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Effect of compressive load on the martensitic transformation from austenite to 5M martensite in a polycrystalline Ni–Mn-Ga alloy studied by *in-situ* neutron diffraction

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A B S T R A C T

In this study, the influences of uniaxial compressive load on martensitic transformation from austenite to 5M martensite were studied in a directionally solidified Ni–Mn-Ga polycrystalline alloy with coexisting $\langle 0\ 0\ 1\rangle_A$ and $\langle 1\ 1\ 0\rangle_A$ preferred orientations parallel to the solidification direction (SD). Based on the neutron diffraction, the direct evidence on the variant redistribution induced by the thermal-mechanical treatment was presented and the selection of preferential variants was found to be strongly dependent on the austenite orientation. For the austenite with a $\langle 0\ 0\ 1\rangle_A$ preferred orientation, the compressive loading direction (LD) along the SD can promote the formation of preferred variants with $\{0\ 2\ 0\}_{5M} \perp$SD (LD). On the other hand, for the austenite with a $\langle 1\ 1\ 0\rangle_A$ preferred orientation, the variants with $\{1\ 2\ 5\}_{5M}/\{\bar{1}\ 2\ 5\}_{5M} \perp$SD (LD) are more favorable after the thermal-mechanical treatment. Such variant selection is originated from the accommodation between the anisotropic lattice distortion in martensitic transformation and the external constraint. The present investigations may offer some fundamental information on variant selection subject to external stress field and the necessary guidelines for microstructure optimization of polycrystalline Ni–Mn-Ga alloys through external field training.

1. Introduction

Heusler type Ni–Mn-Ga ferromagnetic shape memory alloys, with the combination of the ferromagnetism with a diffusionless, reversible martensitic transformation, recently have attracted great attention due to their significant magnetic field controlled functional behaviors [1]. Because of the strong coupling between the magnetic and structural orders, these alloys have demonstrated giant magnetic field induced strains (*i.e.* magnetic shape memory effect) up to ~12% in single crystals [2]. Such effect takes place in the martensitic state and is attributed to the reorientation of martensitic variants under a magnetic field [3]. With the integration of large output and fast dynamic response under the external magnetic field, Ni–Mn-Ga alloys are conceived as the promising candidates for the actuation and sensing applications [4].

The parent phase of Ni–Mn-Ga alloys is a cubic $L2_1$ ordered austenite, which transforms to the lower symmetry martensitic phase on cooling. Three types of martensite are frequently observed [5–8], namely five-layered modulated (5M) martensite, seven-layered modulated (7M) martensite and non-modulated (NM) martensite. Which type of martensite is prevalent after the phase transformation is strongly dependent on the chemical composition. So far, the 5M martensite has been extensively studied [9–16], since it may offer a lower twinning stress required for variant reorientation in comparison with 7M and NM martensites [11–14]. It has been reported that the twinning stress of 5M martensite can be as low as ~0.1 MPa [14]. Clearly, the lower twinning stress is more attractive since it results in the better actuation performance, thus

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the higher potential for practical applications.

In general, the martensitic transformation is a kind of shear deformation dominant diffusionless solid-state phase transformation with symmetry break. The lower symmetry of the product phase may result in the formation of several oriented variants with self-accommodation characteristics to compensate the elastic strains associated with the transformation [17]. However, such self-accommodated microstructure with multi-variants is not favorable for the achievement of magnetic shape memory effect in Ni–Mn-Ga alloys. The co-existence of multi-variants would greatly enhance the resistance for the variant reorientation, resulting in the loss of output under the magnetic field. To improve the magnetic shape memory properties, the microstructure modification through external field training to reorganize the variant configuration is supposed to be an essential prerequisite [18]. It has been established that the so-called thermo-mechanical treatment [18,19], *i.e.* the application of external load during the martensitic transformation, is an effective method to perform microstructure modification. For example, James et al. reported the enhancement of the magnetic field induced strain from 0.5% to 4% after the specimen was submitted to a thermo-mechanical treatment [19].

In fact, the external stress applied during the martensitic transformation would promote the formation of some favorable variants but prevent other unfavorable ones. Such an effect should be attributed to the coupling between the anisotropic external constraint (*e.g.* unidirectional tension or compression) and the crystallographic anisotropy of lattice deformation accompanying the martensitic transformation. Therefore, the application direction of external constraint determines the formation of the final martensitic microstructure. Deep insights into the variant redistribution feature caused by thermo-mechanical treatment as well as its correlation to the applied direction of external load should not only enrich the martensitic transformation crystallography, but also offer some necessary guidelines for the design and optimization of the external training process.

