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# Characterization of the microstructures and the shape memory properties of the Fe-Mn-Si-Cr-Ni-C shape memory alloy after severe plastic deformation by differential speed rolling and subsequent annealing

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ABSTRACT

The Fe-13.51Mn-4.82Si-8.32Cr-3.49Ni-0.15C shape memory alloy was subjected to severe plastic deformation by high-ratio differential speed rolling and subsequent annealing (between 873 and 1373 K) to find the factors that affect recovery stress and recovery strain. As the annealing temperature increased (i. e., as grain size increased), the recovery strain increased. This was attributed to decrease in the density of grain boundaries and the density of twins with increasing grain size. The recovery stress, however, showed a different behavior. It increased with annealing temperature up to 973 K, at which an ultrafine grain size near 1.6 μm was obtained, and then decreased with further increase in the temperature. The difference between the recovery strain and the plastic strain induced by recovery stress under constrained displacement during cooling was proposed to determine the amount of recovery stress. There was an optimum grain size for the largest difference between the recovery strain and the plastic strain. The current study shows that grain-size reduction is not good for having a high recovery strain but it is beneficial for having a high recovery stress in the Fe-Mn-Si shape memory alloys.

## 1. Introduction

Ni-Ti based shape memory alloys (SMAs) have major technological applications in the industrial market, but their high cost has limited their applications in civil engineering structures where a large volume of material is consumed. Fe–Mn–Si SMAs, which have low material and production costs, wide transformation hysteresis, high elastic stiffness and strength compared to Ni-Ti SMAs, have attracted attention for potential use in civil constructions [1], and since 1982, new iron-based SMAs with improved shape memory properties have been developed for this purpose [2–10]. For prestressing applications, the iron-based SMAs have several advantages compared to traditional prestressing technologies because they can prestress concrete without the need for the anchor heads that are required for applying the force with a hydraulic device in prestressing conventional steels [11]. The parent austenite phase γ (FCC) in the Fe-Mn-Si SMAs experiences stress-induced transformation to ε martensite (HCP) during the prestraining. The reversion of this ε to γ by heating produces the shape recovery strain or recovery stress depending on the constraint conditions. The Fe-Mn-Si SMAs typically exhibit smaller recovery strains than the Ni-Ti based SMAs, but the formers are able to generate higher recovery stresses [8,9]. This is important because for using the Fe-Mn-Si SMAs in prestressing applications, a high recovery stress is advantageous.

Grain refinement has been suggested as a strategy to enhance shape memory properties. Severe plastic deformation (SPD) is one of the effective methods for grain refinement. Several researchers have studied the effect of SPD on the shape memory effect of Fe-based SMAs using equal channel angular pressing (ECAP) followed by annealing at various times and temperatures. Zhang et al. [12] obtained an improved shape recovery ratio and recovery stress in an Fe-19.04Mn-4.98Si-8.50Cr-4.59Ni-0.013C (by wt%) alloy through ECAP (with a single pass at 573 K) followed by annealing at temperatures ranging from 573 to 1073 K for 1 h. The best improvement was achieved after annealing at 873 K. Käfer et al. [13] measured the shape recovery ratios of a Fe-7.4Mn-4.6Si-13.12Cr-6.35Ni12.18Co-0.4Ti-0.03C alloy prepared by ECAP (with 1–2 passes at 523 K) followed by annealing at different temperatures ranging from 723 to 1323 K. The best shape recovery was achieved after annealing at 923 K, where the recrystallized grain size was 12–20 μm. Bernardi et al. [14] examined the shape recovery ratios in Fe-14.2Mn-5.3Si-8.8Cr-4.65Ni-0.008C and Fe-8.26Mn-5.25Si-12.8Cr-5.81Ni-0.009C-11.84Co alloys after deformation by ECAP (with a single pass at 523 K) followed by annealing at 923 and 1123 K for 1 h.

