Role of Carbide in the Recovery Process of 0.2% Carbon Lath Martensitic Structure*

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The changes in carbide morphology and matrix microstructure that occur during 973 K annealing have been investigated for a low carbon lath martensitic steel by optical and transmission electron microscopy. For optical metallographic observation, striking changes in the lath martensitic structure were not found even in long-time annealed specimens because carbides preferentially precipitate on block and lath boundaries to maintain the structural character of lath martensite. The morphology of carbides and matrix microstructure, however, are gradually changed during the annealing.

Stick type carbides, which were formed along lath boundaries in the early stage of annealing, play an important role in suppressing the recovery of dislocations within the lathes. Dislocation density within the matrix pronouncedly decreases when the morphology of lath boundary carbides changes from stick to oval or sphere, leading to an unusual change in tensile properties. It was also confirmed that the deformation to a pre-annealed specimen containing the stick type lath boundary carbides does not greatly affects on recovery behavior during the further annealing. The result also indicates the fact that the growth of lath boundary carbides controls the recovery of lath martensite matrix.

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I. Introduction

Low carbon lath martensitic steels are characterized by the stable structure against recrystallization even though they contain a high density of dislocations⁽¹⁾⁻⁽³⁾ as well as cold-rolled ferrite. The recrystallization is difficult to occur even at high temperatures just under the A₁ point⁽⁴⁾ and this is the reason why excellent ductility can be obtained without expense of strength after the high temperature annealing. Besides, an embrittling behavior by stress relief annealing has been found in low carbon lath martensitic steels served for pressure vessel⁽⁵⁾⁻⁽⁷⁾. The embrittlement is also associated with the recovering process of the lath martensite matrix⁽⁸⁾.

For the reason of retarded recrystallization in low carbon lath martensitic steels, it is believed that carbides obstruct⁽⁹⁾⁽¹⁰⁾ the movement of dislocations. In addition to such a retarding effect of carbides, it is also suggested that the difference in dislocation arrangement⁽¹¹⁾ between martensite and deformed ferrite results in the difficulty of recrystallization of lath martensite. Details for the recovery and recrystallization processes of lath martensite, however, have not been cleared yet.

In this study, the changes in carbide morphology and dispersion during high temperature annealing were in-

vestigated for a 0.2% C low alloy (SNCM 420) steel in connection with the microstructural changes of the lath martensite matrix, in order to make the recrystallization retardation mechanism clear.

II. Experimental Procedure

1. Materials and heat treatment

A commercial 0.2% C low alloy (SNCM 420) steel with the chemical composition shown in Table 1 was used. A 50-mm diameter rod material was rolled to 10 mm at 1473 K and then cold rolled to 1 mm thick plates at room temperature. The steel plates were solution treated at a high temperature of 1573 K for 1.8 ks in order to make carbides perfectly dissolve into the matrix. Full lath martensitic structure was obtained by water quenching from the solution treatment temperature. Since a reversion temperature from the lath martensite to austenite (A_{c1} point) was 1006 K for the steel used, annealing was performed at 973 K below the A_{c1} point. For the annealing treatment, salt baths were used to enable rapid heating to 973 K. Isothermally annealed specimens were water quenched from the annealing temperature and then served to several examinations.

Table 1 Chemical composition of the steel used. (mass %)

Alloy	C	Si	Mn	Ni	Cr	Mo
SNCM420	0.21	0.24	0.57	1.59	0.50	0.16

others: P < 0.03, S < 0.03

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2. Examinations

Microstructures were observed by means of optical and transmission electron microscopy (TEM). Specimens were prepared by 2% nital etching for optical observation and by jet electropolishing method with an ethyl alcohol solution of 20% sulfuric acid for TEM observation.

Tensile tests were carried out for platelet specimens of 6 mm width, 0.8 mm thickness and 35 mm gauge length with an Instron type testing machine at the cross head speed of 0.02 mm/s. Displacement was detected with a displacement meter of 20 mm gauge length attached to a tensile test piece. Ductility was estimated by total fracture elongation. Hardness was represented by the average of more than five measurements in 9.8 N load Vickers hardness.

