Nanocrystalline structures and tensile properties of stainless steels processed by severe plastic deformation

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Abstract. The development of nanocrystalline structures in austenitic stainless steels during large strain cold rolling and their tensile behavior were studied. The cold rolling to total equivalent strains above 2 was accompanied by the evolution of nanocrystalline structures with the transverse grain size of about 100 nm. The development of deformation twinning and martensitic transformation during cold working promoted the fast kinetics of structural changes. The development of nanocrystalline structures resulted in significant strengthening. More than fourfold increase in the yield strength was achieved. The strengthening of nanocrystalline steels after severe plastic deformation was considered as a concurrent operation of two strengthening mechanisms, which were attributed to grain size and internal stress. The contribution of internal stresses to the yield strength is comparable with that from grain size strengthening.

1. Introduction

The development of ultrafine grained and/or nanocrystalline structures by severe plastic deformation is one of the most promising methods for obtaining various steels and alloys with superior mechanical properties [1, 2]. It has been shown that the ultrafine grained and/or nanocrystalline structures could be obtained in almost all metallic materials after sufficiently large strains at relatively low temperatures [3]. By now a number of special deformation methods have been successfully utilized for severe straining [1, 4-6]. It should be noted that the effectiveness of deformation treatment for the development of ultrafine grained or nanocrystalline metals/alloys depends significantly on the susceptibility of the processed material to the grain fragmentation/subdivision upon plastic working. The high efficiency of cold working should be expected for processing of metallic materials exhibiting deformation twinning and/or strain-induced martensitic transformation [7-10]. Typical representatives of such materials are austenitic stainless steels. The low values of stacking fault energy in austenitic stainless steels promote the deformation twinning, whereas the metastable austenite experiences martensitic transformation under cold working conditions. Therefore, the austenitic stainless steels can be easily processed in ultrafine grained or nanocrystalline states by using conventional method of metal working without application of any special techniques.

The development of ultrafine grained or nanocrystalline structures is of the utmost importance for austenitic stainless steels. These materials are characterized by relatively low yield strength after ordinary thermomechanical processing [11]. The structural strengthening by means of large strain deformations has been shown as an advanced approach for production of high strength austenitic stainless steels [12, 13]. The aim of the present study is to clarify the mechanisms of structural
changes leading to the development of nanocrystalline structures in austenitic stainless steels during cold rolling and to elucidate the relationship between the developed nanocrystalline structures and the mechanical properties. Two types of steels, i.e. 316L and 304L, are used as typical representatives of frequently used austenitic stainless steels.

2. Experimental

A 316L-type austenitic stainless steel (Fe-0.04%C-0.4%Si-1.7%Mn-17.3%Cr-10.7%Ni-2%Mo-0.04%P-0.05%S) and a 304L-type austenitic stainless steel (Fe-0.05%C-0.4%Si-1.7%Mn-18.2%Cr-8.8%Ni-0.05%P-0.04%S) were used as starting material. The steel samples were hot rolled and then annealed at 1100°C for 10 min. The initial annealed grain sizes were 21 and 24 µm in the 316L and 304L steel samples, respectively (figure 1). The cold working was carried out by caliber rolling 9.2 mm × 9.2 mm square bars into 1.25 mm × 1.25 mm square bars at ambient temperature, leading to a total equivalent strain of 4.

Structural investigations were performed close to the sample axis on sections parallel to the rolling axis, using a Nova Nanosem 450 scanning electron microscope (SEM) incorporating an orientation imaging microscopy (OIM) system and a JEM-2100 transmission electron microscope (TEM). The grain sizes were measured on perpendicular to the rolling axis by a linear intercept method, including all boundaries with misorientation θ ≥ 15° revealed by OIM micrographs. The volume fractions of the austenite were averaged through OIM, X-ray analysis and magnetic induction method. The hardness of rolled samples was studied using Vickers hardness tests with a load of 3N with a corresponding indent diagonal of about 35 µm. The tensile tests were carried out by using specimens with a gage length of \( L_0 = 5.65\sqrt{S_0} \), where \( S_0 \) is the cross-sectional area.

![Figure 1. Initial microstructures of 316L (a) and 304L (b) stainless steels.](image)

3. Results and discussion

3.1. Strain hardening

The effect of cold rolling on the hardness is shown in figure 2. Qualitatively, almost the same strain dependencies of the hardness are obtained for both the 316L and 304L steel samples. An early deformation is accompanied by significant hardening. In the both steels, the hardness rapidly increases to about 3500 MPa after rolling to a strain of 0.4. Then, the rate of strain hardening gradually decreases to approx. 350 MPa per a strain, leading to almost linear dependence of the hardness on the strain in the strain range of 1 < ε < 4. It should be noted that the 304L steel samples are characterized by somewhat higher hardness values at all the total strains studied.
Figure 2. The strain effect on the hardness of 316L and 304L steels.

