Recrystallization of Lath Martensite with Bulge Nucleation and Growth Mechanism

T. TSUCHIYAMA, Y. MIYAMOTO and S. TAKAKI

Department of Materials Science and Engineering, Kyushu University, Hakozaki, Higashi-ku, Fukuoka 812-8581 Japan. E-mail: toshi@zaiko.kyushu-u.ac.jp 1) Corporate Research & Development Center, NSK Ltd., Kugenuma Shinmei, Fujisawa 251-8501 Japan.

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The recrystallization behavior of lath martensite during tempering was investigated in high-chromium martensitic steels by means of hardness testing, optical and transmission electron microscopy. The role of carbide particles on the recrystallization was also discussed in terms of the grain boundary pinning effect. The hardness of tempered specimens was plotted as a function of the tempering parameter, T log t, for a low-carbon steel (Fe–9Cr–0.1C mass%) and an ultra-low carbon steel (Fe–9Cr–1Ni–0.006C mass%). The low-carbon steel exhibited gradual softening with recovery but did not undergo recrystallization. However, the ultra-low carbon steel suffered abrupt softening owing to the discontinuous recrystallization of lath martensite. Microstructural observations in the ultra-low carbon steel indicated that the recrystallization of lath martensite occurs with the ‘bulge nucleation and growth (BNG) mechanism’. The possibility of recrystallization via this mechanism depends upon both the spacing of carbide particles on grain boundaries and the dislocation density of martensite. An energetic analysis on the formation of a recrystallized grain revealed the critical carbide spacing minimum required for the occurrence of recrystallization as a function of dislocation density. In the case of the low-carbon steel, carbide precipitates on grain boundaries with spacing smaller than the critical value, thus suppressing recrystallization.

KEY WORDS: ultra-low carbon steel; lath martensite; recrystallization; grain boundary; carbide; dislocation density; bulge nucleation and growth mechanism.

1. Introduction

Upon tempering, lath martensite in low-carbon steels undergoes recovery,*1 but never recrystallizes*1–4 despite of its high dislocation density of 10^{14–15}/m^2 which corresponds to that of heavily cold-worked materials.5 Therefore tempering of martensite can be effective for improving ductility and toughness without significant reductions in strength. However, it is reported that ultra-low carbon martensitic steels soften markedly on tempering with recrystallization. For example, Galibois and Dube6) examined the relation between softening behavior and microstructural change of martensite during tempering at several temperatures using some ultra-low carbon steel wire (Fe–0.021,
0.03C mass%), and found that equiaxed recrystallized grains are formed within the martensitic structure after tempering at around 600°C.

These results suggest that carbide particles suppress recrystallization during tempering in carbon-bearing martensite although lath martensite can be susceptible to recrystallization. Some previous work using low-carbon steels revealed that carbides precipitated during tempering pin the grain boundaries3) or dislocations4) in the martensitic structure. However, the process of nucleation and growth during the recrystallization of lath martensite has never been discussed. Thus the detailed role of carbide particles is unclear with respect to the suppression of recrystallization.

In this study, the recrystallization mechanism of lath martensite was clarified by means of hardness testing, optical and transmission electron microscopy. The role of carbide particles on the recrystallization was also discussed in terms of the grain boundary pinning effect. The hardness of tempered specimens was plotted as a function of the tempering parameter, T log t, for a low-carbon steel (Fe–9Cr–0.1C mass%) and an ultra-low carbon steel (Fe–9Cr–1Ni–0.006C mass%). The low-carbon steel exhibited gradual softening with recovery but did not undergo recrystallization. However, the ultra-low carbon steel suffered abrupt softening owing to the discontinuous recrystallization of lath martensite. Microstructural observations in the ultra-low carbon steel indicated that the recrystallization of lath martensite occurs with the ‘bulge nucleation and growth (BNG) mechanism’. The possibility of recrystallization via this mechanism depends upon both the spacing of carbide particles on grain boundaries and the dislocation density of martensite. An energetic analysis on the formation of a recrystallized grain revealed the critical carbide spacing minimum required for the occurrence of recrystallization as a function of dislocation density. In the case of the low-carbon steel, carbide precipitates on grain boundaries with spacing smaller than the critical value, thus suppressing recrystallization.

KEY WORDS: ultra-low carbon steel; lath martensite; recrystallization; grain boundary; carbide; dislocation density; bulge nucleation and growth mechanism.

