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Compressive deformation of polycrystalline Ni-Mn-Ga alloys near chemical ordering transition temperature

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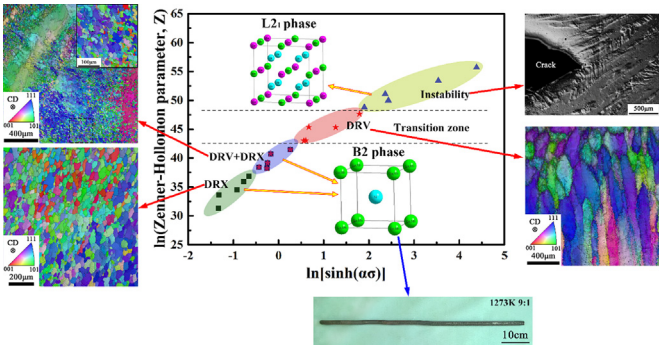
HIGHLIGHTS

- Plasticity in a $Ni_{51.3}Mn_{27.6}Ga_{21.1}$ alloy was demonstrated over $L2_1 \rightarrow$ B2 transition temperatures.
- DRV process occurred around the $L2_1 \rightarrow$ B2 transition temperature range.
- DRX and DRV/DRX mixed processes existed in B2 phase.
- Partially ordered crystal structure and DRX contributed to the good plasticity of B2 phase.

GRAPHICAL ABSTRACT



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ABSTRACT

Ferromagnetic shape memory Ni-Mn-Ga alloys exhibit an ordered $L2_1$ to partially ordered B2 transition at temperatures 873–1073 K. Based on the isothermal compressive results, it is demonstrated that $L2_1$ phase was brittle near ordering temperature, but the B2 phase was excellent above the chemical ordering temperature. In the compressive mode at temperatures of 473–1273 K and strain rates of 0.001–1 s^{-1}, the plastic deformation mechanism was found to be strongly dependent on the Zener-Hollomon parameter *Z*: the dynamic recrystallization was the dominant mechanism at the low *Z* region in the B2 state, then with increasing Z the dynamic recovery and ordering transition from B2 to $L2_1$ occurred, finally only dynamic recovery existed at the high *Z* region in the $L2_1$ state. Based on the processing map constructed from the isothermal compressive curves, hot extrusion of Ni-Mn-Ga alloy with ratio as high as 9:1 was achieved at 1273 K. This proved that polycrystalline Ni-Mn-Ga alloys are strikingly facile for large plastic deformation in the B2 state by dynamic recrystallization at temperatures around 1273 K and strain rate 0.03 s^{-1}, which affords a viable temperature and strain rate processing window for this intrinsically brittle alloy.

1. Introduction

Ni-Mn-Ga ferromagnetic shape alloys (FMSAs) exhibit large magnetic-field-induced strain (MFIS) and are attractive for applications in sensors, actuators and dampers operating at high frequencies [1]. MFIS as high as 7.1–11.2% in single crystalline Ni-Mn-Ga alloys [2–5]

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and 8.7% in polycrystalline foams [6] has been reported. Compared with single crystalline Ni-Mn-Ga alloys that are expensive and prone to compositional segregation during the crystal growth process, polycrystalline alloys are cost effective because they can be produced by conventional casting technique. However, constraints caused by the grain boundaries may reduce the mobility of the twin boundary and lead to a significantly reduced MFIS.

Several strategies exist in reducing the grain boundary constraints and thus enhancing the MFIS: (1) Fabrication of micron-sized materials such as microwires [7–9], ribbons [10] and thin films [11], in which the high surface to volume ratio and bamboo grains formed by annealing [7,8] were responsible for the reduced constraints and thus enhanced MFIS in polycrystalline Ni-Mn-Ga alloys [12]. (2) Introducing pores into bulk alloys in order to create micron-sized nods and struts surrounded by pores [13]. As martensitic twins covered the whole width of the nods/struts, the constraints caused by the grain boundary were significantly reduced. As a result, a maximum MFIS of 8.7% may be achieved in such polycrystalline foams [6]. (3) Creation of textures into polycrystalline Ni-Mn-Ga alloys by unidirectional solidification [14–16] and plastic deformation such as hot extrusion [17–19], rolling [20–22], forging [23,24] and torsion [25]. For example, ⟨100⟩ texture might be produced by unidirectional solidification [14–16], yielding a 1% MFIS in polycrystalline $Ni_{50}Mn_{29}Ga_{21}$ alloy [15]. Compared with the unidirectional solidification technique, plastic deformation is more convenient and efficient in forming a texture that favors MFIS [26]. In addition, the homogenous composition after hot deformation also benefits for the MFIS.

However, polycrystalline Ni-Mn-Ga alloys are extremely brittle at ambient temperature because of the strong elastic anisotropy, long-range ordered $L2_1$ crystal structure and mixed metallic-covalent bond, leading to their low deformability and thus difficulties for a large plastic deformation. These characters imply that dislocation slip process of $L2_1$ phase is difficult at room temperature, similar to many ordered intermetallic compounds such as Ni_3Al [27], TiAl [28] and Fe_3Al [29]. Conversely, Ni-Mn-Ga alloys exhibit a chemical ordering transition, i.e. from the a full ordering $L2_1$ to partial ordering B2 transition, at temperatures 873–1073 K upon heating [30,31]. The transition process of the atomic ordering in Ni-Mn-Ga alloys is different from that in binary intermetallic compounds such as Ni—Al, Ti—Al and Ni—Ti whose transformations into the ordered state occur at temperature just below the melting point. The B2 phase exhibits a partially ordered crystal structure and has much smaller Burgers vector than that of the $L2_1$ structure, which are responsible for the enhanced dislocation mobility and thus the recovery/recrystallization process during plastic deformation [32]. More importantly, the $L2_1$–B2 transition in Ni-Mn-Ga alloys is far below their melting points. For example, in stoichiometric Ni_2MnGa alloy, the $L2_1$ - B2 ordering temperature is 1071 K while the melting point is 1382 K [30], possibly leading to a wide working temperature window allowing for the plastic deformation. By contrast, conventional intermetallic compounds, i.e. Ni_3Al [27] and TiAl [28], become ordered state upon solidification, implying that the working temperature window may be small. As a result, the hot deformation mechanisms of Ni-Mn-Ga alloys should be different from the well-known binary ordered intermetallic alloys such as Ni—Al and Ti—Al.

