10°C higher due to the simultaneously applied 2% deformation during the transformation. A lenticular shaped martensite plate is also seen in the micrograph (marked A) between the wings of the butterfly-shaped martensites which was formed under the same conditions and exhibits a triangular configuration with the wings. The selected-area diffraction pattern obtained from one of plates (marked M) is also shown in the figure which consists of [10$\bar{1}$] austenite and [100] martensite patterns and reveals the existence of a Kurdjumov-Sachs type orientation relationship. Crystallographic analysis indicated that all of the three martensites appeared with the triangular configuration have the same habit plane as {225}. As shown in the figure the lenticular shaped martensites were also formed individually (marked B) and they were found to have similar crystallographic parameters. These martensites contain fine transformation twins as it is seen in the plate marked B and they were found to be formed on the {112}⟨111⟩ system of the martensite structure. Fig. 4 shows a TEM micrograph of another butterfly-like martensite plates formed at the M_s temperature in a 2% deformed austenitic sample of Fe-17.1%Ni-0.81%C alloy which was austenized at 1350°C prior to the transformation. The larger plates formed in a butterfly-like configuration with a twinned substructure and as shown in the figure a small

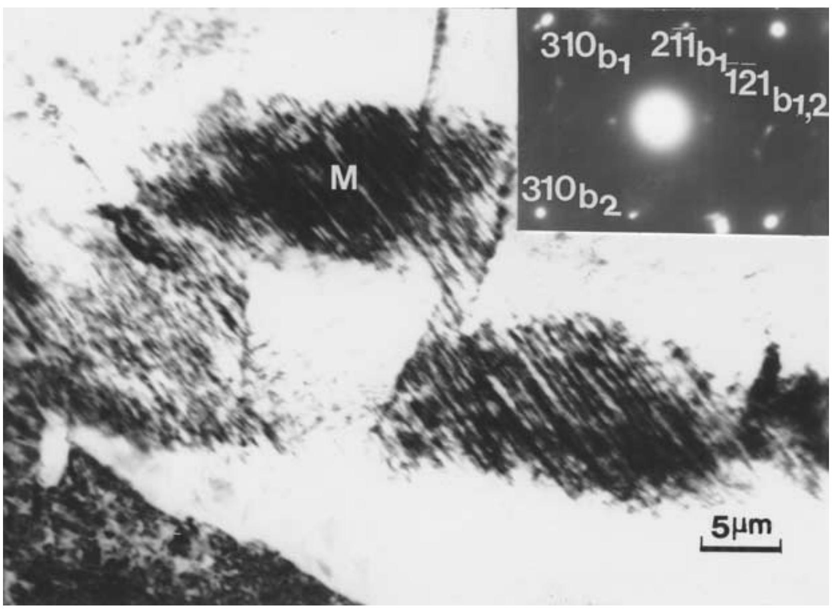

Figure 2 TEM micrograph of the coarse martensites formed with nearly rectangular shapes and twinned substructures in Fe-40.2%Ni-0.36%C alloy and electron diffraction pattern obtained from the plate marked M which shows twin reflections (b_1, b_2).