III. Experimental Results

1. Lath martensitic structure of annealed specimens

The steel quenched from 1573 K (as-quenched specimen) has large pre-austenite grains of about $200\text{ }\mu\text{m}$ and lath martensitic structure characterized by laths with high density of dislocations, as shown in Fig. 1(a) and 1(b) respectively. The laths are the minimum unit in lath martensitic structure and the morphology is thought to be strip-like⁽¹²⁾⁽¹³⁾. The thickness and width of laths were about

$0.2\text{ }\mu\text{m}$ and $1\text{ }\mu\text{m}$, respectively. The length of a lath is as long as thirty times⁽¹²⁾⁽¹³⁾ of the thickness. Needle-like structure in Fig. 1(a) is the block, which is a complex of the same variant laths in crystallography. The block size is several microns in the cross section. Such a microstructure is characteristic of the low carbon lath martensite containing about 0.2% C and has already been classified as “the lath martensite composed of undeveloped blocks”⁽¹¹⁾.

Figure 2 shows optical micrographs of specimens annealed at 973 K for (a) 0.1 ks and (b) 10 ks. The annealing temperature, 973 K, is high enough for cold-rolled ferritic steels to recrystallize. However, the micrograph (b) shows almost the same microstructure as micrograph (a), despite a large difference in the annealing time. No area where recrystallization takes place was observed in both micrographs. TEM micrographs in Fig. 3 indicate the microstructures of (a) as quenched and (b) 10 ks annealed specimens where a couple of blocks impinge each other. In the photograph (a), block boundary lines are easily found out because of the discontinuity of lath structure. Comparison between photographs (a) and (b) reveals the fact that carbides preferentially precipitate on the boundary between blocks and laths, and the growth of block boundary carbides is faster than that of the lath boundary carbides. The microstructural characteristic of the as-quenched specimen is maintained by the carbides on block and lath boundaries even after 10 ks annealing. The reason why optical microstructure does not change

Fig. 1 Microstructures showing lath martensite in the as-quenched 0.2% C steel. (a) optical and (b) transmission electron micrograph.

Fig. 2 Optical micrographs of 0.2% C lath martensite annealed at 973 K for (a) 0.1 ks and (b) 10 ks.

Fig. 3 Transmission electron micrographs showing the microstructure around block boundaries. (a) as-quenched and (b) 973 K-10 ks annealed.

even after the long time annealing is attributed to such a carbides precipitation. However, the substructural changes within the matrix such as the changes in the dislocation density and arrangement, gradually proceed during annealing.

2. Changes in the carbide morphology and substructure

Figure 4 shows the changes of 0.2% proof stress, tensile strength and elongation during 973 K annealing. The changes in mechanical properties are classified into three stages: the earliest stage showing an abrupt change, the subsequent flat stage to 0.3 ks and the final stage with slow change after 0.3 ks. The earliest abrupt change is mainly due to a substantial decrease in dislocation density⁽¹⁴⁾. Although carbides can precipitate in a short time annealing of 0.28 s at 963 K⁽¹⁰⁾, dislocation pinning by the carbides is hard to occur because the stored strain energy is so high that the movement of dislocation might not be stopped by the carbides⁽¹⁵⁾. The subsequent change of mechanical properties, however, must be closely associated with the growth of carbides and the recovery behavior of the matrix. Especially, we should pay attention to the unusual change between 0.3 and 1 ks. It seems to suggest that a significant microstructural change in the matrix takes place around the annealing condition.

TEM micrographs (a) and (b) of Fig. 5 show the microstructures of the steel annealed 30 s at 973 K. The

Fig. 4 Changes in mechanical properties by 973 K annealing in the 0.2% C lath martensitic steel.

Fig. 5 Transmission electron micrographs of 0.2% C lath martensite annealed at 973 K for 30 s. Showing (a) carbide precipitates and (b) dislocations within lathes.

photograph (a) seems to be a microstructure observed from the thickness direction of laths. In the micrograph, two kinds of precipitates are observed: One is fine carbides within the lath matrix and the other is stick-like carbides on lath boundaries. Dislocations exist within the matrix, surrounded by the stick-like lath boundary carbides, as shown in the photograph (b). The subsequent changes in substructures within the matrix are represented in Fig. 6. In a 0.3 ks annealed specimen (photograph (a)), a large number of dislocations were observed within the matrix yet. Especially, it should be noted that the dislocation density is substantially higher in the area where the stick-like carbides densely retain

than in the area where lath boundary carbides change the morphology to oval or sphere. In a 1 ks annealed specimen (photograph (b)), the mean dislocation density is found to considerably decrease although there are some local differences in the dislocation density. The unusual change in mechanical properties has been recognized between 0.3 and 1 ks, as shown in Fig. 4. The experimental result must reflect the significant decrease in dislocation density. After 1 ks, recovery of the matrix gradually proceeds with annealing time and subgrains are formed in places. A 10-ks annealed specimen is composed mostly of such subgrains as shown in photograph (c). The subgrain boundaries are pinned by coarsened

Fig. 6 Transmission electron micrographs of 0.2% C lath martensite annealed at 973 K for (a) 0.3 ks, (b) 1 ks and (c) 10 ks.

