Annealing-induced Hardening in a Nanostructured Low-carbon Steel Prepared by Using Dynamic Plastic Deformation

L.X. Sun\textsuperscript{1)}, N.R. Tao\textsuperscript{1)*}, M. Kuntz\textsuperscript{2)}, J.Q. Yu\textsuperscript{3)}, K. Lu\textsuperscript{1)}

1) Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, China
2) Corporate Sector Research and Advance Engineering Production Technology 1, Materials and Process Engineering Metals, Robert Bosch GmbH, P.O. Box 30 02 40, 70442 Stuttgart, Germany
3) Research and Technology Center Asia Pacific, Bosch (China) Investment Ltd., Shanghai 200335, China

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Lamellar nanostructures were induced in a plain martensitic low-carbon steel by using dynamic plastic deformation at room temperature. The nanostructured steel was hardened after annealing at 673 K for 20 min, with a tensile strength increased from 1.2 GPa to 1.6 GPa. Both the remained nanostructures and annealing-induced precipitates in nano-scale play key roles in the hardening.

KEY WORDS: Nanostructure; Annealing; Precipitation hardening; Low-carbon steel; Dynamic plastic deformation

1. Introduction

Plastic deformation has been proven to be an effective approach to strengthen metallic materials due to the strain-induced grain refinement and high density of defects. The grain size of ferrite steel can be reduced to 100–200 nm by using plastic deformation methods, such as high pressure torsion (HPT)\textsuperscript{[1]}, equal-channel angular pressing (ECAP)\textsuperscript{[2]}, accumulative roll-bonding (ARB)\textsuperscript{[3]}. These ultrafine-grained (UFG) ferritic steels exhibit an enhanced tensile strength as high as 1 GPa. After annealing, strengths of these UFG ferritic steels usually decrease monotonously with an increasing annealing temperature\textsuperscript{[4,5]}. In the plain low-carbon steels with martensite, nano-scale lamellar structures were obtained through cold rolling, resulting in the tensile strength as high as 1.5 GPa\textsuperscript{[6]}. Consistent with the UFG ferritic steels mentioned above, softening was also observed in the nanostructured low-carbon steel with martensite after annealing. The annealing-induced softening was usually attributed to a decrease of defect density and microstructure coarsening at elevated temperatures.

Nanostructures were obtained in various metals and alloys by using dynamic plastic deformation (DPD)\textsuperscript{[7,8]}. In a DPD-processed nanostructured plain low-carbon steel, we found annealing-induced hardening, rather than softening as usually observed in deformed metals. The objective of the present study is to identify the origin of such an annealing-induced hardening.

2. Experimental

A commercial plain low-carbon steel with a carbon concentration of 0.2 wt% was used in this work. Its chemical composition is listed in Table 1. The as-received material was annealed at 1173 K for 2 h and then quenched in water to obtain the microstructure of lath martensite. The initial samples before DPD treatment are cylinders of 10 mm in diameter and 17 mm in height. The details of DPD set-ups and process can be found in our previous work\textsuperscript{[7,9]}. During DPD processing, the cylindrical samples are compressed with impact loading repeatedly. The DPD strain is calculated by the expression $\varepsilon = \ln(h_0/h)$, where $h_0$ and $h$ are the initial and final heights of DPD samples, respectively. The strain rate of each impact is estimated to be $10^2$–$10^3$ s$^{-1}$. In this work, the low-carbon martensitic steel sample was deformed to a strain of 1.7 by using DPD treatment at room temperature. Three groups of DPD samples were annealed at 673 K, 773 K and 873 K for 20 min, respectively. The water quenched samples without deformations (WQ for short) were also annealed with corresponding parameters for comparison.

Transmission electron microscopy (TEM) was used to characterize the microstructures of the steel samples, with a JEOL JEM-2010 microscope operated at 200 kV. The TEM samples were cut parallel to the loading directions and prepared with...
twin-jet. Back-scattered electron (BSE) signals in scanning electron microscopy (SEM) are used to obtain electronic channeling contrast (ECC) from the electrochemically polished surfaces which depend on the orientation of grains in the matrix and could show annealed microstructures clearly for large areas. Dog-bone tensile samples with the gauge dimensions of $5 \text{ mm} \times 1 \text{ mm} \times 0.5 \text{ mm}$ were cut perpendicular to the loading directions, which means parallel to the surface of samples. The tensile tests under a strain rate of $5 \times 10^{-3} \text{s}^{-1}$ were conducted on an Instron 8848 micro-tester with a laser extensometer at room temperature.

