High-strength state of ultrafine-grained martensitic steel produced by high pressure torsion

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Abstract The paper presents the study results on the effect of severe plastic deformation (SPD) via high pressure torsion (HPT) on the structure and properties of martensitic steel. The contribution of different strengthening mechanisms in the strength of steel has been analyzed. It is shown that independently of the deformation temperature the main contribution in hardening belongs to grain boundaries (about 50 %), whereas the dislocation and solid solution components achieve 15 and 25 %, respectively.

1 Introduction

High strength in martensitic steels at room and elevated temperatures is traditionally ensured by two-stage thermal treatment. At the first stage as a result of quenching strengthening takes place due to phase hardening during martensitic transformation. At the second stage during heating of the quenched steel, the strength grows according to the mechanism of dispersion hardening at the expense of formation of precipitates [1-4].

It has been recently shown that the strength of steels can be enhanced using severe plastic deformation, for example by high pressure torsion (HPT) [5-8]. During HPT an ultrafine-grained microstructure with grain sizes less than 1 μm can be observed, which allows additional strengthening in accordance with the Hall-Petch ratio [9,10]:

\[ \sigma_y = \sigma_0 + K_y d^{-\frac{1}{2}} \]  

(1)

where \( \sigma_0 \) – the friction stress of a crystalline lattice, \( K_y \) – the coefficient of strengthening at the expense of grain boundaries, \( d \) – the grain size.

A grain size depends on the parameters of deformation and thermal treatment. Usually when a deformation temperature increases, grain sizes grow [11]. Therefore, in order to achieve the maximum strengthening effect due to microstructure refinement, one should reduce the deformation temperature. However, several publications have appeared recently [7,8,12], which demonstrate that the dependence of strength on the deformation temperature is not always decreasing for steels. The maximum strength for carbon steels 10 and 45 was observed after severe plastic deformation at 350 °C [7, 8]. The observed strength in the austenite steel after deformation at 300 °C is higher than that after deformation at room temperature [12]. This is explained by a complex microstructure observed in steels as a result of severe plastic deformation. The
deformation temperature affects not only grain sizes, but also such structural features as sizes and distribution of secondary particles, formation of segregations, phase composition, formation of twins [7-8, 11-12]. The influence of temperature on the microstructure and properties of complex-alloyed steels and alloys, for which dispersed particles are one of the main factors for strength enhancement, is especially ambiguous. This work deals with the possibility of strength enhancement in martensitic steel as a result of severe plastic deformation.

2 Material and procedure
Martensitic steel in the shape of a hot-rolled rod is taken as research material. Table 1 displays the chemical composition of the steel under study.

| C    | Si  | Mn       | Ni   | S    | P    | Cr       | Mo  | W      | V    |
|------|-----|----------|------|------|------|----------|-----|--------|------|
| 0.1 - 0.16 | to 0.6 | to 0.6  | 1.5 - 1.8 | to 0.03 | to 0.03 | 10.5 - 12 | 0.35 - 0.5 | 1.6 - 2 | 0.18 - 0.3 |

The steel samples with a diameter of 20 mm and a thickness of 1 mm in the quenched and tempered states are subjected to HPT at room and elevated T=300 °C temperatures. The pressure is 6 GPa, the number of rotations is N=10.

Thin foils for TEM investigations were cut at a distance of 1/2 of the radius from the sample center. After mechanical polishing of foils to a thickness of 0.1 mm, a disc with a diameter of 3 mm is cut out. It is thinned on a jet polishing set "Tenupol-5" in the electrolyte based on n-butyl alcohol. The thinning is performed in the regime: U=60 V at the current stabilization till holing. The fine structure of foils is studied using a transmission electron microscope JEM-2100 at an accelerating voltage of 200 kV and a scanning electron microscope JSM-6390.

