Static Recrystallization Behavior and Mechanical Properties of Heterogeneous Nanostructured Duplex Phase Stainless Steel

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Static recrystallization (SRX) behavior and tensile property of a 92% cold-rolled duplex phase stainless steel were investigated. Lamellae structure, in which austenite (γ) and ferrite (α) phases were complicatedly stacked, was developed by heavy cold rolling. The lamellae gradually changed to equi-axed fine grains during annealing at 1023 K and fully SRXed after 1.44×10^4 s (4 h). Because of precipitation and its impediment of grain-boundary migration, grain coarsening was strongly suppressed even by prolonged annealing to 2.59×10^5 s (72 h). While volume fraction of γ phase was about 30% before annealing, it drastically increased up to 90%. The transformed phase from γ to α by heavy cold rolling re-transformed to stable equilibrium γ at 1023 K and occurrence of SRX. No softening took place even with the occurrence of SRX and exhibited quite high Hv hardness of 5.9 GPa due to precipitation, which effectively suppressed grain growth. Tensile strength was barely changed before and after annealing and stayed about 1.5 GPa on average, while ductility rapidly decreased. [doi:10.2320/matertrans.MT-M2019302]

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1. Introduction

A lot of studies on severe plastic deformation (SPD) have been carried out^{1–7)} on how to attain ultrafine-grained (UFGed) metallic materials to achieve various specific properties as well as high strength. Sample sizes of UFGed metallic materials produced by means of SPD methods are, however, essentially too small to employ as structural ones. New innovative methods to fabricate samples large enough for structural components are therefore needed. The average grain size achieved by SPD methods is approximately 0.1~0.2 μm at minimum, comparable to that of the subgrain. This suggests that the grain-refinement mechanisms taking place during SPD should be identified as continuous dynamic recrystallization, in which dislocation accumulation on to sub-boundaries and a gradual increase in the misorientation angles finally develops UFGs composed of high-angle grain boundaries.³⁾

It is known that mechanical twinning dramatically accelerates grain refinement.⁶⁾ This is because mechanical twinning having boundary spacing around 20 nm or less can instantly subdivide coarse initial grains. By employing nanometer-sized mechanical twinning in a Cu–Zn alloy, a homogeneous nano-structured microstructure with an average grain size of 20 nm can be successfully fabricated by multi-directional forging.⁷⁾ Applying a similar concept for grain fragmentation, dense introduction of mechanical twins can be carried out by simple heavy cold rolling of Cu–Be alloy, resulting in a successfully developed UFGed structure with extraordinary high strength.⁸⁾ This suggests that the evolution of UFGed structures is drastically promoted in metallic materials with low stacking fault energy possibly because of the introduction of dense mechanical twins. Using these methods, Miura *et al.* successfully fabricated UFGed sheet stainless steels, employing mechanical twinning by means of

simple heavy cold rolling.⁹⁾ The heavily cold-rolled stainless steel possessed a specific microstructure subdivided by dense mechanical twinning, which they called a “heterogeneous nano-structure” in which all the microstructures were formed by deformation induced nanometer-scale ones. They exhibited excellent mechanical properties of tensile strength over 2.6 GPa and 5% ductility after ageing. However, UFGed structures are known to be thermally unstable.¹⁰⁾

So far, however, there have been no studies on thermal stability and the static recrystallization (SRX) behavior of the heterogeneous nano-structured stainless steels as far as the authors know. A particular microstructure and mechanical properties originated from the heterogeneous nano-structure may be newly developed. In the present study, SRX behavior and changes in the mechanical properties of heavily cold-rolled duplex phase stainless steel composed of heterogeneous nano-structure are systematically investigated.

2. Experimental Method

A SUS329J3L (DIN1.4462) duplex phase stainless steel with thickness of 6.25 mm was solution treated at 1523 K for 1 h followed by quenching. It was then heavily cold rolled to 0.5 mm thickness, i.e., 92% reduction. The chemical composition is shown in Table 1. The volume fraction of austenite (γ) phase after cold rolling was 70% measured by a ferrite meter. The cold-rolled sample was annealed at 1023 K for 72 h (2.5×10^5 s) in a vacuum furnace. The evolved microstructures before and after annealing were investigated by transmission electron microscopy (TEM) and scanning electron microscopy equipped with electron back scatter diffraction analyzer (SEM-EBSD) after mechanical and electritical polishing. For the observation from transvers direction (TD), foils were prepared using ion milling apparatus. Mechanical properties were examined using micro-Vickers hardness tester and Instron-type mechanical testing machine. Tensile specimens with gauge dimensions of

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Table 1 Chemical composition of a supplied SUS329J3L (DIN1.4462) duplex phase stainless steel in mass%.

C	Si	Mn	P	S	Al	Ni	Cr	Mo	N	O	Fe
0.020	0.47	1.77	0.018	0.0009	0.026	5.50	22.01	3.01	0.1260	0.0018	Bal.