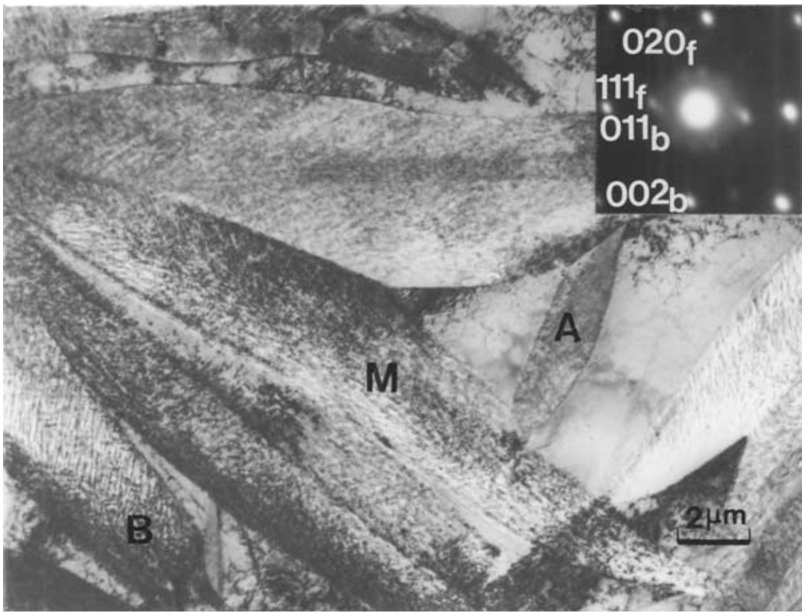

Figure 3 TEM micrograph of the martensite plates formed in Fe-40.2%Ni-0.36%C alloy. Electron diffraction pattern belongs to austenite, and martensite plate marked M.

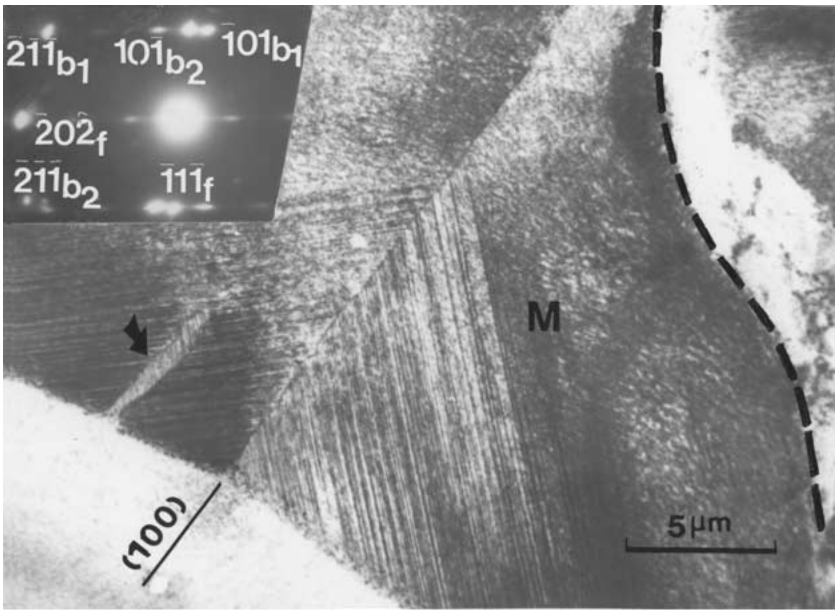

Figure 4 TEM micrograph of the coarse butterfly like martensites formed in Fe-17.1%Ni-0.81%C alloy, and electron diffraction pattern obtained from austenite and twinned martensite (marked M). The broken line indicates the non-planar austenite-martensite interface.

martensite plate was appeared (pointed by the arrow) in one of the large plates. The corresponding electron diffraction pattern is also given in the same figure which consists of $[1\bar{3}1]$ martensite, $[\bar{1}3\bar{1}]$ twin and $[\bar{1}\bar{2}1]$ austenite patterns. The austenite-martensite orientation relationship indicates that $(111)_f$ //$(011)_b$ and $[\bar{1}\bar{1}2]_f$ //$[0\bar{1}\bar{1}]_b$ (subscripts f and b refers to austenite and martensite, respectively). Electron diffraction analysis also revealed that the interface plane between the large martensite wings is the {100} plane of austenite and martensite twinning occurs on the {112}⟨111⟩ system of the martensite crystal structure. The small plate was also formed with the twinned substructure and the habit plane of this martensite was determined as the {225} plane of austenite. Although there was no possibility of finding the twinning system of the small plate with crystallographic determinations due to its size, the direction of the observed twins in this plate was observed as parallel to the twins of the larger plate on the right. Despite the similar appearance of these coarse martensite plates with those of the ones formed in an undeformed austenite structure shown in Fig. 1 the orientation relationship was found to change from Kurdjumov-Sachs to Nishiyama-Wasserman type in these samples which were simultaneously cooled and deformed to induce martensitic transformation. Maki and Wayman [13] noted in an early study that the bulk martensite with low M_s temperature has smooth and planar austenite-martensite interfaces, whereas in thin foil martensite interfaces are generally non-planar and irregular. Comparison of the Figs 1 and 4 reveals that the similar differences are also occurred in the appearance of the austenite-martensite interfaces of the coarse martensite plates formed in deformed and non-deformed bulk austenitic samples. As it is clearly shown in Fig. 4, the large martensite plate formed in the deformed bulk sample was observed to exhibit a non-planar interface with the austenite matrix.