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They observed that the alloy without Co had a smaller grain size (30 μm) and exhibited higher shape recovery ratios compared to the Co-containing alloy with larger grain sizes (64–69 μm). Druker et al. [15] examined the shape recovery ratios of a Fe-15Mn-5Si-9Cr-5Ni alloy after ECAP (with a single pass at 523 K) after annealing at 1073, 1223 and 1273 K. They observed the activation of recrystallization above 1073 K and an increase in the grain size from 18 to 53 μm by increasing the annealing temperature from 1173 to 1273 K. Compared to the initial alloy with a coarse grain size of 290 μm, the annealed samples after ECAP exhibited higher shape recovery ratios.

In all above cases, the grain size from the Fe-Mn-Si alloys after ECAP was larger than 10 μm, indicating that grain refinement by ECAP was not sufficient. This likely resulted due to the use of a limited number of ECAP passes (one or two) on the Fe-Mn-Si alloys. Applying more ECAP passes might have been difficult due to the large increase in brittleness and strength of the Fe-Mn-Si alloys with low stacking fault energies only after a few ECAP pass numbers.

Recently, the current authors reported the achievement of ultrafine grains with a recrystallized grain size of 1.5 μm from the Fe-13.51Mn-4.82Si-8.32Cr-3.49Ni-0.15C (wt%) alloy using high-ratio differential speed rolling (HRDSR) combined with subsequent annealing at 973 K [16]. The HRDSR technique, which generates a large shear deformation during asymmetric rolling with a speed ratio ≥ 2, has advantages over ECAP in producing ultrafine grained materials from hard-to-deform metals because it requires a low roll force for a large thickness reduction [17], so that SPD can be applied to materials at low temperatures.

Extending the previous work, we herein examine the effect of annealing temperature on the recovery stress and recovery strain of the HRDSR-processed Fe-Mn-Si-Cr-Ni-C alloy in a wide range of annealing temperature between 873 and 1373 K. Based on the obtained data, we tried to understand how the strength and microstructural defects influence the recovery stress and recovery strain and establish the relation between the recovery stress and recovery strain.

## 2. Materials and Methods

A Fe-Mn-Si based alloy (Fe-13.51Mn-4.82Si-8.32Cr-3.49Ni-0.15C by wt%) ingot was produced by induction melting under vacuum, using high purity iron, manganese, silicon, chromium, nickel and graphite. After homogenization at 1373 K for 13 h under vacuum, the ingots were hot forged into round bars of 15 mm diameter and then hot rolled to a 2.5 mm thickness at 1273 K. The rolled samples were solution annealed at 1273 K for 3 h and then cooled by water quenching. The material obtained after cooling is hereafter referred to as the solution annealed (SA) sample. The SA sample was heated at 673 K for 10 min and then subjected to HRDSR at 423 K. The speed ratio between the upper and lower rollers was gradually increased from 2 to 3.8 by increasing the speed of the upper roller. Between every pass, the sample was annealed at 673 K for 1 min. HRDSR was conducted to a final thickness of 1 mm. After the final pass by HRDSR, the samples were cooled in air. Annealing after HRDSR was conducted at various temperatures between 873–1373 K for 1 h. The HRDSR-processed samples annealed at 873, 973, 1073, 1173 and 1373 K are hereafter referred to as the A873, A973, A1073, A1173 and A1373, respectively. Dog-bone shaped tensile samples (with a width of 2 mm and a length of 18 mm in the gauge region) were cut along the rolling direction (RD).

The recovery stress of the samples was measured after loading to a prestrain of 4% at an initial strain rate of $5 \times 10^{-4}\,s^{-1}$, followed by unloading with a displacement constraint and heating the sample to 523 K at a rate of 1 K/s with a heating gun. After holding the sample at 523 K for 5 min, the sample was cooled to room temperature (298 K) by turning off the heating gun. After removing the recovery stress, the same test was repeated on the same sample again for reliability of measurement. This procedure was repeated 2–3 times at each condition. The recovery strain of the samples were measured after tensile loading to a prestrain of 4%, followed by unloading and heating up to 523 K at a rate of 1 K/s using a heating gun. During heating and cooling, the crosshead of the tensile testing machine was controlled to move to remove any recovery stress (below 5 MPa) that builds up in the sample due to a reverse martensitic transformation. The same test was repeated on the same sample again (for 2–3 times at each condition).