3. Results

3.1. Microstructures of the DPD steel with nanostructures

The microstructure of the DPD samples with a strain of 1.7 is characterized by lamellae with an aspect ratio of 15, as shown in Fig. 1(a). The lamellae are parallel to each other and perpendicular to the loading direction. Dislocation slipping dominates the plastic deformation and microstructure refinement in the martensitic low-carbon steel during DPD and deformation twinning is not found. The average thickness of lamellae is $84 \text{ nm}$ calculated by Gauss fitting with the thickness distribution in Fig. 1(b). Nanometer scaled microstructures are obtained which resemble those obtained in cold rolled martensitic low-carbon steels and are finer than those in the ferritic low-carbon steels processed by plastic deformations such as ECAP and ARB etc. The nanostructures obtained during DPD are mainly with the benefits of the initial lath microstructures scaled in several hundreds nanometers and the supersaturated carbon in the matrix. High density of dislocations, another typical feature of the cold deformed metals, is also observed accumulating on the lamellar boundaries and tangling in the spaces between the boundaries. It has been confirmed in other metals that higher strain rate in DPD than that in conventional deformation methods with lower strain rate is beneficial to the formation of higher defect density and finer microstructures in the matrix.

3.2. Microstructures of annealed samples

Microstructural observations of the annealed samples were conducted with SEM on the WQ and DPD samples (Fig. 2(a) and (b)) for comparisons. Characteristic size distributions of microstructures of DPD samples are shown in Fig. 2(c). The microstructures of WQ samples do not change much after annealing. Sub-microstructures in the regions of original austenite grains can be observed in the samples annealed at 673 K and 773 K. Fully recrystallized microstructures are obtained after annealing at 873 K for 20 min, but the grain size keeps nearly unchanged. In contrast, remarkable changes of microstructures take place in the DPD samples after annealing. The lamellar structures still exist after annealing at 673 K for 20 min. The average thickness of lamellae measured with SEM-ECC images is about 111 nm. The detailed observations under TEM (Fig. 3(a)) show that there is no obvious change in microstructure. The average thickness of lamellae is 86 nm, identical to that of the as-deformed state. High density of dislocations induced by the DPD is still remaining. Carbide particles with the size of several nanometers were observed at the lamellar boundaries and inside the lamellae, as arrowed in Fig. 3(b). When annealed at 773 K, recrystallization takes place uniformly and equiaxed grains are obtained. The average grain size is about $242 \text{ nm}$. The grain size distributions of equiaxed grains (in DPD samples annealed at 773 K and 873 K) were weighted with the square of grain sizes. Some of ultrafine grains grew into coarse grains embedded in the ultrafine-grained matrix of the sample annealed at 873 K. Carbide particles could be observed with SEM, which are arranged in line perpendicular to loading directions in coarse grains or pinned at the grain boundaries of ultrafine grains.

3.3. Tensile properties and annealing-induced hardening

The as-quenched samples exhibit a lower strength and a higher tensile ductility (Fig. 4(a)) compared with those in martensitic low-carbon steels reported in literature. This is attributed to a long holding duration (2 h) at 1173 K before water quenching, for ensuring a sufficient high ductility for cold deformation and uniform distribution of carbon in the matrix.

<table>
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<tr>
<th>Table 1</th>
<th>Chemical composition of the low-carbon steel studied in this work (wt%)</th>
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<tr>
<td>C</td>
<td>Mn</td>
</tr>
<tr>
<td>0.205</td>
<td>0.510</td>
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Fig. 1 A typical cross-sectional TEM bright-field image of the DPD martensitic plain low-carbon steel (a) and the thickness distribution of lamellar structures measured from cross-sectional TEM bright-field images (b).
Comparing with the as-quenched samples, the as-deformed samples have a higher strength and a much lower uniform elongation (Fig. 4(b)) which are typical features of heavily deformed metals. After DPD, the tensile strength increases from 700 MPa to 1.2 GPa.

After annealing, the strength of WQ samples decreases slightly with increasing annealing temperature. Different from the monotonous decrement on tensile strength with increasing the annealing temperature in the reported UFG or nanostructured low-carbon steels, the strength of the DPD samples in this work rose to 1.6 GPa firstly after being annealed at 673 K, and then decreased at higher annealing temperatures. The ones annealed at 773 K have the average strength of nearly 1.2 GPa, which is close to the strength of the as-deformed ones. After annealing at 873 K for 20 min, the strength drops to 700 MPa. The tensile curves exhibit high yield ratio and low tensile elongation although the tensile strength is close to that of the undeformed ones. All of the DPD tensile samples with and without annealing break with necking and ductile fracture morphology.

4. Discussion

The annealing usually results in softening of the materials strengthened with deformation due to grain coarsening and the reduction of dislocation density, which has been reported in numerous deformed materials\cite{4,5,12-14}. However, a few investigations also showed hardening by annealing in deformed materials, such as HPT Ti\cite{15}, ARB Al\cite{16} and ARB IF steels\cite{17}.