In addition to the formation of thermally induced plate martensites with already known lenticular or needle-like shapes in the austenite grains obtained at the austenitizing temperatures up to 1200°C, martensite structures with unfamiliar morphologies were observed in the austenite matrix structures of the examined Fe-Ni-C alloys which were austenized at higher temperatures. These martensites were found with the twinned substructures and the Kurdjumov-Sachs type orientation relationships. Although there was no possibility of determining their habit planes, it can still be noted that they exhibit similar substructural features and the orientation relationship with those of the plates formed in lenticular or needle-like shapes. The formation of coarse martensite structures was examined for some steels in a previous work by Zeng *et al.* [9] and explained in terms of the austenite lattice imperfections and the more homogeneous nature of the austenite structures at higher austenitizing temperatures. Their discussion indicates that as the alloy reaches to a more homogenized state at the higher temperatures the austenite grains are formed in relatively larger volumes and their crystal structures become more perfect. Obviously the barriers in front of the martensite growth are reduced under these circumstances and much bigger martensite crystals can be formed. The austenite grain sizes also altered the M_s temperatures as expected and average difference of ∼25°C was found between the M_s temperatures of the minimum and maximum grain sizes observed in the present work. Since the coarse martensite plates were formed with internal twins, these twins were considered as transformation type lattice imperfections which shows the presence of transformation inhomogeneity as twinning on the $\{112\}\langle 111\rangle$ system of the martensite in this morphology. The TEM observations also showed the existence of dislocations with high densities in addition to the transformation twins and it was thought they might be created as a result of accommodation distortion which the coarse martensite plate experiences during its growth. The overall appearance of the twinned coarse martensite plates shown in Fig. 4 exhibits some similarities concerning especially their twinned substructures and coupled configurations with those of the so-called self accommodating martensite plates observed in some Fe-Ni-C alloys by Wang *et al.* [14]. These authors examined the martensite formation at the temperatures lower than the usual M_s temperature of the lenticular plate martensite formation and observed some new morphologies. According to their observations, the wedge-shaped thin martensites were formed in the form of coupled plates with twinned substructures and the twin planes of the both martensites were symmetrically distributed on two sides of their boundary. Present results indicate that the coarse martensites plates formed in Fe-Ni-C alloys which were austenized at relatively higher temperatures and transformed at the M_s may also exhibit similar formation characteristics as long as their coupled configurations and twinned substructures are concerned.

The observed difference in twinned substructures of the lenticular and thin martensite plates of some Fe-Ni-C alloys were discussed earlier by Patterson and Wayman [15] in terms of the latent heat released in an individual plate during the transformation. They examined the role of transformation temperature on the plate morphologies and reported that although the martensite plates are formed in elongated, thin shapes with completely twinned structures at lower temperatures, as the local temperature increases sufficiently, the second stage of the formation which does not involve twinning is attained, and the thickness to length ratio of the plates increases. However it was found in the present study that the coarse martensite plates can also be formed with fully twinned substructures and despite the their bulky appearances similar to those of partially twinned lenticular plate martensites, they exhibit similar substructures with the fully twinned thin martensite plates.