Figure 3. Deformation microstructures (a, c) and the corresponding phase maps (b, d) of the 316L stainless steel subjected to cold rolling to total equivalent strain of 1.2 (a, b) and 4 (c, d). The inverse pole figures in (a, c) are shown for the rolling axis (RA).
3.2. Microstructural evolution

Typical deformation microstructures that evolved in the 316L steel samples subjected to cold rolling to different total strains are shown in figure 3. The structural changes at relatively small strains are characterized by the development of deformation twinning (figure 3a), which leads to subdivision of the original grains by frequent network of twin boundaries, and the partial martensitic transformation, which results in formation of strain-induced ferritic nanocrystallites (figure 3b). The strain-induced ferrite appears primarily at the deformation microbands and the deformation twins. The fraction of strain-induced ferrite (bcc-martensite) increases with increasing the total strain. At large strains, the deformation structure consists of mixture of ferrite and austenite grains, which are highly elongated along the rolling axis (figures 3c and 3d).

The 304L steel samples also experience the deformation twinning and the martensitic transformation during cold rolling (figure 4). In contrast to 316L steel, however, the austenite in 304L steel is characterized by a lower stability against martensitic transformation. The volume fraction of the strain-induced ferrite in the 304L steels samples comprises about 0.1 after cold rolling to a strain of 0.4 (figures 4a and 4b). The fraction of strain-induced ferrite (bcc-martensite) in 304L steel rapidly increases upon further cold rolling. Therefore, the deformation microstructure that developed in 304L steel at a total strain of 4 consists of the highly elongated ferrite grains and the separate chains of austenite nanocrystallites (figures 4c and 4d).

Figure 4. Deformation microstructures (a, c) and the corresponding phase maps (b, d) of the 304L stainless steel subjected to cold rolling to total equivalent strain of 0.4 (a, b) and 4 (c, d).

The inverse pole figures in (a, c) are shown for the rolling axis (RA).
Typical fine structures that evolved in the 316L and 304L steel samples subjected to cold rolling to a total strain of 4 are shown in figures 5a and 5b, respectively. The nanocrystalline structures developed at large strains are characterized by high dislocation densities, which exceed $10^{15}$ m$^{-2}$ in the both steels. Moreover, the numerous bend contours, which are clearly evident on the TEM images, suggest the large internal distortions that evolved in the nanocrystalline structures at large strains. Such high dislocation densities and corresponding internal distortions can significantly affect the strength of processed samples. Therefore, the strengthening of the rolled samples should be considered as a superposition of the grain size strengthening and the internal stresses aroused by large internal distortions.

**Figure 5.** TEM micrographs of the fine structures evolved in 316L (a) and 304L (b) steels subjected to cold rolling to total equivalent strain of 4.
The strain effect on the structural changes is quantitatively represented in figures 6 and 7. In the 316L steel samples, the austenite fraction gradually decreases during rolling in the strain range of 0.4 to 4, finally approaching 0.44 at a strain of 4 (figure 6). On the other hand, the martensitic transformation develops more readily in the 304L steel samples (figure 6). The austenite fraction rapidly decreases to about 0.17 during cold rolling to a strain of 2 and then slightly decreases to 0.16 upon further processing to a strain of 4. It should be noted that the equilibrium austenite fractions with partitioning of alloying elements at ambient temperature are about 0.15 and 0.13 in the 316L and 304L steels, respectively, as calculated by ThermoCalc. The sluggish kinetics of strain-induced ferrite formation in the 316L steel as compared to 304L steel may be associated with larger amount of nickel in the former that suppress the martensitic transformation.

Commonly, the cold rolling results in significant reduction in the transverse grain size (figure 7). In the 316L steel samples, the transverse austenite grain size rapidly decreases to about 160 nm during rolling to a total strain of 2 and then hardly changes during subsequent rolling. Similarly, the transverse austenite grain size in the 304L steel samples decreases to 70 nm after straining to 2 followed by slight decrease to 60 nm at a total strain of 4. The strain-induced ferrite appears with the transverse grain size of about 200 nm in the both steels. Then, the transverse ferrite grain size decreases to about 100 nm after straining to 2 and 1.2 of the 316L and 304L steel samples, respectively, and does not vary remarkably during further rolling.
3.3. Tensile behaviour

A series of engineering stress-strain curves for the nanocrystalline stainless steels subjected to cold rolling with different total rolling strains are shown in figure 8. Portions of the flow curves for initially annealed steel samples are also displayed in the figure for reference. Commonly, the tensile stress-strain curves are characterized by a peak stress at relatively small strain of about 1% followed by a decrease of the stress that is associated with a quick necking of tensile specimens. The tensile strength increases with increase in the previous rolling strain. The cold rolling to a total strain of 2 results in tensile strength above 1500 MPa, and further rolling to a total strain of 4 leads the tensile strength to above 2000 MPa in the both steels, although the 304L steel samples exhibit somewhat higher tensile stress levels than the 316L ones. Correspondingly, the strengthening by cold rolling is accompanied by a remarkable degradation of the plasticity. The tensile elongation does not exceed several percents in the severely strained samples.

