Effect of Magnetic Field on Deformation Properties in Ferromagnetic Ni₂MnGa Shape Memory Alloy

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Ferromagnetic shape memory alloy Ni-25 at%Mn-23 at%Ga prepared by melting in an electric furnace was investigated to study compression properties at various temperatures with and without magnetic field. The magnetic field is applied parallel to the compression axis at a temperature below martensite finish temperature and at a temperature above austenite finish temperature. Triggering stress necessary for rearrangement of martensite variants is reduced by 0.76~1.68 MPa (*i.e.*, 2.5~5.5%) when a magnetic field of 0.4~0.6 T is applied to martensite state. The stress reduction is larger at a higher magnetic field. On the other hand, triggering stress for stress-induced martensite transformation is essentially unchanged under the same magnetic field applied to austenite state. This is due in part to very small changes in Gibbs free energy under magnetic field because of large temperature differences between M_s and T_c , and because of essentially no difference in magnetization between martensite and austenite phases.

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1. Introduction

Ferromagnetic shape memory alloys (FSMAs) have received considerable attention in recent years for potential use in sensors and actuators as robust high-performance device elements. It is because such FSMAs are expected to show rapid actuation response caused by magnetic field-induced shape change, *i.e.*, magnetic field-induced shape memory effect (SME). The magnetic field-induced shape change has advantage over conventional thermally induced SME with respect to heat dissipation, which is a drawback for response speed.

Two mechanisms¹⁻⁷⁾ govern magnetic field-induced shape change. One involves the rearrangement of martensite variants through the movement of their variant interface caused by the application of magnetic field. The variant rearrangement happens when a magnetic field is applied at a temperature below martensite finish temperature (M_f). At such a temperature, each martensite variant has a magnetic moment oriented to the easy magnetization axis, *i.e.*, the *c*-axis in the case of Ni₂MnGa.⁸⁾ When a magnetic field is applied, the magnetic moment tends to turn from the easy direction to the direction of magnetic field, causing variant interface to move if change in magnetic energy is bigger than that for interface motion.⁸⁾ In this mechanism, magnetic anisotropy energy is an important factor along with elastic energy arising from the transformation strain when martensite is formed thermally.^{2-5,7)} The strain associated with this mechanism was observed to be 0.2–6%.^{2,4,7)} It should be noted that, in the mechanism, reversible shape change occurs only when the magnetic field is applied and removed under a constant stress.⁹⁾ No shape recovery occurs without stress. It is because rearranged martensite variants by a magnetic field are stable at that temperature. Remembered is the shape of

martensite phase, which is not the case for conventional shape memory alloys, where the shape of austenite phase is remembered.

Another mechanism involved field-induced martensite transformation, which can take place when a magnetic field is applied at a temperature above austenite start temperature (A_s) preferably above austenite finish temperature (A_f). When a magnetic field is applied, Gibbs free energy is reduced by magnetic energy causing field-induced martensite to stabilize. In this case, the stability of martensite can be increased under an applied magnetic field if there are large differences in magnetization between martensite and austenite phases.^{1,7)} Total Gibbs free energy changes for austenite (*i.e.* parent) phase to transform to martensite phase under an applied stress (σ_a) and under a magnetic field (H_a) can be expressed⁷⁾ as

$$\begin{aligned} \Delta G(T, \sigma_a, H_a)_{\text{total}}^{\text{P} \rightarrow \text{M}} = & \Delta G(T)_{\text{chemical}}^{\text{P} \rightarrow \text{M}} \\ & + \Delta G(T)_{\text{el}}^{\text{P} \rightarrow \text{M}} + \Delta E_{\text{friction}}^{\text{P} \rightarrow \text{M}} \\ & - \int \sigma_a d\varepsilon + \Delta G(H_a)_{\text{mag}} \end{aligned} \quad (1)$$

where $\Delta G(T)_{\text{chemical}}^{\text{P} \rightarrow \text{M}}$ is chemical energy change upon the austenite-to-martensite transformation, $\Delta G_{\text{el}}^{\text{P} \rightarrow \text{M}}$ elastic strain energy generated in the transformation, $\Delta E_{\text{fr}}^{\text{P} \rightarrow \text{M}}$ frictional energy spent on the movement of the interface between the two phases and $\Delta G(H)_{\text{mag}}$ magnetic energy given to the specimen. Here, the magnetic energy can be expressed as