Diffraction patterns were recorded on X-ray diffractometer DRON-4 with Co Ka emission using a monochromator, 0.02 ° step and a holding time of 5 seconds for each step. Analysis of diffraction patterns was performed using «MAUD» program [13] which is in the public domain[14]. The dislocation density $\rho_{XRD}$ is determined according to the results of X-ray studies via the formula:

$$\rho_{XRD} = \frac{2\sqrt{3} (\epsilon^2)^{1/2}}{bd_{XRD}},$$

where $(\epsilon^2)^{1/2}$ – the level of elastic microdistorsions of the crystalline lattice; $b$ – the Burgers vector of dislocations; $d_{XRD}$ – the coherent scattering domain sizes.

The average grain size is measured by Random Intercept method. The microhardness is measured on a Micromet-5101 set at a load of 100 g for 10 seconds.

3 Results
The microstructure of the steel in the initial state is a ferrite matrix with non-uniformly distributed particles of strengthening phases, which can be clearly seen on the image taken with the SEM (Fig.1a). The average size of the matrix phase is 1400±100 nm. The majority of particles is localized on grain boundaries of the ferrite matrix. The average size of particles is 171±30 nm, the volume fraction is 5 %. Clear grain boundaries can be observed in the fine structure (Fig. 1b). The majority of grains are free of dislocations. Pile-ups of dislocations can be seen in separate grains.
Non-uniform distribution of particles in the volume, as a rule, decreases the deformability of the material. Therefore, prior to deformation the samples are quenched in oil after heating and holding at 1050 °C, for 1 hour, then they are tempered at 800 °C for 1 hour.

After quenching martensitic microstructure can be observed (Fig.1 c, d). The average thickness of martensite plates is 518 nm. The martensite is characterized by high dislocation density within a plate. Separate dislocations as well as wide dislocation walls are observed within martensite plates. There are no secondary phase particles, which are typical of the initial state, in the martensitic structure.

**Figure 1.** Microstructure of steel in the initial (a, b); quenched (c, d) and tempered (e, f) states.
After tempering uniform distribution of secondary phase particles in the volume is observed (Fig. 1 e, f). The particles precipitate in the boundaries of prior austenite grains and along the boundaries of martensite plates. Only about 25% of particles are located in martensite plate bodies. The average size of particles is 250 nm, the volume fraction achieves 19%. The plate structure of the martensite retains. The average size of plates is 556 nm.

As a result of HPT the microstructure changes significantly. The sizes of structural components are determined by the deformation temperature. Thus, after HPT at room temperature an ultrafine-grained microstructure with an average grain size of about 140 nm forms (Fig. 2 a, c). After torsion under HPT at an elevated temperature T=300°C the average grain size is 200 nm (Fig. 2 d, f). The diffraction pattern with spots located around the circle (Fig. 2c) testifies to high-angle misorientations. The dark-field images display (Fig. 2e) that after HPT at room temperature grains have the shape close to the equi-axed one.

After HPT at an elevated temperature the spots on the diffraction pattern are located around the circle, but they have azimuthal blurring indicating to high internal stresses (Fig. 2d).
Examination of the microstructure in SEM at a lower magnification (Fig. 2 a, b) demonstrates a significant reduction of the volume fraction of secondary phase particles in the deformed samples as compared to the quenched and tempered states (Fig. 1 e). The volume fraction of particles is 4.5 % after HPT at 20 °C and 3.6 % after HPT at 300 °C. The average particle size is 109 nm after HPT at room temperature and 160 nm after HPT at 300 °C (Fig. 2 a, b). This is confirmed by the data of X-ray analysis of the samples, which are listed in Table 2.

