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Article

Martensitic Transformation in Ultrafine-Grained Stainless Steel AISI 304L Under Monotonic and Cyclic Loading

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Abstract: The monotonic and cyclic deformation behavior of ultrafine-grained metastable austenitic steel AISI 304L, produced by severe plastic deformation, was investigated. Under monotonic loading, the martensitic phase transformation in the ultrafine-grained state is strongly favored. Under cyclic loading, the martensitic transformation behavior is similar to the coarse-grained condition, but the cyclic stress response is three times larger for the ultrafine-grained condition.

Keywords: martensitic transformation; UFG; cyclic loading; AISI 304L; stainless steel; equal channel angular pressing; tensile test

1. Introduction

Stainless steels are among the most widely used materials in structural applications and are very well characterized. Especially metastable austenitic steels gained a lot of interest as these offer the possibility to tailor the strength and ductility by introducing different ratios of soft austenite and hard martensite via heat treatments. This phase transformation also enhances the fatigue life significantly, as crack propagation is retarded due to local strengthening and crack closure effects [1,2]. These effects make this material perfectly suited for structural applications where high damage tolerance is required, *i.e.*, for usage as pipes or reactor pressure vessels in power plants [3,4].

It is well known that the strength of metallic materials can be enhanced by the reduction in grain size to an ultrafine-grained (UFG) condition.

In the literature, investigations on fine-grained or ultrafine-grained metastable steels [2,5–7] produced by the so called reverse transformation in annealing treatments [2,5,6] can be found. Another possibility for the introduction of an ultrafine-grained microstructure is severe plastic deformation (SPD). Among the different SPD techniques, equal channel angular pressing (ECAP) [8–12] is the most widely used. The advantage of SPD processing is that nearly all metallic materials can be processed to obtain ultrafine-grained microstructures. In contrast to reverse transformation, SPD processing is nearly independent of the chemical composition of the steel. Due to the advantages, SPD processing gained industrial attention and upscaling of SPD processes is currently a major field of research. Both processes, reverse transformation and SPD processing, lead to similar grain sizes but significantly different microstructures in terms of defect concentrations, dislocation density, chemical and grain size homogeneity, *etc*. To date, there have only been a few investigations reported on homogeneous metastable austenitic steels processed by SPD, e.g., [7], where the authors primarily investigate the processing-microstructure relationships. Almost two decades ago it was already shown by Maier and Bayerlein *et al*. [13–15] in low cycle fatigue (LCF) tests that the coarse grained (CG) condition of the AISI 304L is not stable under cyclic loading at room temperature (RT) and a martensitic transformation occurs if the plastic strain amplitude $\frac{\Delta\varepsilon_{pl}}{2}$ exceeds 3×10^{-3} [15]. The transformation rate increases with increasing plastic strain amplitude and decreasing temperature [13].

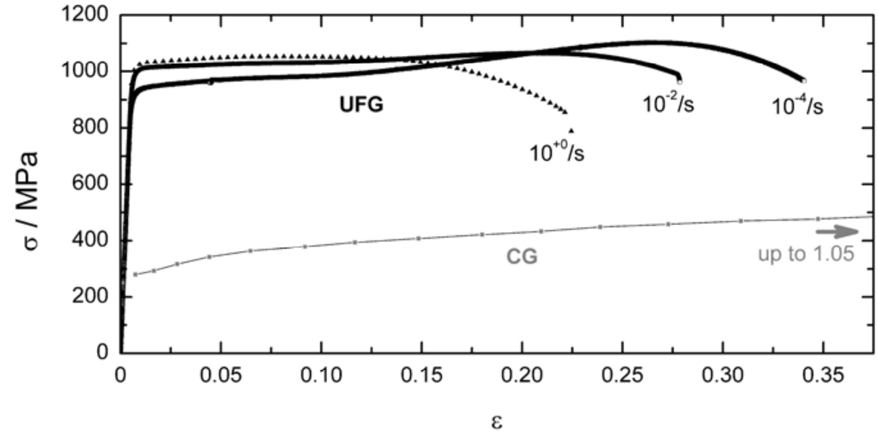
In the high cycle regime, where this threshold amplitude is not reached, cyclically-induced martensitic transformation due to local strain concentrations is reported by Krupp *et al*. [16]. Additionally, it was already shown that the fatigue strength and the fatigue life are strongly dependent on the grain size. This was demonstrated by [17,18] for 304L with grain sizes in the range from 2 to 50 µm, where the fatigue life is improved with decreasing grain size.