$$\Delta G(H)_{\text{mag}} = - \int H dI \quad (2)$$

$$dI = I_{\text{S(M)}} - I_{\text{S(P)}}. \quad (3)$$

where dI is difference of saturation magnetization between martensite and austenite. As seen in eqs. (2), (3), in an

extreme case the difference of magnetization can be maximized when ferromagnetic martensite is formed directly from paramagnetic austenite,^{1,7,10)} where $I_{S(P)}$ is zero. In the case of Ni-19.5 at%Mn-27 at%Ga, such a condition is nearly satisfied and M_s increases linearly with applied magnetic field⁷⁾ and it has been observed that M_s increases by 6 K under a magnetic field of 800 kA/m.⁷⁾ In this mechanism, the shape to be remembered is that of austenite phase, as is the case for conventional shape memory alloys.

There are several FSMA's available at the present moment, and they are Heusler-type Ni₂MnGa-based,^{1–16)} Fe-Ni-Co-Ti,¹⁷⁾ Fe~30 at%Pd^{18–21)} and Fe-25 at%Pt¹⁹⁾ and Fe-Pt-Pd.²²⁾ Among FSMA's, Ni₂Mn-Ga-based alloys are particularly interesting in part because such alloys have extensively been studied recently to try to understand features related to SME and its related phase transformation behavior with and without magnetic field. Notable features observed, in addition to the above-mentioned features, are as follows. (1) The shape change in the [221] direction for a Ni-19.5 at%Mn-27 at%Ga single crystal is extremely small when martensite transformation occurs thermally without magnetic field, while it is about 1.3% under a magnetic field of 240 kA/m applied along the [672] direction.³⁾ This field-assisted thermally-induced shape change is reversible and associated with the formation of favorably oriented martensite variants under magnetic field.³⁾ (2) Shape change also occurs at a constant compression stress when magnetic field is applied. In this case, there is a threshold stress, above which stress-assisted strain increases linearly with compression stress at a magnetic field of 800 kA/m.⁷⁾ The threshold stress of polycrystalline Ni-19.5 at%Mn-27 at%Ga (A_f : 336 K) at 346 K is 126 MPa when a magnetic field of 800 kA/m is applied.⁷⁾ This stress-assisted field-induced shape change is not reversible upon removal of magnetic field and this behavior remains unexplained.

In addition to the above effects of magnetic field, it is necessary to understand SME and its related mechanical properties for FSMA's including Ni₂MnGa if they are to be used in applications. Since FSMA's are new materials, investigations are mostly concerned about demonstration of how much shape change occurs on what alloys. Hence, only a few reports are available on mechanical properties and behavior of FSMA's even without magnetic field.^{11,16,23)} We have recently carried out compression tests of Fe-bearing Ni₂MnGa alloys with a magnetic field at temperatures below M_s and the effect of magnetic field on triggering stress for variant rearrangement have been reported.¹⁴⁾ In the present

study, compression tests are also carried out for a ternary Ni-25 at%Mn-23 at%Ga alloy at temperatures above A_f and below M_f , with and without a magnetic field. The effect of magnetic field on triggering stress for field-induced variant rearrangement and field-induced martensite transformation are reported.

2. Experimental Procedure

Pure metals, Ni (purity 99.9%), Mn (99.9%) and Ga (99.99%), were put in an alumina crucible (9 mm in diameter and 600 mm long) to make a Ni-25Mn-23Ga alloy ingot (by nominal atomic percentages). After alloying in a high-purity argon atmosphere using an electric furnace for heating to 1623 K and holding at the temperature for 3 hours, the ingot was cooled down and then crashed into small pieces to try to make compositionally uniform ingots. Crashed alloy pieces were then put in an alumina crucible for re-melting in a similar way. This process was repeated a few times and an alloy ingot in a cylinder shape of 9 mm diameter and 60 mm length was fabricated. Thus fabricated ingot was sectioned to five columns 9 mm high and they are named specimens D, E, F, G and H from the top to the bottom (In a previous report,²³⁾ specimens made in a similar manner were named specimens A, B, and C). Primarily employed in the present study was specimen D, which was wire-electro-discharge machined into four rectangular-shaped compression specimens with dimensions of 2.7 mm × 2.7 mm × 9 mm. The compression specimens were all vacuum encapsulated in quartz tubes, annealed at 1173 K for 3.6 ks and then quenched into water without breaking the quartz tubes.