Table 2. X-ray analysis results.

| State                        | Lattice parameter, Å | CSD size, nm | Microdistorsions, % |
|------------------------------|----------------------|--------------|---------------------|
| Quenching 1050°C+tempering 800 °C | 2.87818(3)           | 1500         | 0.039               |
| HPT at 20°C, 10 rot, 6 GPa    | 2.88043(5)           | 35           | 0.27                |
| HPT at 300°C, 10 rot, 6 GPa   | 2.88021(8)           | 40           | 0.32                |

The results of X-ray analysis by the Rietveld method using the MAUD software package are listed in Table 2. These results demonstrate that in samples after HPT the level of microdistorsions of the crystalline lattice increased by an order as compared to the quenched and tempered steel. A much higher level of microdistorsions is observed after SPD at an elevated temperature: 0.32 % against 0.27 % for steel after SPD at room temperature. Besides, HPT results in increase of the lattice parameter as compared to the state after quenching and tempering. A significant reduction of CSD dimensions is observed (Table 2) as well.

Thus, the deformation temperature impacts not only the grain sizes, but also the quantity and sizes of secondary phase particles. In particular, after the deformation at an elevated temperature the...
particle sizes are larger, the volume fraction is lower than those after the deformation at room temperature. The microstructure change as a result of severe plastic deformation affects considerably the steel microhardness (Fig. 3). The microhardness in the initial state is 3200 MPa, after quenching the microhardness increases up to 5100 MPa, and after tempering it returns practically to the value typical of the initial state – 3500 MPa.

![Figure 3. Microhardness of steel in different states.](image)

As a result of SPD the microhardness increases practically two times as compared to the initial state and by 25 % as compared to the quenched state of steel (Fig. 3). The deformation temperature insignificantly effects the microhardness. Thus, the microhardness value after HPT at room temperature is 6700 MPa, it is 6300 MPa after HPT at an elevated temperature.

### 4 Discussion

Achievement of a high-strength state in steels as a result of severe plastic deformation is ensured by formation of a unique microstructure, in which a number of strengthening factors is combined. First of all, it is an ultrafine-grained matrix that ensures grain boundary hardening in accordance with the Hall-Petch ratio [9, 10]. Besides, solid solution hardening of an alloyed ferrite and dispersion hardening at the expense of high-dispersion particles of the strengthening phases take place for the complex-alloyed steel.

Solid solution hardening \( \Delta \sigma_{SS} \) is determined by the content of the alloying element and its strengthening effect [15]:

\[
\Delta \sigma_{SS} = k_i c_i,
\]

where \( k_i \) – the strengthening coefficient for ferrite, which is an increment of the yield stress when 1 % (wt) of the \( i \)-th alloying element dissolves in it; (for carbon \( k = 4670 \) MPa/ % [16]) \( c_i \) – the concentration, % (wt), \( i \)-th alloying element.

Hardening at the expense of dispersion particles can be determined in accordance with the Orowan’s ratio [17]:

\[
\Delta \sigma_{or} = 1.2Gbf^{1/3}d^{-1},
\]
where $G=78 \text{ GPa}$ – the shear modulus of the ferrite, $b=0.25 \text{ nm}$ – the Burgers vector for dislocations $\frac{1}{2}<111>$ of the ferrite, $f, d$ – the volume fraction and size of particles.

The dislocation hardening can be calculated via the formula:

$$
\Delta \sigma_d = \alpha M b G \rho^{1/2} \tag{5}
$$

where $\Delta \sigma_d$ – the dislocation hardening, where $\alpha$ is the coefficient depending on the character of dislocation interaction in the course of work hardening; $M$ is the orientation multiplier: for $\alpha$-iron $M = 2.75$, and the product $\alpha M = 0.5$, $b$ – the Burgers vector, $\rho$ – the dislocation density[16].

In general at first approximation the contribution of different mechanisms in the hardening is additive:

$$
\sigma_y = \Delta \sigma_0 + \Delta \sigma_{ss} + \Delta \sigma_{or} + \Delta \sigma_{gb} + \Delta \sigma_d \tag{6}
$$

where $\sigma_0$ - the friction stress of the $\alpha$-iron lattice; $\Delta \sigma_{ss}$ - solid solution hardening; $\Delta \sigma_{or}$ - dispersion hardening; $\Delta \sigma_{gb}$ - grain boundary hardening; $\Delta \sigma_d$ - dislocation hardening.