2. Experimental Procedures

The chemical compositions of the steels used are listed in Table 1. The ultra-low carbon steel (ULC steel) was designed to satisfy the following conditions: (1) the M_s temperature is below 800 K so that a fully lath-martensitic structure can easily be obtained at low cooling rates, and (2) the reversion temperature (A_1) is above 1000 K so as not

*1 In this paper, the term ‘recrystallization’ has been defined as a discontinuous process, that is, new dislocation-free grains are formed within a recovered structure and they grow consuming old grains. Continuous processes such as the formation of subgrain structure or the increase in misorientation of grain boundaries have been referred to as ‘recovery’.
to form austenite during tempering even at a temperature which is high enough for the diffusion of iron atoms. The addition of chromium is effective for depressing $M_s$ temperature without depressing $A_1$ temperature. The low-carbon steel (LC steel) was also used to study the effect of carbide particles. These specimens were solution-treated at 1273 K for 1.8 ks, followed by water quenching to obtain a fully martensitic structure. Tempering was generally carried out at around 973 K\(^*2\) for various times in an argon atmosphere. These tempered specimens were subjected to Vickers hardness testing and microstructural observations by means of optical and transmission electron microscopy. In order to find the nucleation site in the recrystallization, in-situ observation was also carried out for the ULC steel as follows; the microstructure of the as-quenched specimen was observed at the same area before and after tempering, which was performed in a hydrogen atmosphere to avoid oxidization of the specimen surface.

The dislocation density of the ULC steel was estimated from hardness. The hardness of this material, $HV$, may be roughly expressed by Eq. (1) as a function of dislocation density, $\rho$, because the strength of martensite without carbon should be mainly due to dislocation hardening.\(^7\)

\[ HV = H_{V_o} + \alpha G b \sqrt{\rho} \]  

where $H_{V_o}$ is the hardness of dislocation free material, $\alpha$ is a constant, $G$ is the shear modulus, and $b$ is the burgers vector. We defined the parameter $P$ which is a function of $HV$: 

\[ P = (HV - H_{V_{min}})/(H_{V_{max}} - H_{V_{min}}) \]  

where $H_{V_{max}}$ and $H_{V_{min}}$ are the hardness of as-quenched material and that of fully recrystallized material, respectively. By using the relation of Eq. (1), $P$ can be rewritten as:

\[ P = \left( \frac{\sqrt{HV_{o} + \alpha Gb \rho}}{\sqrt{H_{V_{max}} + \alpha Gb \rho}} \right) \left( \frac{\sqrt{H_{V_{min}}}}{\sqrt{\rho}} \right) \]  

where $\rho_{max}$ is the dislocation density of the as-quenched specimen and $\rho_{min}$ is that of fully recrystallized material. After all, the dislocation density of the material with hardness $HV$ is given by

\[ \rho = \left( \frac{\sqrt{\rho_{max} - \sqrt{\rho_{min}}} + \sqrt{\rho_{min}}} {\sqrt{\rho_{max}}} \right)^2 \]  

where the values of $\rho_{max}$ and $\rho_{min}$ used in this study are $10^{15}/m^2$ and $10^{12}/m^2$ respectively.

3. Results and Discussion

3.1. Softening Behavior and Microstructure Development during Tempering

Figure 1 shows optical micrographs of the LC (a) and ULC (b) steels quenched from the solution treatment temperature. Both specimens have a typical lath martensitic structure composed of martensite-packet and -block structures although the etching depth varies because of their corrosion resistance. The hardness of this specimen was HV394 in the LC steel and HV270 in the ULC steel ($HV_{max}$). These specimens were subjected to tempering at various temperatures for various times, and then Vickers hardness were measured as a function of the tempering parameter, $T(\log t + 20)$, (Fig. 2) where $T$ is absolute temperature and $t$ is tempering time in hours.\(^10\) The hardness of the LC steel gradually reduces as the martensite recovers, however, it did not recrystallize as is the case of plain-carbon steels.\(^10\) In the ULC steel, hardness drops abruptly when the parameter became about 19 300 (for example, 973 K–2.7 ks), and then settles at a constant value of HV120 ($HV_{min}$). Such softening behavior is similar to the case of

\[ P = \left( \frac{\sqrt{H_{V_{o}}} - \sqrt{H_{V_{min}}} + \sqrt{H_{V_{min}}}} {\sqrt{H_{V_{max}}} - \sqrt{H_{V_{min}}}} \right)^2 \]  

Table 1. Chemical compositions of steels used (mass\%)

<table>
<thead>
<tr>
<th></th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>P</th>
<th>S</th>
<th>Cr</th>
<th>Ni</th>
<th>N</th>
</tr>
</thead>
<tbody>
<tr>
<td>Low-carbon (LC)</td>
<td>0.11</td>
<td>0.06</td>
<td>0.40</td>
<td>0.012</td>
<td>0.012</td>
<td>8.95</td>
<td>–</td>
<td>0.0077</td>
</tr>
<tr>
<td>Ultra-low carbon (ULC)</td>
<td>0.006</td>
<td>0.02</td>
<td>0.21</td>
<td>0.005</td>
<td>0.012</td>
<td>8.98</td>
<td>0.99</td>
<td>0.0090</td>
</tr>
</tbody>
</table>

\(^*2\) The equilibrium carbide at around 973 K is $M_23C_6$ type. The volume fraction of the carbide is roughly estimated at 0.1 vol% in the ULC steel and 2.2 vol% in the LC steel, respectively.\(^9\)
general recrystallization in heavily deformed steels.

