Effects of cold rolling deformation on microstructure in 18/8 grade stainless steel

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Abstract. 18-8 stainless steel is one of the most versatile materials. Some of its uses are related to structural applications; in this case, the high mechanical properties required are obtained by controlled lamination after re-crystallisation annealing. In this work, detailed analyses by FEG-SEM and EBSD have been done to correlate the mechanical properties, the amount of martensite and the structural misorientation generated. A linear relation between yield strength and local misorientation has been obtained.

1. Introduction

AISI 304 (EN 1.4301) is the most versatile and widely used stainless steel. It is characterized by an austenitic fcc crystallographic structure with a nominal composition of 18 wt% of chromium and 8 wt% of nickel as alloying elements. This grade is used for structural applications where a stainless steel is required and high strength is needed, which is obtained by cold rolling after an intermediate annealing. In addition, Type 304 stainless steel is an austenitic grade that can be severely deep drawn. This property has resulted in AISI 304 being the dominant grade used in architectural applications.

The deformation-induced martensite transformations have been studied for many years and it is known that the austenite structure or phase is unstable during cold deformation and breaks down to the much stronger, less ductile martensite phase. So, the stainless steel SS304 is a metastable austenite material and its crystallographic structures can transform to martensite phase under plastic deformation. The presence of martensite phase produces significantly different properties compared to the austenitic steel, both in plastic deformation and failure. The phase transformation changes and disorders the microstructure of grains, resulting in resistance to dislocation motions. The material with martensite phase has higher strength and less ductility [1-2].

In this context, it is necessary to improve the basic knowledge about cold rolling for fcc structure, as well as the effect on microstructure. This required the use of techniques such as electron channelling contrast imaging, ECCI (backscattered electron Mott detection [3, 4]), which allows the analysis of variations in sample structures, and electron backscatter diffraction (EBSD) to study textures and secondary phases [5].

In this study, cold rolled AISI 304 (EN 1.4301) steel samples, subjected to different degrees of deformation controlled by thickness reduction. An undeformed sample, with final conventional annealing treatment, was used as a reference. The resulting samples were first analysed by using conventional techniques available in stainless steel work in order to determine bulk composition, mechanical properties, structure and grain size. In a second step, textures and secondary phases were characterized by EBSD [6-9], and crystallographic orientation and defects were studied by ECCI.
(backscattered electron Mott detection [10, 11]). The correlation between strain level obtained, mechanical properties, structure and EBSD results are discussed.

2. Experimental procedure
In this study, EN 1.4301 steel samples of thickness 0.70 - 0.75 mm, taken from the daily production of ACERINOX EUROPA SAU (www.acerinox.es), with the same chemistry, were submitted to different degrees of deformation as determined by thickness. The rolling reduction was 22.0 % (R1) and 37.5 % (R2). As a reference, another sample without deformation and with final conventional annealing treatment is taken. The summary of the samples is listed in table 1.

| Sample Id | Final thickness (mm) | Rolling reduction (%) | Content of martensite (%) | Percentage of not indexed EBSD patterns |
|-----------|----------------------|-----------------------|---------------------------|----------------------------------------|
| A         | 0.70                 | 0                     | 0.04                      | 1.1                                    |
| R1        | 0.70                 | 22.0                  | 2.4                       | 16.1                                   |
| R2        | 0.75                 | 37.5                  | 4.3                       | 26.7                                   |

The chemical composition of the samples was analysed depending on the element, specifically Si, P, Ti, Cr, Mn, Co, Ni, Cu, Nb, Mo and Sn were analysed by X-ray fluorescence (XRF) using a PANALYTICAL PW 2606 XRF spectrometer (www.panalytical.com); finally, C and S, and N were analysed using LECO CS 600 and TC 600 analysers, respectively (www.leco.com).

The mechanical tests were carried out using different equipment, depending on the parameters to be measured. For the study of high-tensile strength, yield strength, elongation, coefficient of plastic anisotropy and hardening coefficient, a RK100 ROELL+KORTHAUS Universal Testing Machine was used; to obtain the Erichssen, KW1 and LDR parameters, a ZWICK ROELL BUP600 drawing machine (www.zwick.com) was used.

Volume fraction of ferromagnetic phase along the surface was measured with a Feritscope Fischerscope MMS (www.helmut-fischer.es). The measured values were converted to martensite contents with a correlation factor of 1.7 defined by Papula et al. [12]; the results are shown in table 1.