The friction stress of the $\alpha$-iron lattice ($\sigma_0$) is determined by the Peierls–Nabarro stress [18]:

$$
\sigma_0 = 2 \times 10^{-4} G. \tag{7}
$$

Table 3 lists the results of calculation of the contribution of different strengthening mechanisms in the yield stress of steel in accordance with equations (1),(3-5), (7). The comparison with the experimental results is made with account of the empirically set correlation between strength and hardness: $\sigma_y = \frac{1}{3} HV$ [19]. This relation can be used to analyze the yield stress, as the yield stress of steels with an ultrafine-grained microstructure produced by HPT is, as a rule, close to the ultimate strength value.

The following assumptions were accepted during calculation of solid solution hardening. One-phase structure, namely solid solution of all the alloying elements, is taken for the quenched structure. The carbon content, which most significantly hardens $\alpha$-Fe, in the tempered structure is taken as equilibrium (0.006 % according to diagram “iron-cementite”). The results of X-ray analysis are taken into account for steel after HPT (Table 2), which showed the increase of the lattice parameter as a result of deformation by 0.002Å, which corresponds to the growth of the carbon content in the solid solution by 0.02 %.

| state       | $\Delta \sigma_{gb}$ | $\Delta \sigma_d$ | $\Delta \sigma_{ss}$ | $\Delta \sigma_{or}$ | $\sigma_y$ | $1/3 \text{HV}$ | HV |
|-------------|----------------------|-------------------|----------------------|----------------------|------------|---------------|-----|
| quenched    | 555                  | 185               | 1049                 | -                    | 1789       | 1700          | 5100|
| tempered    | 536                  | 20                | 448                  | 356                  | 1360       | 1166          | 3500|
| HPT 20°C    | 1069                 | 319               | 541                  | 166                  | 2095       | 2233          | 6700|
| HPT 300°C   | 894                  | 325               | 541                  | 51                   | 1811       | 2100          | 6300|

The results listed in the table show that the estimated values are very close to the experimental ones, which proves the possibility of considering the principle of linear superposition when
evaluating the contribution of different mechanisms in the yield stress in accordance with (4). Thus, the strength of the quenched steel is ensured mainly by solid solution hardening (58 %), and the contribution of grain boundaries is significant (30 %). After tempering the solid solution hardening reduces to 38 % with the dispersion hardening increase to 30 %.

Another situation is observed during HPT. Independently of the deformation temperature the main contribution belongs to grain boundaries. Nevertheless, this contribution is only about 50 %. The dislocation (to 15 %) and solid solution (to 25 %) components increase significantly.

An interesting fact is that the estimate values of the yield stress for the quenched steel and quenched and tempered steel exceed the experimental value of HV/3, whereas the values are reduced by 10% for the ultrafine-grained steel after HPT. This can be caused by the inaccuracy connected with evaluation of microstructural parameters as well as the content of alloying elements in the solid solution. However, one can suppose synergetic effect or some new strengthening mechanism, for example, at the expense of segregations formation on grain boundaries [20, 21].

The results obtained counts in favor of the complex structure formed during SPD that gives possibility of simultaneous activation of several mechanisms of hardening. As a result of such collective effect, the superstrength is achieved in steels with the ultrafine-grained structure.

5 Conclusions

1. As a result of HPT the ultrafine-grained microstructure has been produced in martensitic steel. Significant reduction of the volume fraction of secondary phases from 19 % in the tempered state to 3.6–4.5 % in the deformed state is observed. When the deformation temperature increases from the room one to 300°C, the grain sizes grow from 140 nm to 200 nm, the sizes of particles increase, and the volume fraction of secondary phases decreases.

2. Evaluation of the contribution of different strengthening mechanisms in the strength properties showed that grain boundary hardening ensures only about 50% of strength. Only the combination of various strengthening mechanisms of the complex ultrafine-grained structure produced by HPT makes it possible producing superstrong steels.

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