The top and bottom portions of the ingot, 60 mm apart, were chemically analyzed by an inductively coupled plasma atomic emission spectroscope and it was found that these portions were almost identical compositionally. The measured chemical compositions of the top and bottom portions (D and H) are listed in Table 1, along with martensite and austenite transformation temperatures determined by compression testing, differential scanning calorimetry (DSC) and magnetization measurements. For comparison, Table 1 also shows alloy compositions for specimens A and C of a previous study.²⁴⁾ The DSC measurements were carried out at cooling and heating rates of 5 K/min using a Seiko-made calorimeter (model DSC6100). Magnetization measurements were carried out as a function of temperature in a temperature range between 298 K and 383 K in a constant magnetic field of 166 kA/m applied by a magnetic balance (SHIMADZU,

Table 1 Alloy compositions (at%) of the Ni-Mn-Ga system investigated and phase transformation temperatures (K) obtained by compression testing. Curie temperature was determined by DSC and magnetization experiment.

Alloy	Ni	Mn	Ga	M_s	M_f	A_s	A_f	T_c	Remarks
Ni-25Mn-23Ga: A	51.9	25.0	23.1	298	287	300	311	365	Ref. 22)
Ni-25Mn-23Ga: C	52.1	24.6	23.3	296	293	312	326	365	Ref. 22)
Ni-25Mn-23Ga: H	52.4	24.5	23.1	293	288	300	306		present work
				294	293	308	311		mechanical test
Ni-25Mn-23Ga: D	52.6	24.2	23.2	319	288	296	326	365	DSC
				320		316	326	366	magnetization

MB-2). Although the data is not shown in the present study, optical microscopy and X-ray diffraction showed that the high temperature austenite phase of the alloy is single phase having an ordered Heusler-type structure with a lattice parameter of 0.5808 nm.

Compression tests were carried out at a strain rate of $9.3 \times 10^{-5} \text{ s}^{-1}$ using an Instron-type testing machine (SHIMADZU, IS-5000) equipped with a temperature-controlled bath, as shown in Fig. 1. During testing, test temperatures were controlled within an accuracy of 1 K by monitoring and controlling the temperature of the bath, which contained ethanol for a temperature range between 238 K and 268 K and water between 278 K and 328 K. Compression specimens were all heated to 373 K (above A_f) for shape recovery after unloading. Optical microscope observations were carried out using an optical microscope (Nikon, OPTIPHOT) after mechanical polishing with emery papers and Al₂O₃ powders and electro-polishing in a solution of 50% perchloric acid and

50% ethanol to remove damages induced during mechanical polishing.

The effect of magnetic field on deformation properties was investigated by placing compression specimens between paired NdFeB permanent magnets (8 mm or 14 mm in diameter) in a parallel configuration of magnetic field with respect to the compression direction. Figure 1 is a schematic illustration of compression test apparatus showing the parallel configuration, where paired magnets were mounted on the top and bottom compression grips. Two-dimensional distributions of magnetic fields created by paired NdFeB magnets were measured and the results were drawn in Fig. 2. In this figure, the position of specimens is indicated by dotted lines. As seen, magnetic fields applied are center-symmetric, ranging (a) from 0.2 to 0.5 T for paired magnets of 8 mm in diameter and (b) from 0.4 to 0.6 T for paired magnets of 14 mm in diameter. When compression specimens are set between the paired magnets, as seen in Fig. 2, large portion of the specimens are under a field of 0.2–0.3 T for ϕ 8-mm magnets and under that of 0.4–0.5 T for ϕ 14-mm magnets. After compression, specimens were all unloaded and demagnetized followed by heating up to 423 K for shape recovery.