Thus it is of major importance for potential industrial applications of SPD-processed ultrafine-grained 304L whether the transformation behavior is influenced by an ultrafine-grained microstructure (<1 µm) introduced by SPD. Therefore, the monotonic and cyclic deformation behavior, as well as the microstructural changes in ultrafine-grained stainless steel AISI 304L, has been investigated. Special emphasis is put on the martensitic transformation behavior.

2. Monotonic Loading

The *monotonic* stress-strain curves of the UFG AISI 304L are presented in Figure 1. Compared to the CG condition [19], the UFG condition exhibits nearly a three times higher yield strength and also a much higher ultimate tensile strength. Additionally, a secondary hardening regime due to significant martensitic transformation occurs after 10% of strain which is not observed for the CG condition where only very little martensitic transformation occurs at very large strains [3,19]. The ductility in the UFG condition is decreased compared to the CG condition, but still exceeds 30% for tests at similar strain rates.

Figure 1. Tensile true stress-true strain diagram of coarse grained (CG) and ultrafine-grained (UFG) AISI 304L, performed at three different nominal strain rates of $10^{-4}\cdot s^{-1}$, $10^{-2}\cdot s^{-1}$ and $10^{+0}\cdot s^{-1}$. Data for the CG condition has been recompiled from [19].



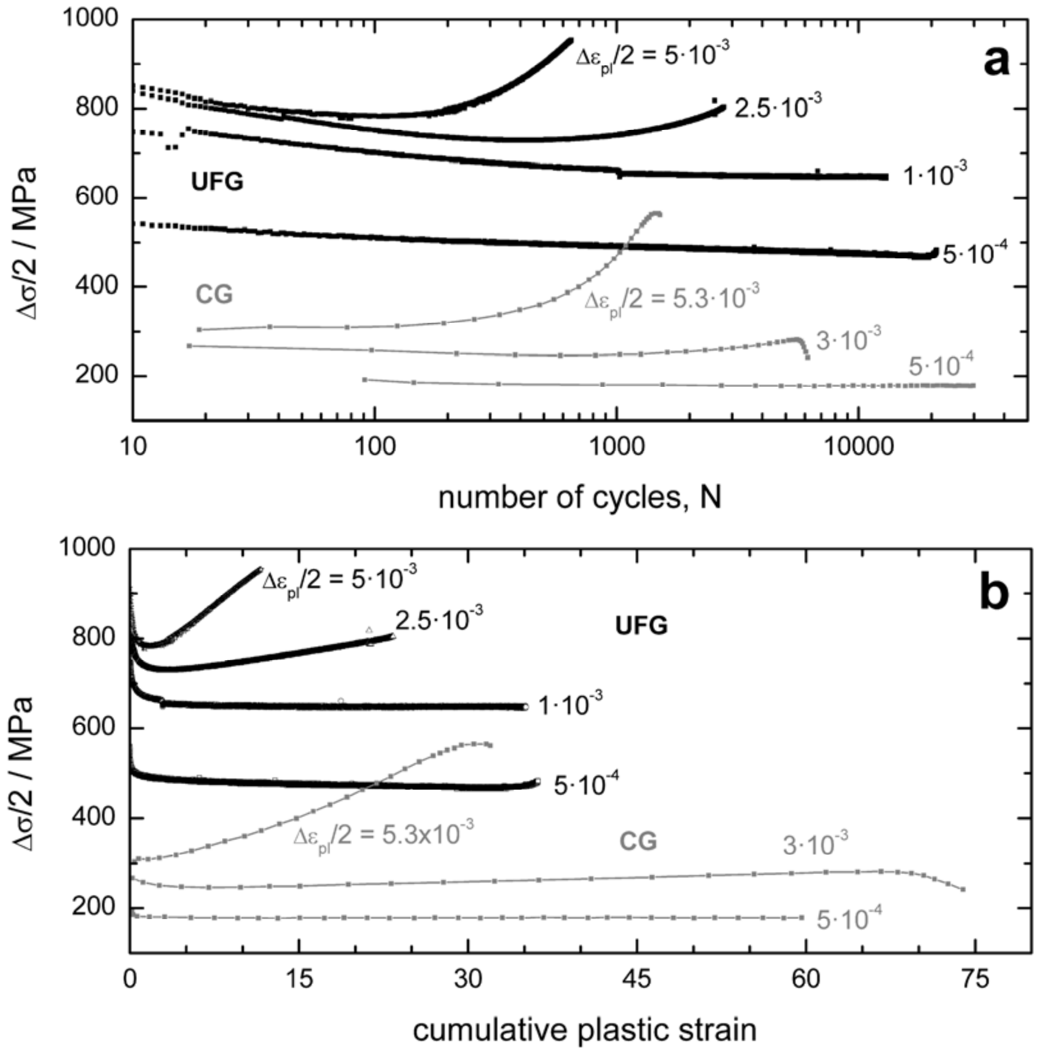
The UFG material shows a strain rate sensitive behavior, which is typical for UFG metals, and the yield strength increases with increasing strain rate and the ductility decreases (Figure 1). For all strain rates applied, the material exhibits the UFG-typical initial hardening up to a plateau value. Upon further straining significant secondary hardening is observed for the test at the lowest strain rate of 10^{-4} s^{-1}. This hardening seems to be based on martensitic transformation as it is in good agreement with earlier studies by Hamada *et al.* [2,20] on CG 301LN. This hardening is still prominent but much less pronounced at the medium strain rate of 10^{-2} s^{-1} and not present for the highest strain rate of 10^{+0} s^{-1}. The onset appears to be independent of the strain rate at about 10% true strain. Thus, the martensitic transformation rate decreases with increasing strain rate.