![Fig. 2](image1.png) Typical cross-sectional SEC-ECC images of the as-quenched (WQ) (a) and DPD martensitic low-carbon steel samples (b) after annealing at different temperatures, and the grain size distributions of DPD samples after annealing (c).

![Fig. 3](image2.png) Typical cross-sectional TEM bright-field images of DPD samples after annealing at 673 K for 20 min (a and b) and the distribution thickness of lamellar structures measured from TEM bright-field images (c).
The strength is raised by 25 MPa in ARB Al and 43 MPa in ARB IF steel, respectively, after annealing at low temperatures. Dislocation source limited strengthening\(^{[17]}\) is considered to work, which means after the dislocation sources have decreased in annealing at low temperatures, higher stress is required to activate the alternative dislocation sources to deform the samples. However, dislocation source limited strengthening is not the only mechanism that induced hardening in the DPD martensitic low-carbon steel after annealing, especially in the 673 K annealed ones.

The microstructures of the matrix in the as-DPDed samples and the DPD ones annealed at 673 K are characterized by lamellar structures with nearly the same size (Figs. 1 and 3) and similar dislocation density, between which the distinct difference is that there are carbide precipitates dispersed in the matrix of the annealed samples which scaled in several nanometers or even smaller. The pinning effects of the nano-scale precipitates could keep the deformation-induced defects, such as dislocations and boundaries stable during annealing. The retained deformation microstructures in the annealed sample prevent the matrix from softening. In addition, these carbides could provide obvious precipitation hardening, which enhanced the tensile strength from 1.2 GPa to 1.6 GPa. Precipitation hardening can also be identified in the samples annealed at 773 K. The strength of the sample annealed at 773 K only decreases slightly from the level of the as-deformed one though recrystallization has taken place and the average size of microstructures has risen from 84 nm to 242 nm. In other words, the precipitation hardening compensated the strength drop induced by microstructure coarsening. The strengthening mechanism is similar to those reported in the nanostructured Al alloy\(^{[18,19]}\). Dislocation source limited strengthening also plays an important role, which gives rise to the appearance of yield drop on the tensile curves when recrystallized grains become main microstructures. Nevertheless, conventional models are not suitable to precisely calculate the precipitation hardening in the nanostructured alloys with high density of defects induced by heavy deformation. A lot of work is still needed for quantitative calculation. When annealed at 873 K, the coarsening of both matrix and precipitates make the strength drop to the level of the undeformed ones.

Precipitation hardening is a common phenomenon some series of Al alloys when the matrix undergoes different degrees of deformation, even when the grain sizes of the matrix are refined into sub-micron or nanometer scales\(^{[18,19]}\). But precipitation hardening was seldom reported in martensitic low-carbon steels with similar compositions. Softening by annealing was reported in the nanostructured martensitic low-carbon steel with 0.13 wt% C prepared by cold rolling\(^{[12]}\). Comparing with the deformation methods with low strain rate, the higher strain rate during DPD induced higher density of deformation defects like dislocation boundaries and dislocation tangles, which provide abundant positions for the nucleation of carbide so that the carbide precipitates are fine and well dispersed in the nanostructured matrix. Therefore, the key factors of the hardening by annealing in the present low-carbon steel are (i) the supersaturation of carbon concentration in the initial martensitic low-carbon steel; (ii) the nanostructures and high density of defects induced by high strain rate deformation, such as DPD; (iii) appropriate annealing temperatures and durations. These factors make carbide particles fine and in dispersed distribution, which result in suppression of decrements of deformation defects including grain growth and provide high precipitation hardening. Further work is in processing to reveal the mechanism for annealing-induced hardening quantitatively and to optimize the properties in the DPD martensitic low-carbon steels.

5. Conclusions

(1) A nanostructured plain low-carbon steel with a high strength of 1.2 GPa was prepared by using dynamic plastic deformation. The microstructure is characterized by lamellar structures with an average thickness of 84 nm and high density of dislocations. Annealing-induced hardening was found in the nanostructured martensitic low-carbon steel. The tensile strength of the DPD sample rose from 1.2 GPa to 1.6 GPa after annealing at 673 K for 20 min.

(2) Compared with the annealing-induced softening usually found in heavily deformed metallic materials, the annealing-induced hardening in the present work is...
attributed to the retained deformation microstructures and the well dispersed fine carbide precipitates in the annealed samples.

(3) The nanostructures and high density of dislocations induced by DPD in martensite facilitate the precipitation of fine carbides with dispersed distributions during appropriate annealing by providing abundant positions for nucleation. The well dispersed carbides promote the stability of the nanostructures by pinning the dislocations and boundaries.

Acknowledgments

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