3. Result and Discussion

3.1 Transformation temperatures

DSC experiments were performed for specimen D to determine martensite and austenite transformation temperatures and DSC curves obtained are shown in Fig. 3. As seen in this figure, M_s and A_f cannot be readily identified because both exothermal and endothermal peaks overlap with reaction peaks for para-to-ferro and ferro-to-para magnetic transitions, respectively. Therefore, M_s and A_f are regarded, respectively, as M^* and A^* , which are the top and bottom of the respective reaction peak associated with transformations.²⁴⁾ The measured transformation temperatures of M_f , M_s , A_s and A_f are 288 K, 319 K, 296 K and 326 K,

Fig. 1 Schematic illustration of compression test apparatus.

Fig. 2 Distribution of magnetic fields parallel to compression direction, created by NdFeB magnets, (a) 8 mm in diameter and (b) 14 mm in diameter. Dotted lines indicate the position of specimen.

Fig. 3 DSC profile for specimen D of Ni-25Mn-23Ga.

respectively, for Ni-25Mn-23Ga, as shown in Table 1.

Transformation temperatures can also be measured from relationships between triggering stress (where stress-induced martensite begins to form) and test temperature using Clausius-Clapayron equation.⁷⁾ Such triggering stresses were measured from compressive stress-strain curves, obtained from a compressive specimen, at various temperatures. The specimen was deformed at various temperatures up to either the completion of rearrangement of martensite variants or that of stress-induced martensite transformation. At each test temperature, the specimen was heated above A_f point after unloading. Compressive stress-strain curves thus obtained from Ni-25Mn-23Ga specimen D1 are depicted in Fig. 4, where N indicates the order of testing and arrows represent shape recovery by heating after unloading. As seen, shape memory effect occurs between 238 K and 318 K and super-elasticity at 328 K. As arrows indicate, 100% of shape

Fig. 4 Compressive stress-strain curves for Ni-25Mn-23Ga specimen D1 deformed at various temperatures. At each test temperature, the specimen was heated to 373 K after unloading, where 100% shape recovery occurred. N indicates the order of testing.

Fig. 5 Triggering stress as a function of test temperature for Ni-25Mn-23Ga specimen D1. σ_{M_s} is a triggering stress necessary for austenite to start martensite transformation, σ_{M_f} stress for austenite to finish the transformation, σ_{Re} is a triggering stress necessary for martensite variants to start rearrangement and σ_{Ref} stress to finish rearrangement.

recovery occurs at all test conditions employed.

From Fig. 4, triggering stresses were measured where either stress-induced martensite occurs at $T > M_s$ or rearrangement of martensite variants occurs at $T < M_s$. Thus measured triggering stresses were plotted in Fig. 5 as a function of test temperature. According to Clausius-Clapayron relationship, the triggering stress for stress-induced martensite transformation holds a linear relationship with test temperature. The intercept of such linear lines with temperature axis gives martensite and austenite transformation temperatures, M_f , M_s , A_s and A_f , which are 293 K, 294 K, 308 K and 311 K, respectively, as shown in Table 1. The result is slightly different from that obtained by DSC in Fig. 3. This difference may be caused by experimental errors caused from insufficient data points of triggering stresses.

The magnetism of the parent and martensitic phase was investigated through magnetization measurements as a function of temperature for a piece of Ni-25Mn-23Ga specimen D, which was placed under a constant magnetic field of 166 kA/m. The result obtained is shown in Fig. 6. As seen from Fig. 6, the magnetization decreases slightly with

Fig. 6 Magnetization as a function of temperature for Ni-25Mn-23Ga specimen D.

increasing temperature, falling rapidly with further increasing temperature and becomes zero at 366 K, showing a typical ferromagnetic to paramagnetic transition. This temperature is regarded as Curie temperature (T_c) of this alloy. From the heating and cooling magnetization curves, martensite and austenite transformation temperatures were also determined using interception method of two tangents and the result is listed in Table 1. As seen in Fig. 6 and Table 1, the austenite and martensite phases of this alloy are ferromagnetic and M_s is about 40 K lower than T_c . It should be noted that the martensite and magnetic transformation temperatures obtained in magnetization measurements are in good agreement with those obtained in mechanical testing and DSC measurements.