3. Cyclic Loading

The results of the *fatigue* tests are shown in Figure 2. Under cyclic loading, martensitic transformation occurs at the higher plastic strain amplitudes while at the lower plastic strain amplitudes it is absent. In order to show the influence of the plastic strain on the transformation behavior, the cyclic stress response is additionally presented as a function of the cumulative plastic strain, $\varepsilon_{cum,pl} = 2*N*\Delta\varepsilon_{pl}$.

The cyclic deformation curves depicted in Figure 2a show that the UFG condition is up to three times stronger than the CG condition at similar test conditions. The fatigue lives of the UFG condition are nearly as long as those of the CG condition [14]. For all amplitudes tested, the UFG condition shows pronounced initial softening due to cyclic recovery after the extreme pre-deformation during SPD processing, see Figure 2a,b, which is not found in the CG condition.

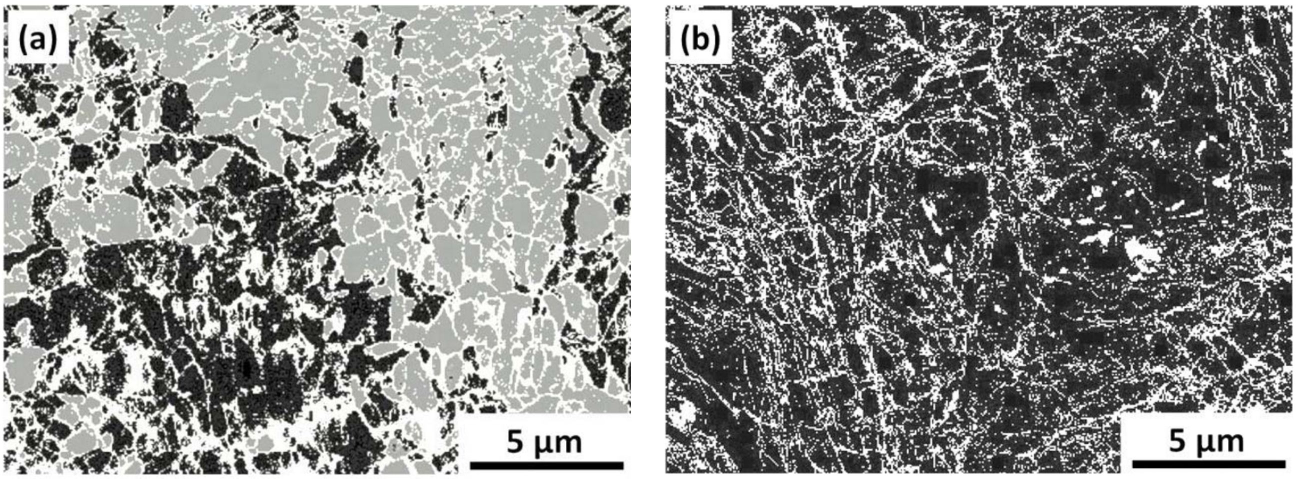
Figure 2. Stress amplitude as a function of the number of cycles (**a**) and of the cumulative plastic strain (**b**) for the UFG condition (black lines). Data for the CG condition (grey lines) has been recompiled from [14].



With increasing plastic strain amplitude initial softening becomes more pronounced. For higher cumulative plastic strains, the influence of the plastic strain amplitude becomes stronger.