Fig. 1 TEM photographs of (a) partially mixed region of ferrite and austenite, and (b) austenite region. Corresponding selected area-diffraction patterns are also shown using circle numbers. RD and ND indicate rolling and normal directions. The microstructure of ferrite only is also displayed in Fig. 4(a) which was observed at a lower magnification.

$6.0 \times 3.0 \times 0.5 \text{ mm}^3$ were electro-discharge machined from the rolled sheet. Tensile test was carried out parallel to the rolling direction (RD) at an initial strain rate of $2.5 \times 10^{-3} \text{ s}^{-1}$ at room temperature.

3. Results and Discussion

3.1 Microstructure before annealing

Figure 1 displays TEM micrographs of the 92% cold-rolled duplex phase stainless steel. The feature of microstructure was different depending on the phase; low-angle lamellae at α region (Fig. 1(a)) and a typical “heterogeneous” nano-structure at austenite (γ region (Fig. 1(b)).⁹⁾ From the selected area diffraction patterns, specific development of substructure in the α region, and twins and low angle lamellae in the γ could be recognized. That is, simple lamellae with an average boundary spacing of about 200 nm, and having dislocation substructure were developed in the α region, whereas a complex “eye”-shaped twin domain was embedded in low-angle lamellae with boundary spacing of an average 170 nm in the γ region. The latter is known to evolve by mechanical twinning during heavy cold-rolling of both austenitic and duplex phase stainless steels.⁹⁾ The twin-boundary spacing inside of the eye-shaped twin domains was about 80 nm. While it is reported that the eye-shaped twin domain is surrounded by shear bands,⁹⁾ this feature was

Fig. 2 Change in the hardness of the heavily cold-rolled duplex phase stainless steel during annealing at 1023 K. The result annealed at 773 K is also displayed for comparison.⁹⁾

not clearly observed in Fig. 1(b). Therefore, a quite complicated nanometer-sized structure was evolved by alternative stacking of α and γ phase lamellae. Furthermore, their feature drastically changes depending on the phase.

3.2 Annealing behavior and microstructural change

Change in the hardness during annealing at 1023 K was investigated and the result as well as that at 773 K for comparison⁹⁾ is shown in Fig. 2. Hardness rapidly increased to a peak around $1.44 \times 10^4 \text{ s}$ (4 h), and then gradually decreased by prolonged annealing. As will be shown later, SRX extensively took place after $7.2 \times 10^3 \text{ s}$ (2 h) and the observed hardening in spite of SRX was affected by dense precipitation. SRX was not detected at all at 773 K.⁹⁾

Figure 3 shows the summarized results of SEM-EBSD observation. Because of the large strains stored and the high dislocation density, no detected area indicated by black contrast appeared before annealing (Fig. 3(a)). Although the texture appears also to change, the reason is not discussed here because of quite small areas analyzed and insufficient number of grains for statistical analysis. After annealing for $1.44 \times 10^4 \text{ s}$ (4 h), the sample seemed to be fully SRXed and had an average grain size approximately of $2 \mu\text{m}$ (Fig. 3(b)). It was interesting to see that most of the α phase/grains transformed into γ one (Fig. 3(d)).

The evolved microstructure before and after annealing was observed by TEM from TD and the typical microstructures are displayed in Fig. 4. The as-rolled sample showed a typical α lamellae structure with an average boundary spacing of 200 nm (Fig. 4(a)). Of course, some areas were composed of alternative stacking of α and γ phases, as already shown in Fig. 1. However, most of them were changed to equi-axed fine grains by SRX. The phase transformation from α to γ could be recognized by the

Fig. 3 Microstructural changes on the normal plane during annealing at 1023 K observed using SEM-EBSD; IPF maps of (a) before annealing and (b) annealed for 1.44×10^4 s (4 h), and the corresponding phase maps; (c) before annealing and (d) annealed for 1.44×10^4 s (4 h), respectively. α and γ phases are indicated by red and green colors in (c) and (d). Observation and color decoding are parallel to N.D.

Fig. 4 Change in the microstructure during annealing at 1023 K; (a) as rolled, and annealed for (b) 2 h and (c) 72 h.

formation of annealing twins in the SRXed grain interior (Figs. 4(b), (c)) as well as the phase maps in Figs. 3(c), (d). It was revealed by the microscopic observation using TEM that recrystallization was not still completed after annealing for 7.2×10^3 s (2 h). Full SRX was actually achieved after 1.44×10^4 s (4 h) annealing (see Fig. 5). It is also interesting to note that the SRXed grains hardly coarsened even by the prolonged annealing. The average grain sizes after annealing for 7.2×10^3 s (2 h) and 2.59×10^5 s (72 h) are roughly 1 and 2 μm respectively. The above results suggest that a duplex phase stainless steel composed of thermally stable equi-axed fine grains could be attained by occurrence of a truly homogeneous nucleation.

