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Key criterion for achieving giant recovery strains in polycrystalline Fe-Mn-Si based shape memory alloys

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ABSTRACT

In this study, it is proposed that coarsening austenitic grains is a key criterion for achieving giant recovery strains in polycrystalline Fe-Mn-Si based shape memory alloys. In order to verify the hypothesis, the relationship between recovery strains and austenitic grain-sizes in cast and processed Fe-Mn-Si based shape memory alloys was investigated. The recovery strain of cast Fe-19Mn-5.5Si-9Cr-4.5Ni alloy with the coarse austenitic grains of 652 μm reached 7.7% while the recovery strain of one with the relatively small austenitic grains of 382 μm was only 5.4%. Moreover, a recovery strain of 5.9%, which is the highest previously published value for solution-treated processed Fe-Mn-Si based shape memory alloys, was obtained by coarsening the austenitic grains through only solution treatment at 1483 K for 360 min in a processed Fe-17Mn-5.5Si-9Cr-5.5Ni-0.12C alloy. However, its recovery strain was still 5.9% after thermo-mechanical treatment consisting of 10% tensile strain at room temperature and annealing at 1073 K for 30 min. This happens because annealing twins play a negative role, refining the austenitic grains, limiting the recovery strains to below 6%. In summary, coarse austenitic grains enable the achievement large recovery strains by two mechanisms. Firstly, the grains are bigger, and consequently there are fewer grain boundaries, and thus their suppressive effects of grain boundaries on stress-induced ε martensitic transformation is reduced. Secondly, coarse austenitic grains are advantageous to introduce ε martensite with single orientation and reduce the collisions of different martensite colonies, especially when the deformation strain is large. As such, the ceiling of recovery strains is dependent on the austenitic grain-sizes.

1. Introduction

Shape memory alloys (SMAs) exhibit the shape memory effect (SME) and super-elasticity, and thus are a kind of intelligent functional material combining perception and driving functions [1–6]. As such, the SMAs are promising for a wide range of applications in biomedicine, actuation, energy conversion, aerospace, robotics, civil construction, damping, and micro-electromechanical systems (MEMS), among other fields [1,7–12]. Ni-Ti based SMAs possess an excellent SME, *i.e.* a large recovery strain of around 8% [13]. However, they suffer from high processing cost due to low cold workability [1,11]. As an alternative, Fe-Mn-Si based SMAs seem to be more favorable for many applications due to their low cost, good workability, good machinability, and good weldability [14–16]. This field has emerged since Sato *et al.* discovered a giant recovery strain of 9% in a monocrystalline Fe-30Mn-1Si alloy [17]. For the purpose of practical applications, polycrystalline Fe-Mn-Si based SMAs have to be manufactured and are generally subjected to processing techniques, such as forging [18], rolling [19–21], and drawing [22]. Unfortunately, the processed polycrystalline Fe-Mn-Si based SMAs only achieve a low recovery strain of 2–3% after solution treatments at temperatures from 1273 K to 1473 K [18,23–25]. A range of studies have, however, showed that the recovery strains could be improved up to around 5% using training, that is, several cycles of straining at room temperature (RT) and subsequent annealing at 873–923 K [25–28]. In addition to the training, the recovery strains can be enhanced significantly by thermo-mechanical treatments (TMTs), consisting of cold-rolling/deformation at RT and subsequent annealing/aging, and the aus-forming at 973 K [24,25,29–33]. To our knowledge, however, there are no published reports of recovery strains exceeding 6% for processed polycrystalline Fe-Mn-Si based SMAs after treatments such as the training, TMTs and aus-forming.

Recently, Wen et al. [34] demonstrated a bending recovery strain of

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8.4% and a tensile recovery strain of 7.6% in a cast and annealed polycrystalline Fe-20.2Mn-5.6Si-8.9Cr-5.0Ni alloy with coarse austenitic grains of about 1100 μm. This result was a breakthrough in attaining the large recovery strains of above 6% for polycrystalline Fe-Mn-Si based SMAs. There are two key reasons why the giant recovery strain was produced by the simple synthesis-processing of casting and annealing. One reason is that strong interactions occur between annealing twins and stress-induced ε martensite during deformation, but the formation of annealing twins is heavily suppressed by casting followed by annealing. The other is that this cast alloy primarily consisted of coarse austenitic grains. However, it is important to note that a cast Fe-17.5Mn-5.29Si-9.68Cr-4.2Ni-0.09Ti alloy with small austenitic grains reached a recovery strain of just 4.5% [35]. Thus, the above results raise the question of whether coarse austenitic grains play a more crucial role than annealing twins in achieving the large recovery strains of > 6% for polycrystalline Fe-Mn-Si based SMAs.

In this paper, we test the hypothesis that coarsening austenitic grains is a key criterion for achieving a giant recovery strain in Fe-Mn-Si based SMAs. In order to do so, we produced cast Fe-Mn-Si based SMAs alloys with different sized austenitic grains by controlling the solidification rates, and then investigated the effect of austenitic grain-sizes on the stress-induced ε martensitic transformation and the recovery strains. Specifically, we investigated these effects in a processed Fe-17Mn-5.5Si-9Cr-5.5Ni-0.12 C alloy and demonstrated clearly that austenitic grain-sizes determine the recovery strains but that the key parameter is the effective grain-size once the spacing of annealing twins has been taken into account.

2. Criteria of achieving giant recovery strains in polycrystalline Fe-Mn-Si based SMAs

It is beyond doubt that the SME in Fe-Mn-Si based SMAs originates from the stress-induced transformation of γ austenite to ε martensite and its reverse transformation [1]. Therefore, the basic rules for obtaining a good SME are to facilitate the stress-induced ε martensitic transformation and suppress dislocation-mediated plastic slip during deformation, as well as to promote the crystallographic reversibility of the reverse transformation on subsequent heating. To our knowledge, there are four criteria following the basic rules in published literature.