Figure 7. The strain effect on the transverse austenite/ferrite grain size in 316L and 304L steels.
Generally, the grain size strengthening can be represented by the Hall-Petch relationship between the yield strength ($\sigma_{0.2}$) and the grain size (D) [14]:

$$\sigma_{0.2} = \sigma_0 + k_y D^{-0.5} \quad (1)$$

In the case of duplex microstructures like an austenite/ferrite mixture, the yield strength should result from the fractional strengths of the both components:

$$\sigma_{0.2} = f_{\text{aust}} \sigma_{0.2}^{\text{aust}} + f_{\text{ferrite}} \sigma_{0.2}^{\text{ferrite}} \quad (2)$$

where $f_{\text{aust}}$ and $f_{\text{ferrite}}$ are the volume fraction of austenite and ferrite, respectively. Taking the $\sigma_0 = 200$ MPa [15], the following Hall-Petch relationship can be obtained for austenite, $\sigma_{0.2} = 200 + 130 D^{-0.5}$, which is quite similar to that reported for grain size strengthening in an S304H-type austenitic stainless steel [16]. The grain size strengthening of ferrite was studied in Fe – 15%Cr steel [17], and the following Hall-Petch relationship was reported, $\sigma_{0.2} = 120 + 240 D^{-0.5}$.

Figure 9 represents the values of the yield strengths obtained in the 316L and 304L steels after cold rolling to different total strains along with the Hall-Petch strengths calculated by equations (1) and (2). It is clearly seen in figure 9 that the experimental values of the yield strength are significantly higher than those predicted by Hall-Petch strengthening. Therefore, the effect of internal stresses should be taken into account in addition to the Hall-Petch strengthening, while the strengthening of severely deformed material is considered. Thus, the strength of severely deformed material results from Hall-Petch strengthening ($\sigma_{\text{Hall-Petch}}$) and additional strengthening by internal stress ($\Delta \sigma$).

$$\sigma_{0.2} = \sigma_{\text{Hall-Petch}} + \Delta \sigma \quad (3)$$

The high internal stresses in the studied materials after large strains are associated with high dislocation densities including an inhomogeneous distribution of misfit grain boundary dislocations, which are reaction products of dislocation fluxes crossing over the neighboring grains [17, 18]. Since the misfit dislocation density is in direct proportion of dislocation flux, which in turn depends linearly...
on the strain, the additional strengthening by the internal stresses should be directly related to the strain, i.e., \( \Delta \sigma = K \varepsilon \).

\[ \Delta \sigma = 300 \varepsilon \]

**Figure 9.** The effect of cold rolling strain on the yield strength of 316L and 304L steels.

**Figure 10.** Relationship between the internal stresses (\( \Delta \sigma \)) and the rolling strain (\( \varepsilon \)) for 316L and 304L steels.

All the experimental data obey the following expression for the internal stress, \( \Delta \sigma = 300 \varepsilon \) (figure 10). The relationship between the experimental yield strengths and those calculated by equations (1-3)
is shown in figure 11. It is clearly seen that yield strengths obtained by a summation of the Hall-Petch and internal strengthening are coincident with the experimental results.

![Figure 11](image)

**Figure 11.** Relationship between the experimental and calculated yield strengths of 316L and 304L steel subjected to cold rolling.

4. Summary
The development of nanocrystalline structures in 316L and 304L austenitic stainless steels during large strain cold rolling was studied. The present steels are characterized by fast kinetics of grain refinement. The cold rolling to total strains above 2 resulted in the development of nanocrystalline structures with the transverse grain sizes of about 100 nm. The rapid grain refinement was promoted by the development of deformation twinning and martensitic transformation. The martensitic transformation developed more readily in the 304L steel samples, leading to somewhat finer average austenite/ferrite grains as compared to 316L ones. The development of nanocrystalline structures resulted in significant strengthening, which was considered as a result of the grain size strengthening (Hall-Petch relationship) and the internal stresses, which were attributed to the large internal distortions involved by severe deformation.

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