Grain size analysis was performed after polishing and etching the samples (Vilella’s reagent), for 60 to 75 s. The observation and measurement of grain size were performed with an OLYMPUS GX71 optical microscope equipped with an “AnalySIS Inclusion Inspector” image analysis system (www.olympus.co.uk).

The characterisation by EBSD and BSE Mott detectors was performed on a ZEISS ULTRA 55 FEG-SEM (www.zeiss.es), using polished longitudinal sections of the samples. The sample sections were prepared by conventional metallographic procedures, with a final polish with colloidal silica suspension, OPS (www.struers.com). The FEG-SEM used is equipped with an HKL CHANNEL 5 EBSD system, from Oxford Instruments (www.oxinst.com). Backscattered electron (BSE) images were obtained at an accelerating voltage of 20 kV and a working distance of 1-2 mm; EBSD maps were acquired at 20 kV and a working distance of 10 mm.

3. Results and discussion

3.1. Composition, structure and mechanical properties
The materials selected for this study are summarized in Table 2, together with their global chemical composition and type of annealing treatment applied. The composition of the samples is typical for this grade, with lower contents of interstitial elements such as C and N.
Table 2. Composition, annealing treatment and metallurgical properties of the materials.

| Sample Id | Chemical composition (wt%)* | Treatment | Mechanical tests | Grain size |
|-----------|-----------------------------|-----------|-----------------|------------|
|           |                             |           | Rm (Mpa) | Rp (Mpa) | A (%) | OM (ASTM) | EBSD (ASTM) |
| A         | C: 0.049; N: 0.051; Si: 0.35; Cr: 17.7; Ni: 8.0; Mo: 0.30; Cu: 0.32 | Annealing | 638 | 274 | 59 | 8.0 | 9.9 |
| R1        | C: 0.046; N: 0.048; Si: 0.33; Cr: 17.7; Ni: 8.1; Mo: 0.26; Cu: 0.32 | Rolling | 1,004 | 857 | 14 | 8.7 | 15.3 |
| R2        | C: 0.049; N: 0.050; Si: 0.37; Cr: 17.7; Ni: 8.1; Mo: 0.31; Cu: 0.37 | Rolling | 1,108 | 929 | 11 | 8.7 | 15.3 |

*Other elements in their residual conventional levels.

Optical microscope images showing the structure of samples A, R1 and R2 are displayed in figures 1, 2 and 3 respectively. The structure of material A shows complete re-crystallisation, with pits at grain boundaries (distribution of carbides within the grains well differentiated); material R1 can be seen to show a partial re-crystallisation with texture marked and distortion preserved; and in material R2 the texture is very marked and there is more evident deformation.

The results of the mechanical tests performed on the A, R1 and R2 samples are listed in table 2, which include the tensile strength, Rm, the yield strength, Rp0.2 [13], and the elongation test, (A). The results obtained for parameters Rm and Rp0.2 indicate that these properties are directly proportional with rolling reduction. In addition, as it is known, the A parameter, which gives valuable information on plastic deformation, is correlated with rolling reduction (inversely proportional) in these materials.
The results of grain size measurements obtained by conventional optical microscopy (OM) and EBSD are listed in table 2 [14, 15]. EBSD measurements respond only to variations in the relative misorientation between adjacent areas, and they are not affected by the action of chemical reagents as in conventional analysis. Grain sizes obtained by OM and EBSD are correlated [16] but not closely agree with each other.

3.2. High angle BSE information
Backscattered electron Mott detection provides information on crystallographic orientation and defects in the material under study [17]. ECCI images are usually obtained by tilting the sample, which increases the BSE yield and enhances electron channelling. High-angle BSE information is used to locate areas of tension, dislocations, or other crystalline defects [18, 19].

Here it should be noted that the high accelerating voltage (20 kV) and the short working distance (1 - 2 mm) used allow the observation of fine structures in grain boundaries, which largely coincide with the dislocations that can be observed by transmission electron microscopy (TEM). In these conditions, tilting the sample is not required to get the ECCI effect.

Figures 4 and 5 show general BSE micrographs corresponding to the reference sample, A. The contrast observed is the result of the different crystallographic orientation of the grains with respect to the impinging electron beam (different zone axis). For all the samples, a crystalline structure of equiaxed grains randomly oriented can be observed. Twins are clearly observed.
Figures 6 and 7 show general and detailed BSE micrographs corresponding to the rolling sample R1. The grains present changing contrast due to stress associated with the presence of dislocation networks by accumulated tension in grain boundaries, which present complex structures of shear and slip bands [20-23].