At the lowest plastic strain amplitude used ($\frac{\Delta\varepsilon_{pl}}{2} = 5 \times 10^{-4}$) the material softens continuously with a very low rate and no steady state condition was reached during the fatigue test of the material. At intermediate plastic strain amplitude ($\frac{\Delta\varepsilon_{pl}}{2} = 1 \times 10^{-3}$), a stable plateau value was observed. At large amplitudes, *i.e.*, $\frac{\Delta\varepsilon_{pl}}{2} > 1 \times 10^{-3}$, the cyclic deformation curves show a minimum in the stress response, which is shifted to smaller cumulative plastic strains for further increasing plastic strain amplitudes. At the same time secondary hardening becomes more prominent, see Figure 2b. Subsequently, the test revealing a constant stress response is assumed to reflect the threshold value for the onset of martensitic transformation at this specific strain rate of $\dot{\varepsilon} = 1 \times 10^{-2}$ s^{-1}. These results are supported by the microstructural investigations of the lowest and highest plastic strain amplitude via phase contrast measured by EBSD, depicted in Figure 3.

Figure 3. Phase contrast images obtained by EBSD showing the microstructure evolution following tests at the largest plastic strain amplitude of 5×10^{-3} (**a**) and smallest plastic strain amplitude of 5×10^{-4} (**b**). Black = austenitic phase, light grey = martensitic phase.



The martensitic fraction increases considerably in the fatigue tests with large plastic strain amplitudes, as shown in Figure 3a for the test at a plastic strain amplitude of $\frac{\Delta\varepsilon_{pl}}{2} = 5 \times 10^{-3}$. The microstructure shown in Figure 3b depicts a nearly fully austenitic condition. The image is from the UFG 304L after fatigue with the smallest plastic strain amplitude and it is very similar to the material directly after ECAP processing (not shown for the sake of brevity).

Additionally it is shown that slight coarsening occurs alongside the martensitic transformation which is in good agreement with the observed softening for high plastic strain amplitudes (Figure 2a).

4. Discussion

The obtained monotonic as well as the cyclic results of the UFG condition indicate strain-induced transformation of the soft austenitic to the strong martensitic phase.

Under *monotonic loading* it could be shown that the hardening seems to be related to the martensitic phase transformation which strongly depends on the applied strain rate. This can be related to the temperature sensitivity of this material [15,20,21]. With increasing deformation rate, adiabatic heating becomes more pronounced and can reduce or even fully suppress the martensitic transformation. This also explains that only a small fraction of martensite was found in the heavily deformed ECAP condition as the ECAP process was performed at 400 °C. This is in good agreement with work by Huang *et al*. [7] who investigated the influence of ECAP processing temperature on the monotonic mechanical behavior of the AISI 304L. In addition, the ductility of the material decreases with increasing strain rate. Thus, the test at the highest strain rate failed already before the necessary cumulative plastic strain level for the onset of the martensitic transformation of about 10% was reached.

For the CG condition, no secondary hardening is observed for the 304L [3,19]. Still, martensitic transformation can occur to a very small extent at very high plastic strains [19]. In other metastable austenitic steels like CG 301LN, significant martensitic transformation is present already at similar low strains leading to a secondary hardening regime as shown in the studies by Hamada *et al*. [2,20]. On the one hand this behavior can be attributed to the sensitivity of transformation activity to minor changes in chemical composition as the chemical composition influences the probability of the formation of deformation bands [2,20–22]. The results of the present study show that the reduction in grain size reduces the influence of the chemical composition significantly. This is in good agreement with Di Shino *et al*. [17,18] and Hamada *et al*. [2] who showed that the reduction in grain size down to 2 µm has a strong effect on the fatigue strength due to the suppressed formation of deformation bands. Thus chemical compositions which promote or hinder the formation of such deformation bands are no longer dominant if the microstructure is transformed to the ultrafine-grained regime by SPD processing. On the other hand, the number of initially stored transformation nuclei has to be taken into account. Krupp *et al*. [16] showed in high cycle fatigue tests (HCF) that local straining related to high anisotropy due to grain misorientations have to be considered as transformation nuclei. This perfectly fits to the observations in the ultrafine-grained condition, where the mean grain size is reduced drastically and the number of anisotropy related transformation nuclei is increased significantly.

This is additionally supported by the very high dislocation density introduced by SPD processing. For the investigated UFG condition, 10% of strain is needed to provide the conditions for the onset of the martensitic transformation. The relevant microstructural features could be strain concentrations and a high density of stacking faults [14] near grain boundaries, which are typical for UFG materials [23–25]. The formation of these structures is strain rate sensitive and thus may additionally influence the occurrence of martensitic transformation.