Figure 3 represents microstructural change on the abrupt softening of ULC steel. Before the softening starts (a), the lath-martensitic structure is fully retained and block structures can still be observed within grains. The prior austenite grain boundaries and packet boundaries are characterized by their linear geometry. However on softening (b), some grains with wavy grain boundaries appear in the microstructure. It should be noted that martensite block structure cannot be seen within these grains. This indicates that they are recrystallized ferrite grains which have grown by consuming martensite during tempering. The recrystallized grains gradually multiply and coincident with the hardness reaching a stable minimum (c), the martensitic structure disappears to be replaced completely by recrystallized grains.

In order to clarify where the recrystallized grains nucleated, the microstructure was observed before and after recrystallization at the same area in a specimen (in-situ observation). Figure 4 shows an example of the results of in-situ observations. The upper optical micrograph and trace of grain boundaries are of an as-quenched specimen and the lower ones are of the same specimen after tempering at 1023 K for 3.6 ks, during which time partial recrystallization has occurred on the specimen surface. It was found that the packet boundary shown by the hatched line in the schematic illustrations migrated upward during tempering and the martensite block structure has been removed in the region swept by the boundary. The same phenomenon was observed for prior austenite grain boundaries but not for block and lath boundaries. The observation may be explained by differences in boundary mobility. Illustrated in Fig. 5 are a set of transmission electron micrographs which represent the process of grain boundary migration. Figure 5(a), which was taken just after the onset of the migration, shows a dislocation free region behind the migrating grain boundary. This corresponds to the nucleus of the recrystallizing grain. It can also be seen that the grain boundary is pinned by carbide particles and bulges between them. It is easy to imagine that, if carbide particles densely decorate the grain boundary, this would restrict the bulging process and hence obstruct grain boundary migration. This seems to be the reason why carbon-bearing steels do not recrystallize during tempering. Once the grain boundary unpins, as shown in the Fig. 5(b), the recrystallized grain grows rapidly, consuming the martensite which contains a high density of dislocations whose strain energy drives the migration of grain boundary.

3.2. Role of Carbide Particles in the Bulge Nucleation and Growth (BNG) Mechanism

Similar recrystallization process due to bulge nucleation was reported in deformed aluminum by Beck and Sperry, and the energetics of the process have been analyzed by Bailey and Hirsh. The mechanism is termed the ‘strain-induced boundary migration (SIBM)’. The model explains the possibility of bulge nucleation in recrystallization of deformed materials, but it does not specify how the bulging grain boundary escapes from the pinning and causes the growth of recrystallized grain. In this study, the condition for the bulging grain boundary to escape from the pinning
by carbide particles was expressed as a function of the critical carbide spacing. This condition was regarded as the requirement for the occurrence of recrystallization. We describe this model as the ‘bulge nucleation and growth (BNG) mechanism’.

The BNG mechanism can be understood by the schematic illustrations in Fig. 6. Carbide particles are assumed to exist on a grain boundary with the spacing, \( \lambda \) (a). If the grain boundary bulges to one side, the strain energy, \( \Delta G_1 \), will be released because dislocations are removed by sweeping with the grain boundary, while the energy due to grain boundary, \( \Delta G_2 \), will be increased owing to the expansion.
sion of the grain boundary. Therefore the grain boundary can migrate under the condition of $\Delta G_1 > \Delta G_2$ with a decrease in the total free energy of the system. Letting the shape of bulging grain boundary be a cylindrical cap with a depth of unit length, when the grain boundary bulges with the angle $q(b)$, the value of $\Delta G_1$ and $\Delta G_2$ are expressed as:

$$\Delta G_1 = \Delta V / \rho E_d$$
$$\Delta G_2 = \Delta S \sigma$$

$$= \lambda^2 \rho E_d (\theta - \sin \theta \cos \theta) / 4 \sin^2 \theta$$

where $\Delta V$ is the volume of the swept region, $\rho$ is the dislocation density of the tempered martensite, $E_d$ is the strain energy of a dislocation per unit length, $\Delta S$ is the change in area of the grain boundary, and $\sigma$ is the interfacial energy per unit area of the grain boundary. For bulging to occur, we know that $\Delta G_1 > \Delta G_2$, therefore the condition for grain boundary migration, given as a function of angle $\theta$ is as follows:

$$\lambda > \sigma (\theta / 4 \sin \theta - 1) / (\rho E_d (\theta - \sin \theta \cos \theta) / 4 \sin^2 \theta)$$

If the angle $\theta$ exceeds $\pi / 2$, adjacent cylindrical caps coalesce, hence the bulging grain boundary will continue to advance by escaping the pinning of carbide particles (c) (d). Therefore substitution of $\pi / 2$ for $\theta$ gives the critical carbide spacing required for the occurrence of recrystallization (or complete suppression of recrystallization) in lath martensite as:

$$\lambda_c = 4 \sigma (\pi - 2) / \rho E_d$$

When the carbon content is high enough to produce carbide precipitates on the grain boundaries with the spacing smaller than $\lambda_c$, the recrystallization should be completely suppressed.