Firstly, composition design or deformation-temperature selection should be done to ensure that the stacking fault energy is as low as possible, in order to facilitate the stress-induced ε martensitic transformation. Generally, deformation mechanisms, including dislocation glide, mechanical twinning and the ε martensitic transformation, depend on the stacking fault energy in austenitic high-Mn alloys [36,37]. Studies indicate that the ε martensitic transformation can occur if the stacking fault energy is below 18 mJ/m^2; mechanical twinning can take place if the stacking fault energy is in the range of 12–35 mJ/m^2; and therefore both ε martensitic transformation and mechanical twinning can occur simultaneously when the stacking fault energy is in the range of 12–18 mJ/m^2; dislocation glide becomes the dominant deformation mechanism when the stacking fault energy is above 35 mJ/m^2 [37–41]. Therefore, for the best SME, it is necessary to ensure that the stacking fault energy is below 12 mJ/m^2 for Fe-Mn-Si based SMAs.

Secondly, a high density of stacking faults should be distributed uniformly inside the austenitic matrix [20,24,42–47]. The atomic arrangement of a stacking fault in the austenite is equivalent to a thin ε martensite with two atomic layers. As such, the stacking faults can act as embryos for the growth of ε martensite [48,49]. Therefore, the stress-induced ε martensite preferentially nucleates and grows at these preexisting stacking faults during deformation. In this case, the critical stress inducing martensitic transformation is significantly reduced and the ability to suppress the plastic slip during stress-induced ε martensitic transformation is also enhanced. Some ways of improving the SME of Fe-Mn-Si based SMAs involve introducing a high density of uniform stacking faults *via* training, TMTs and aus-forming [24–28,30,31,42–47]. It should, however, be noted that reported recovery strains of processed polycrystalline Fe-Mn-Si based SMAs are still below 6%, even after the use of these methods.

Thirdly, the austenitic matrix may be strengthened through solid solution hardening with interstitial atoms such as carbon and nitrogen, or by dispersion hardening with second-phase precipitates. However, the effectiveness of carbon and nitrogen on the SME seems to be limited [50–57]. It is well known that the starting temperature of the thermally-induced martensitic transformation (M_s) can be significantly reduced by adding a small amount of carbon or nitrogen into Fe-Mn-Si based SMAs [53,54,57]. The deformation temperature is, in most cases, at around room temperature [50–57]. Note that a good SME is generally obtained as the deformation temperature is close to the M_s [58]. As such, the strengthening effect of carbon and nitrogen on the austenitic matrix is blinded by the improper selection of deformation temperatures. As a result, the improvement of the SME is limited, and the SME may even deteriorate after the addition of carbon and nitrogen. Recently, it was found that the shape recovery ratio increased from 42% at a deformation temperature of 293 K to 81% at a deformation temperature of 77 K when the deformation strain was 3.7% in the processed Fe-17Mn-5.5Si-9Cr-5.5Ni-0.12C alloy subjected to solution treatment at 1373 K for 30 min [59]. This result clearly revealed that the addition of interstitial atoms can improve the SME of Fe-Mn-Si based SMAs radically, since the proper deformation temperature is selected. In addition to the addition of interstitial atoms, the precipitation of second-phase particles, such as NbC [25,31,60], VN [61,62], VC [63,64], TiC [65], and $Cr_{23}C_6$ [22], can also effectively strengthen the austenite and markedly improve the SME in FeMnSi based SMAs. Furthermore, it was reported that the precipitation of second-phase particles during the training or the TMTs is beneficial for further improving the recovery strain [66,67]. Unfortunately, polycrystalline Fe-Mn-Si based SMAs treated as above still cannot achieve the stated aim of a recovery strain more than 6%.

Fourthly, the formation of annealing twins should be suppressed in Fe-Mn-Si based SMAs [34,68,69]. Our previous research indicated that the interactions between annealing twins and stress-induced ε martensite not only distort the twin boundaries heavily, but also significantly inhibit the stress-induced ε martensitic transformation [34]. Consequently, processed polycrystalline Fe-Mn-Si based SMAs show low recovery strains without special treatments. The number of annealing twin boundaries can be significantly reduced by training, TMTs, and aus-forming [34]. However, even then, the recovery strains cannot exceed 6% in processed polycrystalline Fe-Mn-Si based SMAs.

As summarized above, training and TMTs do not just introduce a uniformly high density of stacking faults, but also significantly reduce the amount of annealing twins. Furthermore, second-phase particles could be precipitated in the austenitic matrix after the training or the TMTs when a certain amount of carbon is added in Fe-Mn-Si based SMAs, which are beneficial for the SME. However, their recovery strains are still below 6%. In other words, it has not been possible hitherto to produce a large recovery strain of above 6% only based on the above four criteria. The reason for this may be associated with the austenitic grain-size. In general, it is easy to reach austenitic grain-sizes of about 500 μm, even millimeter-scale, by casting. However, the austenitic grain-sizes are generally below about 200 μm in processed Fe-Mn-Si based SMAs. In this case, a large recovery strain of above 6% can be obtained in cast Fe-Mn-Si based SMAs, whereas, the processed Fe-Mn-Si based SMAs cannot achieve such a high level of recovery strains. It may, therefore, be hypothesized that the maximum recovery strain is dependent on the austenitic grain-size for polycrystalline Fe-Mn-Si based SMAs. Consequently, we propose that austenitic grain growth is a key step towards achieving giant recovery strains in Fe-Mn-Si based SMAs, in addition to the above four criteria.