Under *cyclic loading*, the phase transformation is observed for both, the CG and UFG condition in AISI 304L. The initial stress-strain response is, however, different. In contrast to the CG condition the UFG material exhibits strong initial softening which seems to be based on recovery of the high dislocation density introduced by SPD processing as no significant grain coarsening is observed.

The threshold amplitude of $\frac{\Delta\varepsilon_{pl}}{2} = 2.5 \times 10^{-3}$ for the onset of the phase transformation in the UFG condition is very similar to the value determined by Bayerlein *et al*. [14]. Similarly, both the UFG and CG conditions show that the hardening rate, and thus, the transformation rate increases with increasing plastic strain amplitude. Moreover, the onset of the martensitic transformation occurs at similar levels of cumulative plastic strain for the UFG and CG condition. This indicates that the transformation nuclei are formed by the local accumulation of plastic strain and—in contrast to the observations under monotonic loading—are not dominated by the ultrafine-grained microstructure or the high dislocation density introduced by SPD processing. This is supported by the strong initial softening due to recovery under cyclic loading prior to the onset of martensitic transformation, see Figure 2a,b.

For both monotonic and cyclic loading a minimum amount of (cumulative) plastic strain is needed for the evolution of the martensitic microstructures. Furthermore, the cyclic tests show that not only the absolute amount of plastic strain but the plastic strain amplitude needs to exceed a threshold value. It has to be pointed out that these thresholds are only valid for this particular test conditions as it is known that the thresholds disappear at low temperatures [14,21].

5. Experimental

Metastable austenitic stainless steel AISI 304L (CrNi18-9) was investigated in an ultrafine-grained condition. The material was ECAP processed at the Institute of Physics of Advanced Materials (Valiev) at the SEI HPE Ufa State Aviation Technical University (Russian Federation). Cylindrically shaped samples with a diameter of d = 20 mm and a length l = 110 mm were pressed up to 4 passes using Route B_c at 400 °C in a die with a 90° angle. All mechanical tests were conducted with the loading axis parallel to the processing direction. Details of the ECAP process can be found in [11,12]. From these billets cylindrical shaped specimens with a gauge section of 4.5 mm in diameter and a length of 10 mm were machined.

All samples were ground down to 5 µm prior to the mechanical tests. For the microstructural investigations the samples were sectioned transverse to the loading direction. The samples were ground and polished down to 1 µm and subsequently electro polished using a perchloric acid solution.

Tensile tests were performed at RT on a servo hydraulic testing system MTS810 in displacement control with additional measurement of the strain in the gauge length of the samples up to a strain of 4% using an extensometer with a gauge length of 8 mm. After 4% of strain the extensometer was removed from the sample and the tests were continued up to 25% stress drop after reaching the ultimate tensile strength. The deformation rates in displacement control were chosen to achieve mean nominal strain rates of $10^{-4}{\cdot}s^{-1}$, 10^{-2} $s^{-1}$and $10^{+0}{\cdot}s^{-1}$. True stress and true strain were calculated from the load-displacement data. All strain rates were tested at least two times.

The cyclic deformation behavior was investigated in symmetric push-pull fatigue tests (R = −1) on a servo hydraulic testing system, MTS810. All tests were performed at room temperature in total strain control with a mean total strain rate of $\dot{\varepsilon} = 1 \times 10^{-2}\ s^{-1}$ with limited plastic strain amplitudes in the range of $\frac{\Delta\varepsilon_{pl}}{2} = 5 \times 10^{-4}$ to $\frac{\Delta\varepsilon_{pl}}{2} = 5 \times 10^{-3}$.

Microstructural *investigations* were carried out in a high resolution SEM, a Zeiss ESB 1540 SEM equipped with an electron back scatter diffraction unit using a step size of 0.05 µm.

6. Conclusions

In the current work it could be shown that the reduction in grain size down to the UFG regime influences the martensitic transformation behavior of the AISI 304L significantly under monotonic loading. In contrast to the CG condition the martensitic transformation occurs at much smaller plastic strains of about 10%. The transformation is much more pronounced, indicated by a secondary hardening regime. These observations can be attributed to the increased number of transformation nuclei in the ultrafine-grained condition. The large fraction of highly misoriented grains as well as typical dislocation arrangements in UFG materials act as strain raisers or transformation nuclei. Under cyclic loading, no significant differences could be observed between the CG and UFG condition regarding the martensitic transformation or fatigue life, although the stress amplitude is about a factor of three higher for the UFG material at similar plastic strain amplitudes. Consequently, the active transformation nuclei under cyclic loading seem to be independent of the ultrafine-grained microstructure.

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