Fig. 7 Change in the morphology of lath boundary carbides. Annealed at 973 K for (a) 0.3 ks, (b) 1 ks and (c) 10 ks.

Fig. 8 Changes in the morphology and size of lath boundary carbides observed in the annealed 0.2% C lath martensite. The “diameter” in the figure represents the carbide size in the perpendicular direction to lath boundaries.

carbides on block or lath boundaries.

Considering the role of carbides on the recovery process, lath boundary carbides can play an important role throughout the recovery process, while block boundary carbides contribute to the suppression of the recrystallization through the subgrain boundary pinning only in the latter annealing stage. Therefore, the role of lath boundary carbides is more important than that of block boundary carbides, because low alloy carbon steels such as the steel used in this investigation are usually provided for the use in the high strength level of 0.7–0.9 GPa, which is obtained in the early recovery stage for 973 K annealing. The typical morphologies of lath boundary carbides are shown in Fig. 7. The morphology changes from stick-like to oval or sphere, as shown in the photograph (a), (b) and (c). Figure 8 summarizes the changes in the morphology and size of lath boundary carbides as a function of annealing time. The carbide size is represented by the diameter in the perpendicular direction to lath boundary for stick-like and oval carbides. The carbide size is under 70 nm before 0.3 ks when the stick-like carbides retains, and gradually increases as the stick-like carbides are split to form the oval or sphere carbides. The annealing time from 0.3 ks to 1 ks, where a significant recovery proceeds within the matrix, just corresponds to the stage where the stick-like carbides disappear. This result indicates that the recovery of dislocation is accelerated when the stick-like carbides have been broken, because the dislocation movement across lath boundaries becomes easier.

IV. Discussion

1. Precipitation of lath boundary carbides and change in the morphology

A block is thought to be a complex of many laths as schematically illustrated in Fig. 9. The morphology of lath boundary carbides which precipitate in the early annealing stage was not plate-like but stick-like so that

Fig. 9 Illustration of carbide precipitation along lath boundaries in the 0.2% C lath martensite.

carbides probably precipitate not on lath boundary faces but on boundary lines among three laths, as shown in the illustration. We presume that sphere carbides, which were observed in the latter stage of 973 K annealing, precipitate by the volume of $V \mu\text{m}^3$ within the unit matrix of $1 \mu\text{m}^3$. When the diameter of the sphere carbides is $d \mu\text{m}$, the total number of carbides N within the unit matrix is given by formula (1) because the volume of a carbide is $\pi d^3/6$.

$$N = 6V / (\pi d^3) \quad (1)$$

The total lath boundary line length within the $1 \mu\text{m}^3$ matrix is $10 \mu\text{m}$, so that the carbide spacing $y \mu\text{m}$ on lath boundary lines is also given by formula (2).

$$y = 10 / N = 5\pi d^3 / (3V) \quad (2)$$

Carbides which precipitate in low alloy carbon steels like the steel used is thought to be cementite⁽⁸⁾. Therefore, under no consideration of the influences of alloying elements, the mass fraction of cementite is estimated at 2.85 mass% for the 0.21 mass% C steel used in this investigation. In the volume fraction, it corresponds to 2.90 vol% considering the density of cementite; $7.68 \times 10^3 \text{ kg/m}^3$ ⁽¹⁶⁾. Replacing the value V of formula (2) by $0.029 \mu\text{m}^3$, the carbides spacing y is approximately given by formula (3).

$$y = 180d^3 [\mu\text{m}] \quad (3)$$

When the diameter of carbides d is larger than the spacing y , carbides connect each other to form stick-like precipitates and, in the opposite case, carbides align separately along lath boundary lines. The critical carbide size is given when y is equal to d and the value is about 75 nm. In fact, lath boundary carbides of the size have been observed in the annealing time between 0.3 and 1 ks where the carbide morphology changes from stick-like

to oval or sphere, as mentioned in Fig. 8. The fact demonstrates that the carbides precipitation model illustrated in Fig. 9 is reasonable for low carbon lath martensitic steels.