The research on the hot deformation behavior of shape memory alloys (SMAs) mainly focuses on conventional SMAs such as Ni-Ti-based alloys because their shape memory effect and superplasticity can be enhanced by hot working process. The near equi-atomic NiTi alloy exhibits excellent workability, such as hot forging [33], hot rolling [34], with a hot working temperature ~1273 K at dynamic recrystallization domain [35,36]. The hot deformation mechanism is dislocation glide and climb based on the physically-based constitutive modeling [37,38].

The interesting $L2_1$–B2 transition and the broad B2 phase region provide Ni-Mn-Ga alloy a potential for a favorable plastic deformation capacity. Some plastic deformation routines, such as hot extrusion [17–19], rolling [20–22], forging [23,24] and torsion [25], have been demonstrated. However, the plastic deformation ability has not been fully revealed. For example, the work on the hot extrusion [17–19] has only been accomplished with a small extrusion ratio of 4:1. Furthermore, the underlying mechanisms governing the plastic deformation processes at temperature around $L2_1$–B2 transition temperatures are largely absent from the published literature. Here polycrystalline Ni-Mn-Ga alloys were isothermally compressed with a height reduction of 60%. The hot-working processing map of a polycrystalline $Ni_{51.3}Mn_{27.6}Ga_{21.1}$ alloy was built based on the stress-strain curves obtained in temperatures of 473–1273 K and strain rates of 0.001–1 s^{-1}. The Zener-Hollomon parameter Z was found suitable for describing the transition of the plastic deformation process, i.e. from dynamic recrystallization to dynamic recovery with increasing Z. Based on the processing map, the Ni-Mn-Ga alloy may be extruded at a ratio as high as 9:1. The extruded alloy, with ⟨111⟩ texture favorable for MFIS and small equiaxed grains [39], exhibited superplastic elongation 232.9% at temperature 1073 K and strain rate 0.001 s^{-1}. This confirmed the excellent deformation capacity of the extruded alloy at B2 state, making it possible for net-shaping complex components by conventional plastic deformation strategies, such as bending, tension and punching. Furthermore, the ⟨111⟩ texture formed by compression and extrusion also benefits for the enhanced MFIS, which is still an open question and warrants investigations in the near future.

2. Experimental details

High purity Ni (99.99%), Mn (99.98%) and Ga (99.99%) elements were used for the preparation of the Ni-Mn-Ga ingot by vacuum arc melting and casting. To ensure the homogeneity of the composition, the ingot was re-melted four times and finally suck-cast into a mould with diameter 10 mm. The composition of the cast ingot was $Ni_{51.3}Mn_{27.6}Ga_{21.1}$ (atomic percent) determined by a Zeiss-SUPRA 55 SAPPHIRE SEM equipped with an Oxford EDS. The ingot was then sealed in a quartz tube, evacuated and then back-filled with pure Ar (pressure 10 kPa). Finally, it was annealed at 1173 K for 10 h to further enhance the compositional homogeneity. The crystal structure of $Ni_{51.3}Mn_{27.6}Ga_{21.1}$ was determined by X-ray diffraction to be 5 M martensite at ambient temperature. The $L2_1$–B2 transition was determined using a Netzsch STA 449C differential scanning calorimetry (DSC) at heating rate of 10 K/min under a flowing Ar atmosphere.

Cylindrical samples of 4 mm in diameter and 6 mm in length were prepared by electron discharge machining from the annealed ingot. A NiCr–NiAl thermocouple was welded to the middle of the sample surface. The compressive tests were carried out in a Gleeble 1500D simulator equipped with a vacuum chamber. Testing temperatures of 473–1273 K, strain rates of 0.001–1 s^{-1} and a maximum engineering strain of 0.6 were adopted for the compressive tests. Upon finishing of the compression, the sample was quickly quenched into water to retain the deformation microstructure. The microstructures of the deformed samples, cut perpendicular to the compressive direction, were mechanically polished and examined using an Olympus PMG3 optical microscope under polarization mode. The sample for electron backscattered diffraction (EBSD) analysis was further polished with colloidal silica in a VibroMet2 (Buehler Ltd.) vibratory polisher to reduce the residual stress on the surface. The EBSD analysis of the sample was performed on a Zeiss Sigma™ HDVP field emission SEM with Gemini electron source and Digiview high-speed camera with the associated TSL OIM Analysis 7 software by EDAX™, with accelerating voltage 20 kV, working distance 17 mm and step-size 0.3–6 μm. In order to eliminate the effect of the complex martensite twin variants, the sample was heated in-situ within the SEM to 473 K to reach a full austenite state for subsequent EBSD data collection. A cast ingot with diameter 50 mm was canned with low carbon steel and then extruded into a rod at a temperature 1273 K and extrusion ratio 9:1.