Transformation temperatures were also measured for specimen H using compressive stress strain curves, similar to specimen D, as described above. The result is listed in Table 1. As seen in Table 1, the measured transformation temperatures are very close to those obtained for specimen D. This indicates that the alloy ingot fabricated in this experiment is compositionally very uniform. The alloying method employed in the present study is believed to be very good for making Ni₂MnGa alloys with desired transformation proper-

ties. That is not the case when arc melting and Bridgman methods are used. In these cases, it is extremely difficult to make alloys with desired transformation properties. It should be mentioned that the martensite and magnetic transformation temperatures measured in magnetization measurements are in good agreement with those obtained in mechanical testing and DSC measurements.

3.2 Surface observations

Optical microscopy was carried out to observe the microstructure of martensite in Ni-25Mn-23Ga specimen E, which is considered to represent the transformation properties of specimen D due to its compositional uniformity, as described above. Optical micrographs were all taken at room temperature after various processes, except for one case where micrographs were taken at 393 K. Observed micrographs are shown in Fig. 7, where the exception is Fig. 7(c). Surface observations were carried out in the following process. The specimen was electropolished and observed first at room temperature, showing mostly austenite phase (a); then cooled down to 253 K (below M_f) following warming up to room temperature, showing thermal martensite (b); then heated to 393 K (above A_f) where a micrograph was taken, showing

Fig. 7 Optical micrographs of Ni-25Mn-23Ga (Specimen E) taken at room temperature, a few degrees below M_s (293 K), after various conditions, except for (c) which was taken at 393 K. Arrow indicates the direction of deformation in compression.

austenite phase (c); cooled to room temperature, showing mostly austenite phase with a small amount of thermally formed martensite (d); deformed at room temperature, showing stress-induced martensite (e); heated to 393 K, showing mostly austenite phase (f); cooled to 253 K, showing thermal martensite (g); deformed at room temperature, showing rearrangement of martensite variants (h); and then heated to 393 K, eliminating rearranged martensite variants and showing almost austenite phase.

Optical microscopy can be summarized as follows. As seen in Fig. 7, martensite variants are induced thermally by cooling to 253 K, and they disappear by heating to 393 K. A circle in Fig. 7(d) shows martensite appeared in a small amount at room temperature. This is because this temperature is slightly below M_s of this alloy, which indicates the M_s point determined by mechanical testing is reasonably good, although the M_s point measured by DSC and magnetization measurements seem more accurate. Stress-induced martensite variants are seen in a circle in Fig. 7(e), and such variants are seen more in abundant when rearrangement of martensite variants occurred by deformation of thermal martensite in compression, as seen when compared Fig. 7(g) with Fig. 7(h).

3.3 Magnetic field effect

The effect of magnetic field on triggering stress was investigated both for rearrangement of thermally induced martensite variants and for stress-induced martensite. Triggering stresses for variant rearrangement were determined for Ni-25Mn-23Ga specimen D3 by deforming it at 288 K (below M_f) in compression with and without magnetic field parallel to the compression direction. Figure 8 shows some compressive stress-strain curves obtained without magnetic field and with the majority of a 0.2~0.3 T magnetic field created by ϕ 8-mm NdFeB permanent magnets and that of a

0.4~0.5 T magnetic field created by ϕ 14-mm NdFeB permanent magnets. Compression tests were alternatively performed with and without magnetic field for the single specimen. After testing with a magnetic field the specimen became magnetized and thus demagnetized by heating it to 423 K after unloading.