The reason for the increase in the hardness during annealing and SRX was investigated by TEM observation at higher magnification, and a typical photograph is shown in Fig. 5. It can be seen that precipitates with dark contrast were densely distributed in the SRXed grains. Impediment of grain-boundary migration and grain coarsening by precipitates is now well known.¹¹⁾ Our experiment confirmed, therefore, that suppression of grain coarsening as well as hardening during annealing was induced by precipitation. Although the composition and structure of the precipitates could not be clearly identified by TEM and X-ray

Fig. 5 A TEM photograph of the duplex phase stainless steel fully recrystallized after annealing for 1.44×10^4 s (4 h) at 1023 K. Many precipitates can be seen.

diffractometer analyses due to the complicated chemical compositions and overlap of diffraction peaks, it was assumed that they would be σ phase and/or M_{23}C_6 from the studies on precipitates in stainless steels.¹²⁾ We also carried out a semi-qualitative examination about the formation of σ phase at equilibrium state using Thermo-Calc software and it suggested high mole fraction of the σ phase at 1023 K up to 25% and γ phase rest, furthermore, virtually no formation of α phase and M_{23}C_6 . Nevertheless, the results of SEM-EBSD analysis in Fig. 3 indicated the formation of α phase. In addition, the observed rather coarse grains without annealing twins in Figs. 4(b), (c) and Fig. 5 were presumably α grains. This may imply a non-equilibrium state of the fine-grained SRX structure. Finally, the results confirmed that σ phase strongly impeded grain coarsening.

When comparing Figs. 2, 4 and 5, observation showed that the notable hardening in spite of full SRX during annealing was derived from precipitation. The softening that appeared

after 10^5 s should be, therefore, induced by ripening of precipitates to reduce the distribution density in addition to slight grain coarsening. Both a decrease in density of precipitates and grain growth are generally known to cause softening.

It is reported that a UFGed structure exhibits unusual thermal instability and SRX can take place at temperatures well below $0.5T_m$, i.e., much less than half of melting point.¹⁰ However, in the present heterogeneous nanostructured sample, full SRX occurred after annealing for a long time, even at 1023 K ($0.7T_m$). 1023 K is essentially a high enough temperature for SRX to take place in almost all conventional stainless steels. Furthermore, no SRX was observed by the annealing at 773 K.⁹ This indicates that the development of a heterogeneous nano-structure does not spoil thermal stability. If anything, it possesses rather good thermal stability. A superior property would be affected by high thermal stability of twins.¹³

3.3 Change in the tensile properties by annealing

The samples before and after annealing were tensile tested along RD. The flow curves are shown in Fig. 6. The as-cold-rolled sample exhibited quite high tensile strength of about 1.55 GPa. However, this value is lower than the 1.8 GPa attained in a duplex phase stainless steel with same chemical composition and fabricated by simple heavy cold-rolling to 92%.⁹ Miura *et al.* suggested eye-shaped twin domain played an important role in strength.⁹ Aoyagi *et al.* calculated the effect of the eye-shaped twin domain on strengthening by multi-scale simulation, and theoretically showed that strength increases as the volume fraction of the eye-shaped twin domain increases.¹⁴ While we could not compare the difference between the volume fractions of the eye-shaped twin domains in the corresponding two samples, it is reasonable to suppose that the volume fraction of the eye-shaped twin domains in the present sample must be lower than that in the previous report.⁹ Such differences could easily be caused by the change of rolling process because mechanical twinning is one of stress relief mechanisms.

The tensile strength was not largely changed depending on annealing time, and was roughly 1.5 GPa. This tendency

should be because dense distribution of precipitates and still more full SRX occurred by annealing (Fig. 4). However, ductility rapidly dropped with increased annealing time. The loss of ductility was caused by dense distribution and coarsening of precipitates. Nevertheless, it should be noted that the dense formation of σ phase largely contributed to improve thermal stability of fine-grained structure, which effectively impeded grain coarsening even by prolonged annealing at 1023 K and kept the tensile strength to be around 1.5 GPa.

4. Conclusions

- (1) The cold-rolled sample was composed of a complicated nanometer-scale structure where ferrite (α) and austenite (γ) lamellae were alternatively stacked. The boundary spacings of α and γ lamellae were 200 nm and 170 nm, respectively. Further, eye-shaped twin domains were embedded in the γ lamellae.
- (2) Even while annealing at 1023 K caused full recrystallization, it exhibited good thermal stability. The recrystallized grain size was hardly changed, between 1 and $2\text{ }\mu\text{m}$, even by prolonged annealing for 2.59×10^5 s (72 h). Such high-thermal stability was understood from the impediment of grain coarsening by high densely introduced σ phase precipitation.
- (3) Even after recrystallization and annealing for 72 h, the tensile strength was kept around 1.5 GPa, while ductility rapidly decreased.

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Fig. 6 Results of tensile tests of the cold-rolled duplex phase stainless steels before and after annealing at 1023 K.