The butterfly shaped martensites were observed in both thermally and also strain-induced conditions [16, 17–19], but showed internal twinning only in the strain-induced case [20, 21]. The present results indicate the twinned substructure of butterfly-like plate martensites also in thermally induced conditions. In addition, the fully twinned coarse martensite plates with rectangular like shapes were also observed during the work and general examination of the samples showed the formation of all these coarse martensitic product structures at the early stages of the transformation just after the first burst occurred at the M_s temperature. Obviously the martensite plates appearing at later stages of transformation become much smaller than those forming earlier due to the continuous partitioning of the austenite matrix, thus the plates formed earlier should be relatively bigger. It can be concluded after these observations that the fully twinned coarse martensite plates are formed with unfamiliar shapes in the large austenite grains of the examined Fe-Ni-C alloys which have more homegeneous lattice structures due to their high austenitizing temperatures. The interface plane of the coupled martensite plates in the butterfly-like morphology was determined as the {100} plane of the austenite matrix. Okamoto *et al.* [22] examined the crystallographic features of this plane in terms of the shape strain occurred during the plate formations and considered it as a plane in which the displacement created by the macroscopic shape strains of the butterfly wings are equal. Umemoto and Tamura [18] also studied the nature of the interface plane in some Fe-Ni-C alloys and concluded that the butterfly wings are the kink form of the thin martensite plates which possess the two related variants of a definite habit plane and the observed interface plane is a bisecting plane for the two habit plane variants. According to their results on some Fe-Ni-C alloys the thin martensite plates were formed with {3 10 15} type habits and the interface plane was the {100} plane of austenite. They also examined the habit plane and morphology transitions of the martensite plates caused by the changes in the M_s temperature and reported that the butterfly wings exhibit {225} type habit while the lenticular and lath shaped plates formed at different M_s temperatures having {259} and {111} type habits, respectively. Despite the lack of the habit plane determinations for the observed coarse martensitic structures, the results obtained in the present work indicate that the thermally induced martensite plates have a unique habit plane of the {259} type with the Kurdjumov-Sachs type orientation relationship, and there is no detectable transitions in these parameters caused by the changes in the M_s or austenitizing temperatures.

Early studies on the effect of plastic deformation upon the M_s temperature of thermal martensite formation revealed that the prior plastic deformations below the mechanical stabilization limit of austenite rise the M_s in ferrous alloys [23]. Guimaraes and Shyne [24] investigated the same phenomena in different Fe-Ni-C alloys and measured the maximum increase of the M_s as 15°C at 50% deformation of austenite. It is also known that the stress required to initiate strain-induced martensite nucleations at temperatures slightly above the M_s appreciably smaller than the magnitude of the stress needed for the same transformation below M_s and a stress-assisted type transformation is occurred [6]. Fe-Ni-C samples austenized at various temperatures were simultaneously cooled and deformed in compression up to 3% to observe the combined effects of the temperature and deformation on the martensite formation. The obtained results showed the initiation of the transformation above $\sim$10−15°C of the usual M_s temperatures in 2% deformed samples. Since this deformation range was smaller than the minimum amount of deformation needed for the strain-induced martensite formation, the observed martensitic structures were considered as the stress-assisted type rather than being strain-induced in accordance with the early descriptions [6]. Despite the general formation of these martensite plates with the lenticular or thin morphologies, the coarse martensite plates in butterfly-like configurations were also observed in the samples austenized at >1200°C temperatures, as shown in Fig. 4. Although the overall shapes, and twinned substructures of these martensitic products were found similar with those of the coarse plates formed in adjoined locations to the thermally induced plate martensites, there was an obvious difference in their orientation relationship relative to the austenite matrix which was observed to change from Kurdjumov-Sachs to Nishiyama-Wasserman in the deformed samples. There were also some distinctions on the formation mechanisms of these two product structures. First of all, the plates formed in deformed conditions were observed to form with individual butterfly configurations in seperate locations, i.e. there was no reason to assume that they might be triggered with a plate martensite during the nucleation process. Secondly, small subplates with twinned substructures were observed inside these butterfly wings. The obtained results showed that the habit plane of the small martensitic structure inside the left wing in Fig. 4 is the {225} plane of austenite which is already known as the habit plane of strain-induced martensites in similar alloys [19]. Thus the small plate shown in the figure can be regarded as a strain-induced type martensitic structure formed simultaneously with the coarse butterfly wing under the combined effects of the externally applied deformation and also the internal accommodation strains created during the formation of the coarse plate. A notable difference was also found in the austenite-martensite interfaces of the butterfly-like martensite wings appeared in the deformed austenitic samples and as shown in Fig. 4, a non-planar interface was observed between the matrix and product structures. The austenite-martensite interface was examined by Maki and Wayman [12] for the thin foil samples of some Fe-Ni-(C) alloys in an early study, and contrary to the bulk martensites, thin foil martensites were observed to have generally non-planar interfaces. They explained the observed difference in terms of the lattice constraints and concluded on the basis of their observations that the martensite interface in thin foil samples is not characterized by a crystallographically invariant plane (habit plane), probably due to the different size of the constraints in these product structures. The non-planar nature of the coarse martensite wings observed during the present work may also indicate the similar influence of the lattice constraints. Since the austenite-martensite interfaces were always found smooth and planar in thermally induced martensites, it can be stated that the interface of the coarse martensite plates formed in deformed austenitic samples may differ and exhibit a non-planar nature due to the different sizes of the lattice constraints and thus the austenite-martensite interface of this plates is not characterized by their habit planes.