3.3. Possibility of Recrystallization in Martensitic Steels

Figure 7 shows the recrystallization area map for carbon-bearing lath martensite. The diagonal line bisecting this map is the critical carbide spacing given by the Eq. (8). The values of $\sigma$ and $E_d$ used here are 0.8 J/m$^2$ and 3.2 $\times 10^{-9}$ J/m$^2$, respectively. In the top right region of the map (wide carbide spacing and high dislocation density), where $\Delta G_1$ is greater than $\Delta G_2$, recrystallization can occur at the tempering temperature. In the bottom left region (narrow carbide spacing and low dislocation density), where $\Delta G_1$ is smaller than $\Delta G_2$, only recovery proceeds without occurring the recrystallization. Measurements of $\lambda$ and $\rho$ were taken for both the LC and ULC steels tempered at 973 K for 0.6 ks (early stage of the tempering) and for 2.1 ks (just before the start of recrystallization in the ULC steel), and then plotted on Fig. 7. It is found that the data of ULC steel lie in the ‘only recovery’ area at first but move into the ‘recrystallization’ area before starting the recrystallization, while the data of LC steel remain in the ‘only recovery’ area even after the tempering. This result demonstrates that the possibility of recrystallization in lath martensite can be successfully explained by the BNG mechanism via the spacing of carbide particles on grain boundaries and the dislocation density of tempered martensite.

Generally, the tempering time dependence of both the spacing of grain boundary carbides and the dislocation density in the BNG mechanism should be considered as functions of chemical composition and microstructure. The relationship between $\Delta G_1$ and tempering time is schematically illustrated in Fig. 8. If the mean radius of carbide par-

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*3 The estimation of dislocation density from hardness can not be applied for the tempered LC steel because it is hardened by not only dislocations but also carbide particles. Thus, in this study, the dislocation densities of the LC steel were assumed to be similar as those of ULC steel under the same tempering conditions.
particles on grain boundaries coarsens during tempering in proportion to \((time)^{1/4}\) as Speight\(^{15}\) proposed, carbide spacing, \(\lambda\), is also proportional to \((time)^{1/4}\), and thus the \(\Delta G_1\) and \(\Delta G_2\) will be proportional to \((time)^{1/2}\) and \((time)^{1/4}\), respectively. Supposing the curves of \(\Delta G_1\) and \(\Delta G_2\) intersects on tempering, as thought to happen in the present ULC steel, recrystallization should be possible at times greater than the time of intersection of these curves (i.e. when \(\Delta G_1\) becomes greater than \(\Delta G_2\)). Hence, the time to the intersection corresponds to the incubation period of the recrystallization. The shape of the curves necessarily depend on the coarsening rate of the grain boundary precipitates. Moreover, the recovery rate of martensite significantly influences the incubation period through the reduction of driving force, \(\Delta G_1\). High chromium steels should have lower recovery rate than low alloy steels. This may have contributed to the occurrence of the recrystallization in ULC steel. From these points of view, in steels containing thermally stable precipitates or having high recovery rate, recrystallization should hardly occur although the amount of precipitate is small.

4. Conclusions

(1) Lath martensite in ultra-low carbon high-chromium steels causes discontinuous recrystallization during tempering at around 1000 K with the bulge nucleation and growth (BNG) mechanism: nuclei of recrystallized grains are formed through bulging packet boundaries and prior austenite grain boundaries, they then grow by consuming martensitic structure containing high density of dislocations.

(2) Recrystallization of lath martensite is suppressed by carbide particles precipitated on grain boundaries because they obstruct grain boundary migration by the pinning effect. When the carbide spacing on grain boundaries is smaller than the critical carbide spacing of \(\lambda_c = 4\sigma(\pi-2)/pE_d\pi\), the recrystallization should be completely suppressed in martensitic steels.

(3) The probability of recrystallization of lath martensite should be discussed by consideration of the tempering time dependence of both the spacing of grain boundary carbides affected by the ripening of the carbide particles and the dislocation density depending on the recovery of martensite. In the case of steels containing thermally stable precipitates or having high recovery rate, recrystallization should hardly occur although the amount of precipitate is small.

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REFERENCES