![Figure 6. ECCI (backscattered electron Mott detection) of material R1.](image1)

![Figure 7. Detail image of material R1.](image2)

Figures 8 and 9 show general and detailed BSE micrographs corresponding to the rolling sample R2. In addition to the grain stress contrast, shear bands and other line defects can be observed. These line defects have resulted in dislocation networks defining sub-grains. In comparison to the reference sample, twin content has been considerably reduced, probably by deformation twin [24].

![Figure 8. ECCI (backscattered electron Mott detection) of material R2.](image3)

![Figure 9. Detail image of material R2.](image4)

Therefore, the density of crystalline defects increases with plastic deformation in the grains, especially dislocations. This results in a complex sub-grain structure, which is generated in response to the deformation.
3.3. EBSD

EBSD analysis allowed obtaining information of the whole sample thickness for the main phase, as the observation plane was ND-LD (ND and LD stands for normal direction and line, or rolling, direction). The FEG-SEM characterisation was carried out using a polished sample in longitudinal section (ND-LD plane). After the analysis, a cleaning procedure was applied to the data, which consisted of removing EBSPs that could not be indexed and isolating points that were incorrectly indexed and appeared as Wild Spikes. The removed points were filled in using copies of their neighbouring points (i.e., if a given point had more than the minimum number of indexed neighbours (5-6), then it was replaced by the most common neighbouring orientation).

Figures 10 to 12 show grain, sub-grain, and coincidence site lattice (CSL) maps corresponding to the A, R1 and R2 materials. The grain and sub-grain maps are obtained from misorientation angles (sub-grain, 1° - 10°, in aqua and grain boundaries, > 10°, in black). The CSL maps are obtained from a specific coincidence of the crystal lattices across the boundary. This information is needed to define more accurately the grain size by EBSD (table 2) because CSL are not considered as grain boundaries but special boundaries (i.e., \(\Sigma 3\) is the well-known twin boundary in fcc materials). The lines with different colours correspond to CSLs with different \(\Sigma\)-values (\(\Sigma\) is the reciprocal number density of lattice points that are coincident in two adjacent grains), axis and angle.

**Figure 10.** Grain (black colour), sub-grain (aqua colour and CSL maps (CSL colour code: \(\Sigma 3\), red; \(\Sigma 5\), green; \(\Sigma 7\), blue; \(\Sigma 9\), pink; \(\Sigma 11\) yellow; \(\Sigma 41c\), light blue) of material A. BC+GB+CSL; Grid 1127 x 845 pixels; Step 0.5 \(\mu\)m.

**Figure 11.** Grain (black colour), sub-grain (aqua colour and CSL maps (CSL colour code: \(\Sigma 3\), red; \(\Sigma 5\), green; \(\Sigma 7\), blue; \(\Sigma 9\), pink; \(\Sigma 11\) yellow; \(\Sigma 41c\), light blue) of material R1. BC+GB+CSL; Grid 1127 x 845 pixels; Step 0.5 \(\mu\)m.

In the reference material, sample A (figure 10), areas with a high density of sub-grain boundaries are not observed (except due to polishing artefacts in the sample). However, in rolling materials, R1 and R2 (figures 11 and 12), these areas represent almost the entire analysed zone (aqua colour). In addition, these areas with high densities of sub-grains correlate with deformed grains but not with grain areas with preferred orientation.

Table 3 shows the results of the measurements of sub-grains and twin areas by EBSD in all the studied materials, which are the proportion of grain boundaries analysed that correspond to sub-grain and twin boundaries.
Table 3. Sub-grain and twin area.

| Sample Id | Sub-grain area (%) | Twin area (%) |
|-----------|--------------------|---------------|
| A         | 11.97              | 41.39         |
| R1        | 85.91              | 2.89          |
| R2        | 87.47              | 2.73          |

The sub-grain area increases from material A to R2, being much higher in the rolled samples than in the annealed one. The twin area decreases from material A to R2, being much higher in the annealed sample than in the rolled ones.