Discussing the roles of carbides on the recovery process of dislocations, the carbides spacing is probably one of the important factors but the morphology of carbides seems also to affect the motion of dislocations. On the interaction between dislocations and stick-like lath boundary carbides, dislocations within a lath are easy to move toward the direction parallel to the stick-like carbides but difficult to move toward the vertical direction because the carbides act as obstacles. Especially in a high temperature range, it has been cleared in an investigation for high temperature deformation⁽¹⁷⁾ that dislocations go through precipitates not by the Orowan mechanism but by climbing up mechanism. The shape of stick-like carbides is convenient for suppressing the climb-up of dislocations. Indeed, in the areas where stick-like carbides are densely retained, recovery of the matrix tends to be retarded. Furthermore, a significant decrease in dislocation density occurs in the annealing time between 0.3 and 1 ks where stick-like carbides have been broken. Fine carbides in the matrix have gradually disappeared as lath boundary carbides grow up, but no unusual changes were observed around the annealing time. These results suggest that stick-like carbides play an important role in retarding the recovery of the lath martensite matrix. In the latter annealing stage, the formation of subgrains proceeds as lath boundary carbides grow and this results in a slow decrease in strength and increase in elongation. The size of subgrains is dependent on the dispersion of carbides: The subgrain size is almost the same as or slightly larger than the mean carbides spacing⁽¹⁸⁾⁽¹⁹⁾.

As the results, recovery process of lath martensite matrix is closely associated with the changes in morphology and dispersion of lath boundary carbides. Especially it should be noted that a relatively high density of dislocations has been maintained as long as stick-like carbides remain on lath boundaries and this leads to a high strength of low carbon lath martensitic steels annealed at high temperatures.

2. Influence of carbides on recovery behavior of dislocations induced by cold working

As a reason for the difficulty of recovery and recrystallization in lath martensitic steels, it is also suggested⁽¹¹⁾ that the nature of dislocations induced by martensitic transformation differs from that of dislocations induced by deformation, in addition to the dislocation pinning effect of carbides. When lath martensitic steels were subjected to cold working prior to annealing, even the morphology and dispersion of carbides are changed by the cold working⁽²⁰⁾. In this study, 20% cold working was performed after 973 K–0.3 ks annealing, as shown in Fig. 10, in order not to affect the carbide precipitation behavior. This treatment means that dislocation arrangement was changed after the formation of stick-like lath boundary carbides. The deformation rate

Fig. 10 Effect of cold working on the softening behavior of 0.2% C lath martensite during 973 K annealing.

is thought to be enough to rearrange dislocations within the matrix because the cold-worked specimen is hardened by about 40 in Vickers hardness. The hardness increased, however, abruptly recovers to the level of undeformed specimen in the subsequent short time annealing and then follows almost the same softening curve to 10 ks. There is no outstanding influence of deformation on the recovery behavior, even though local strains which are necessary for recrystallization must be induced by the cold rolling. The result demonstrates that recovery process of low carbon lath martensite mainly depends on the process of carbide growth rather than dislocation arrangement itself. It is also true that deformation to as-quenched lath martensite promotes recrystallization during the following annealing⁽²¹⁾⁽²²⁾. But the reason is probably attributed to the fact that the deformation results in destruction of lath structure itself and this leads to suppression of the formation of stick-like lath boundary carbides⁽²⁰⁾. Thus, it is concluded that the recovery and recrystallization processes of low carbon lath martensitic steels are controlled through the growth of lath boundary carbides, except an abrupt recovery process in the early annealing stage.

V. Conclusion

Relation between the change in carbide morphology and recovery process in a 0.2% carbon lath martensitic steel has been investigated after high temperature annealing at 973 K. The results obtained are as follows.

(1) Since carbides preferentially precipitate on lath and block boundaries to maintain the microstructural characteristic of lath martensite, no conspicuous changes are found in optical microscopic observation even after long time annealing at the high temperature.

(2) The recovery and recrystallization processes of the low carbon lath martensite are controlled through the growth of lath boundary carbides, except an abrupt recovery process in the early annealing stage. Especially, recovery of the matrix tends to be retarded as long as

stick-like carbides remains on lath boundaries.

(3) The stick-like lath boundary carbides change the morphology to oval or sphere with annealing time. A significant decrease in dislocation density occurs when the stick-like carbides has been broken, resulting in an unusual change in 0.2% proof stress, tensile strength and elongation.

(4) With the growth of lath boundary carbides, the matrix gradually recovers. After long time annealing, subgrain boundaries pinned by coarsened lath boundary carbides and block boundary carbides are formed.

(5) 20% cold working to a specimen with stick-like lath boundary carbides, which were pre-induced by a short time annealing at 973 K, does not greatly affect on recovery behavior of the matrix during the further annealing.

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