The phase transformation characteristics of the samples were examined using a differential scanning calorimeter (DSC, Perkin-Elmer DSC7) between 173 and 773 K. The specimens were heated from 293 to 773 K, cooled to 173 K and then heated to 773 K at a rate of 10 K/min.

High-resolution X-ray diffraction (XRD) employing Cu Kα radiation at 40 kV was used to analyze the phases of the samples.

Electron back-scattering diffraction (EBSD) analysis with scanning step sizes of 0.1–2.5 μm was used to characterize microstructures on the longitudinal sections of the rolled sheets. For preparation of the sample for the EBSD measurement, the samples were mechanically ground using SiC paper and then ion-milled. The EBSD data were processed using TSL-OIM analysis software, and the data points with a confidence index ≤ 0.1 were removed from the EBSD data. For the HRDSR sample annealed at 973 K, the EBSD measurement was made on the same sample used in the previous work [16], but on a different position.

The microstructures of the HRDSR samples annealed at 1073 K for 1 h before and after the 4% prestraining (without heating) were observed using transmission electron microscopy (TEM, JEM 2001 F). The TEM specimens were prepared using focused ion beam milling.

## 3. Results

The inverse pole figure (IPF) and grain boundary (GB) maps of the HRDSR samples annealed at various temperatures are shown in Fig. 1(a)–(e). The IPFs (normal direction (ND) and rolling direction (RD)) of the HRDSR samples are presented in the insets. All the annealed HRDSR samples show similar textures with a preferential orientation of {110} ⟨111⟩ , which is similar to a typical {110} ⟨112⟩ brass texture for rolled FCC steels. The mean grain size ($d$), fraction of high-angle boundaries ($f_{HAGB}$), density of twins (= total length of twins divided by the examined area) and the density of grain boundaries (= total length of high angle boundaries divided by the examined area) for all the materials, which were obtained or calculated by analyzing the EBSD data, are presented in Table 1. The SA was composed of fully recrystallized grains (with $d$ = 74.04 μm [16] and $f_{HAGB}$ = 0.88). After deformation by HRDSR, the image quality value, which is the quality of the diffraction patterns that indicates the distortion of crystal lattices in the diffraction patterns, decreased substantially, and remnants of the initial grains contained many low angle boundaries [16], indicating the storage of high strain energy through the accumulation of a high dislocation density during the HRDSR process. The measured $f_{HAGB}$ is low as 0.12. The TEM study [16] showed the uniform distribution of microscale shear bands over the entire longitudinal section of the sample after HRDSR. The HRDSR sample annealed at 873 K shows an ultrafine grained microstructure ($d$ = 1.45 μm), but with a relatively low $f_{HAGB}$ (0.65) compared to that for a fully recrystallized microstructure. Beyond annealing temperatures of 873 K, almost fully recrystallized microstructures (with $f_{HAGB}$ = 0.81–0.91) were obtained. Initially, $d$ slowly increased from 1.45 to 2.38 μm during annealing before increasing rapidly above 1173 K. At 1373 K, the grain size increased to as large as 50.17 μm, but it was smaller than that of the SA (74.04 μm). Many {111} type annealing twins, which are a general feature of recrystallized FCC steels with low stacking fault energies, were found within the parent austenite grains in the SA and the annealed HRDSR samples. Fig. 2 shows the density of high angle grain boundaries and the density of twins as a function of annealing temperature. Both densities decrease with increasing annealing temperature (i.e., with increasing grain size).

The DSC curves for the SA, as-HRDSR and annealed HRDSR samples, showing a single endothermic peak for $\varepsilon \rightarrow \gamma$ in each material, are presented in Fig. 3(a) and (b). These peaks appeared in the course of