Figures 13 to 15 show maximum misorientation per grain, or strain contouring, for each material. This is a component that provides an estimate of the extent of deformation, or strain, in individual grains in an area. The component measures the maximum misorientation between any two points in a grain and then weights this grain according to this misorientation value. The maps show variations in the amount and colour (from green to red) of area with misorientation per grain, which increases from sample A to R2. Red areas, observed mainly in sample R2, indicate higher values of strain in the samples.
Table 4 shows the results of the measurements of maximum misorientation per grain by EBSD in all the studied materials. The strain contouring increases from material A to R2, according to the higher deformation in the samples.

| Sample Id | Max. misorientation (deg) |
|-----------|---------------------------|
| A         | 5                         |
| R1        | 11                        |
| R2        | 16                        |

Figures 16 to 18 show local misorientation average angle histograms for each material. This component is for displaying small orientation changes in the map, highlighting regions of higher deformation. It calculates the average misorientation between each pixel and its surrounding pixels, and assigns the mean value to that pixel. Misorientations over a certain value are discarded, so that the misorientations associated with discrete sub-grain and grain boundaries are excluded.

Figure 15. Strain contouring map of material R2. SC; Grid 1127 x 845 pixels; Step 0.5 µm.

Figure 16. Local misorientation average angle histogram of material A.

Figure 17. Local misorientation average angle histogram of material R1.
Table 5 shows the results of the measurements of local misorientation average angle by EBSD in all the studied materials. The local misorientation increases from material A to R2, corresponding to the higher deformation in the samples.

| Sample Id | Local misorientation (deg) |
|-----------|-----------------------------|
| A         | 0.15                        |
| R1        | 1.65                        |
| R2        | 1.85                        |

3.4. Correlations
Figures 19 to 22 show correlations between properties of each material studied by conventional techniques (mechanical tests and ferritoscope) and EBSD measurements. Figure 19 shows that the percentage of non-indexed EBSD patterns (see the data cleaning procedure outlined in 3.3.) and the content of martensite calculated for each material are directly proportional with rolling reduction. Also, the elongation calculated in each material is inversely proportional to rolling reduction. Figure 20 shows the correlation between rolling reduction and the mechanical properties tensile (Rm) and yield strength (Rp). Figure 21 illustrates that the content of martensite is directly proportional with the percentage of non-indexed EBSD patterns. Figure 22 demonstrates that the yield strength is directly proportional with the average local misorientation (as measured by EBSD).

4. Conclusions
The analyses performed by means of the EBSD technique as well as the results from conventional tests, have shown that AISI 304 austenitic stainless steel after different rolling reduction present an increasing degree of deformation. This is characterized by slip bands, deformation twins (parallel plates characteristic for martensite), strain hardening and the formation of martensite phase grains, which divide the elongated grains of austenite.

Plastic deformation during the rolling reduction of the samples induces a martensitic transformation ($\gamma \rightarrow \alpha'$) over the whole range of applied deformations.

The deformation structure is characterized by elongated austenite grains with $\alpha'$-phase plates and by deformation twins.
Comparison between the materials subjected to rolling reduction and the reference sample, with conventional annealing and without deformation, show correlations between the strain level obtained, mechanical properties, structure and EBSD (linear relation between yield strength and local misorientation angle) as follows:

- There is a correlation of the rolling reduction with mechanical properties, amount of martensite (in percent) and percentage of non-indexed EBSD patterns. The increase in proportion of martensite in percent (A to R2 material) is linearly related to percentage of non-indexed EBSD patterns with a correlation very close to 1, so martensite proportionally increases with deformation.
- There is a correlation between the measurements by means of OM and SEM, both methods evidence very marked texture and lattice rotation in the rolling materials.
- By OM and SEM, the density of martensite needles increases with strain by plastic deformation, therefore, it is confirmed that the volume fraction of martensite formed is a function of the amount of deformation.
- By EBSD, the local misorientation angle presents a linear relationship with the yield strength (Rp). In addition, the strain contouring maps show that the maximum misorientation per grain increases from sample A to R2, while keeping constant the grain size in the samples R1 and R2.

Figure 19. Correlation between rolling reduction % and percentage of non-indexed EBSD patterns, elongation (A) and proportion of martensite in percent.

Figure 20. Correlation between rolling reduction % and tensile (Rm) and yield strength (Rp).

Figure 21. Correlation between proportion of martensite in percent and percentage of non-indexed EBSD patterns.

Figure 22. Correlation between yield strength (Rp) and average local misorientation.
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