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References

1. G. THOMAS, *Metall. Trans.* **2** (1970) 2373.
2. J. NUTTING, *J. Iron Steel Inst.* **6** (1969) 872.
3. P. L. MANGONON and G. THOMAS, *Metall. Trans.* **1** (1970) 1577.
4. R. G. DAVIES and C. L. MAGEE, *ibid.* **2** (1971) 1939.
5. G. KRAUSS and A. R. MARDER, *ibid.* **2** (1971) 2343.
6. P. G. McDOUGALL and C. M. WAYMAN, in Proceedings of Conference on Martensite, edited by G. B. Olson and W. S. Owen (ASM, Ohio, 1992) p. 59.
7. P. VISVESVARAN, *Metall. And Mater. Trans. A* **27A** (1993) 973.
8. R. D. JAMES and K. F. HANE, *Acta Metall.* **48** (2000) 197.
9. D. C. ZENG, Y. H. TAN, G. W. QIAO and Y. C. CHUANG, *Z. Metallkunde* **85** (1994) 203.
10. A. S. SASTRI and D. R. F. WEST, *J. Iron Steel Inst.* **203** (1965) 138.
11. Z. NISHIYAMA, "Martensitic Transformation" (Academic Press, New York, 1978).
12. T. N. DURLU, *J. Mater. Sci. Lett.* **12** (1993) 1378.
13. T. MAKI and C. M. WAYMAN, *Acta Metall.* **25** (1977) 681.
14. S. WANG, Y. H. WEN and X. ZHANG, in Proceedings of the International Conference on Martensitic Transformations, 1992 (ICOMAT' 92), edited by C. M. Wayman and J. Perkins (Monterey Institute for Advanced Studies, Monterey, California, 1993) p. 281.
15. R. L. PATTERSON and C. M. WAYMAN, *Acta Metall.* **14** (1966) 347.

16. I. TAMURA, T. MAKI and H. HATO, *J. Japan Inst. Metals* **33** (1969) 1376.
17. T. N. DURLU, *J. Mater. Sci.* **31** (1996) 2585.
18. M. UMEMOTO and I. TAMURA, in Proceedings of the International Conference on Martensitic Transformations, 1979 (ICOMAT' 79), edited by W. S. Owen (MIT Press, Cambridge, Massachusetts, 1979) p. 53.
19. X. M. ZHANG, E. GAUTIER and A. SIMON, *Acta Metall.* **37** (1989) 487.
20. E. GAUTIER, J. ZHANG, X. ZHANG and T. R. HU, in Proceedings of the International Conference on Solid→Solid Phase Transformations, July 17–22, 1994, edited by W. C. Johnson, J. M. Howe, D. E. Laughin and W. A. Soffa (The Minerals, Metals and Materials Society, Pennsylvania, 1994) p. 847.
21. T. N. DURLU, *Acta Metall.* **26** (1978) 1855.
22. H. OKAMOTO, M. OKA and I. TAMURA, *Trans. Japan Inst. Metals* **19** (1978) 674.
23. T. MAKI, M. SHIMOOKA, M. UMEMOTO and I. TAMURA, *ibid.* **13** (1972) 400.
24. J. R. C. GUIMARAES and J. C. SHYNE, *Metall. Trans.* **2** (1971) 2063.

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