In our previous work, we studied the influence of the compressive load applied during the martensitic transformation on a directionally solidified $Ni_{50}Mn_{30}Ga_{20}$ alloy with $<0\ 0\ 1>_A$ preferential orientation [20]. We found that the compressive load applied during the martensitic transformation along $<0\ 0\ 1>_A$ can result in a strong variant selection, with $<0\ 1\ 0>_{7M}$ preferential orientation of 7M martensite along the loading axis [20]. On the other hand, it still needs further exploration to figure out the orientation effect of the external loading direction applied during the martensitic transformation on the selection of preferential variants. In the present investigation, considering the fact that the 5M martensite with relatively lower twinning stress can provide better actuation performance under the magnetic field, we chose a typical $Ni_{50}Mn_{28.5}Ga_{21.5}$ alloy with 5M martensite as the study object and prepared it by directional solidification. We found that there exist two preferred orientations with $<0\ 0\ 1>_A$ and $<1\ 1\ 0>_A$ parallel to the solidification direction (SD). Such crystallographic feature in the initial austenite orientation may provide an ideal testing condition to analyze the orientation dependence of the external loading applied during the martensitic transformation (*i.e.* thermal-mechanical treatment) on the variant selection. Based on the neutron diffraction, the direct evidence on the variant redistribution induced by the thermal-mechanical treatment was presented and the selection of preferential variants in related to the austenite orientations was discussed.

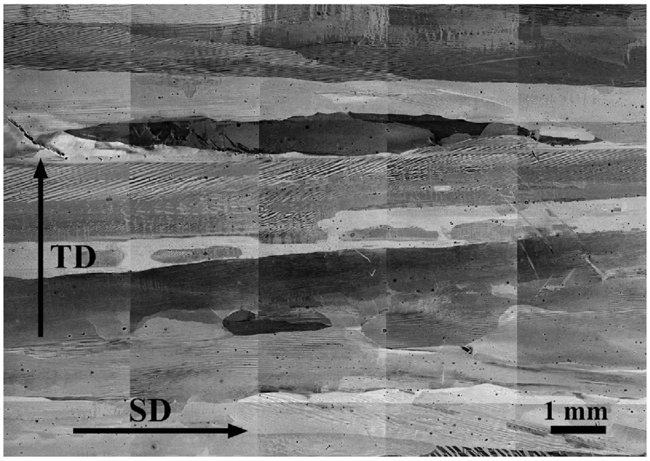

Fig. 1. Microstructure of the directionally solidified $Ni_{50}Mn_{28.5}Ga_{21.5}$ alloy at the room temperature (SD: solidification direction; TD: transverse direction).

2. Experimental methods

A polycrystalline Ni–Mn-Ga alloy with the nominal composition of $Ni_{50}Mn_{28.5}Ga_{21.5}$ (at.%) was prepared by directional solidification. The as-solidified alloy was homogenized at 1173 K for 24 h in a sealed vacuum quartz tube, followed by the quenching in water. Parts of homogenized alloy was ground into powder and then the powder was annealed at 873 K for 5 h to release the internal stress for the subsequent powder X-ray diffraction (XRD) measurements. The rectangular parallelepiped samples (10 mm × 6.5 mm × 6.5 mm) with their longitudinal direction parallel to the solidification direction were cut from the homogenized alloy by wire electrical discharge machining for the neutron diffraction measurements.

The room-temperature crystal structure was identified by powder XRD with Cu K_α radiation in a PANalytical X'Pert Pro MPD diffractometer. The martensitic transformation temperatures were measured by differential scanning calorimetry (DSC, TA Q100) in a temperature range from 183 K to 423 K, with a heating and cooling rate of 10 K/min, respectively. The *in-situ* thermo-mechanical treatment and neutron diffraction experiments were performed using the materials science diffractometer STRESS-SPEC operated by FRM II and HZG at the Heinz Maier-Leibnitz Zentrum (MLZ), Garching, Germany, with a monochromatic wavelength of 2.1 Å [21]. The uniaxial compressive load was applied by a rotatable

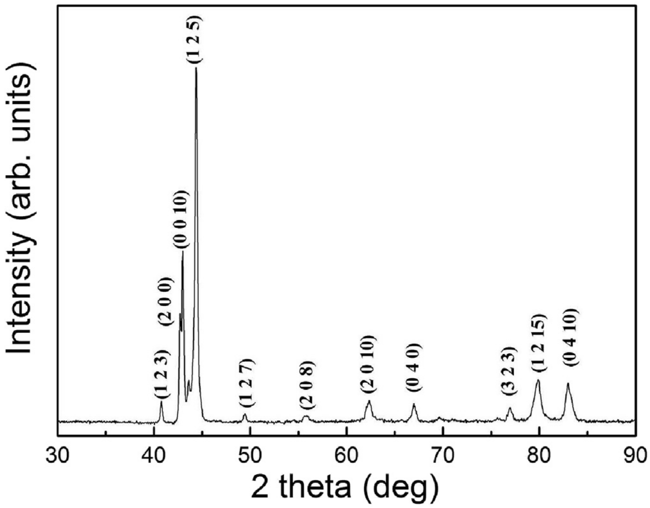

Fig. 2. Powder XRD pattern of the directionally solidified $Ni_{50}Mn_{28.5}Ga_{21.5}$ alloy at the room temperature.