As seen in Fig. 8, stress-strain curves with magnetic fields are very similar to those without magnetic field, although both triggering stress and flow stress after apparent yielding seem slightly lower under magnetic fields than those without magnetic field. To see the change in triggering stress more readily, triggering stress was plotted as a function of the testing order, as shown in Fig. 9. As seen in Fig. 9, magnetic fields applied parallel to the compression direction lower triggering stress for martensite variant rearrangement. Reductions in triggering stress are approximately 0.76 MPa under a 0.2~0.3 T magnetic field and 1.68 MPa under a 0.4~0.5 T magnetic field. Similar magnetic field effect on triggering stress was also found in Ni₂MnGa containing Fe.¹⁴⁾ The present study evidences that the decrease in rearrangement stress is larger for stronger magnetic field. The reduction in triggering and flow stresses is considered to be associated with the alteration of magnetization direction under a magnetic field. To confirm this magnetic field effect on

Fig. 8 Compressive stress-strain curves with and without magnetic field for Ni-25Mn-23Ga specimen D3 deformed at 278 K. After unloading the specimen was heated to 423 K, where 100% shape recovery occurred. N indicates the order of testing.

Fig. 9 Triggering stress for variant rearrangement as a function of number of cycles for Ni-25Mn-23Ga specimen D3.

Fig. 10 Triggering stress for variant rearrangement as a function of number of cycles for Ni-25Mn-23Ga specimen A2.

Fig. 11 Compressive stress-strain curves with and without magnetic field for Ni-25Mn-23Ga specimen D4 deformed at 318 K. After unloading the specimen was heated to 423 K, where 100% shape recovery occurred. N indicates the order of testing.

triggering stress, also tested in a similar way to that employed above was specimen A2, which is shown to have very similar transformation temperatures to specimen D. Results obtained are plotted in Fig. 10. As seen, triggering stress for variant rearrangement is quite evident, similar to that observed from specimen D3.

Similar compression tests were also carried out at 318 K (above A_s) to investigate the effect of magnetic field on triggering stress necessary for stress-induced martensite transformation. Figure 11 shows compressive stress-strain curves obtained from Ni-25Mn-23Ga specimen D4, which was deformed with and without a 0.4~0.5 T magnetic field created by a pair of ϕ 14-mm NdFeB permanent magnets. As seen, superelasticity occurs in all curves regardless magnetic field. Residual strain after unloading was also recovered by heating. As seen, stress-strain curves are almost identical in this case, which can be compared with Fig. 8. Triggering stresses were measured from Fig. 11 and plotted in Fig. 12 as a function of the testing order. As seen in Fig. 12, there is essentially no effect of magnetic field on triggering stress. A large stress drop is seen at $N = 5$ when field is applied. This is not due to magnetic field effect but caused by a crack that occurred during testing. It should be noted that Ni₂MnGa cannot repeatedly be deformed many times, because austenite phase is somehow brittle.²³⁾ No magnetic effect on triggering stress for stress-induced transformation in the present study can be explained as follows. According to our transformation temperature measurements, as shown in Table 1, the difference in transformation temperature between M_s and T_c is large. In addition, magnetization measurements (Fig. 6) showed that there is only slight difference in magnetization between parent and austenite. Hence, Gibbs free energy change due to magnetic field is essentially negligible, leading no magnetic field effect on triggering stress. Appreciably large effect will be expected if Ni₂MnGa-based alloys with T_c nearly equal to M_s are tested.^{3,7)}

Fig. 12 Triggering stress for stress induced martensite transformation as a function of number of cycles for Ni-25Mn-23Ga specimen D4.

4. Summary

Ferromagnetic shape memory alloy Ni-25 at%Mn-23 at%Ga prepared by melting in an electric furnace was investigated to study compression properties at various temperatures with and without magnetic field. The magnetic field is applied parallel to the compression axis at a temperature below martensite finish temperature and at a temperature above austenite finish temperature. It is found that triggering stress necessary for rearrangement of martensite variants is reduced by 0.76~1.68 MPa (*i.e.*, 2.5~5.5%) when magnetic fields of 0.4~0.6 T is applied to martensite state. The stress reduction is larger at a higher magnetic field. On the other hand, triggering stress for stress-induced martensite transformation is essentially unchanged under the same magnetic field applied to austenite state. This is due in part to very small changes in Gibbs free energy under magnetic field because of large temperature differences between M_s and T_c , and because of essentially no difference in magnetization between martensite and austenite phases.

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