RESEARCH ARTICLE | APRIL 10 2018

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AIP Conf. Proc. 1942, 130046 (2018)
https://doi.org/10.1063/1.5029116

Effect of Different Annealing Condition on the Structural and Magnetic Properties of Mn_2NiGa Heusler Alloys

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Abstract. We studied the effect of different annealing conditions on structural and magnetic properties of Mn_2NiGa Heusler alloys. Reitveld refinement of neutron diffraction pattern at RT confirms the tetragonal structure with cubic phase for I-W quenched alloy whereas Le Bail fitting trials performed on neutron diffraction pattern collected for other three alloys confirm 7M monoclinic structure with cubic phase. It is found that starting and finish temperatures associated with martensite and austenite phase transformation depends strongly on the cooling rate corresponding to different cooling techniques. Slow furnace cooled sample possesses the highest martensite start temperature above room temperature ~ 326K which decreases to ~ 198K for ice –water quenched sample. Variation in the drop in the magnetization around M_S obtained upon warming from martensite to austenite phase under ZFC cycle suggests that change in the cooling condition strongly affects the magnetization in the low temperature martensite phase. Present results suggest that by varying the cooling rate, martensite transformation as well as the martensite structure can be tuned.

INTRODUCTION

In recent years, ferromagnetic shape memory alloys have gain considerable attention due to versatile interesting properties exhibited by them such as martensitic transformation[1], larger magnetocaloric effect[2], magnetic field induced strain[3], exchange bias[4], magnetic superelasticity[5], etc. One of the extensively studied ferromagnetic shape memory alloy Mn_2NiGa, which exhibits 4% shape memory effect[6], is potential candidate for technological applications because of high Curie temperature (~588K) and martensite transformation temperature close to room temperature ~270K[7]. It has been established that stress and structural relaxation during the post-annealing can significantly affect the martensitic transition[2]. Generally, thermal induced residual stress arises during the cooling stage. By varying the cooling rate, kinetics of the structural relaxation can be changed. Till date no report exists on the investigation on the effect of the cooling rate in modifying magnetic properties of Mn_2NiGa alloy. In this context we have studied the effect of different annealing conditions having different cooling rate on structural and magnetic properties of Mn_2NiGa Heusler alloys.

EXPERIMENTAL

Four polycrystalline samples of Mn_2NiGa were prepared in arc furnace by melting the stoichiometric quantities of the constituent metals (99.99% purity) under argon gas atmosphere. The as prepared ingots were annealed at 1073K in an evacuated quartz tube for 48 hours and subsequently subjected to different cooling conditions as follows: (1) furnace cooling (FC) (2) controlled furnace cooling with rate of 5°C/min (SC) (3) quenched in ice-water mixture and (I-W) (4) quenched in liquid N_2 (LN_2). Magnetization (M) measurements were performed on a vibrating sample magnetometer (Quantum Design, PPMS-VSM) in the temperature range of 5–390 K and magnetic field H=0.01. Room temperature powder neutron diffraction patterns were collected using using neutrons of wavelength 1.62 A ° at the high resolution neutron diffractometer Echidna at Bragg Institute, ANSTO, Sydney.

DAE Solid State Physics Symposium 2017
AIP Conf. Proc. 1942, 130046-1–130046-4; https://doi.org/10.1063/1.5029116
Published by AIP Publishing. 978-0-7354-1634-5/$30.00

RESULTS AND DISCUSSION

Figure 1 presents the temperature dependence of magnetization measured under zero-field-cooled (ZFC), field-cooled-cooling (FCC) and field-cooled-warming (FCW) protocols at 100 Oe applied magnetic field. In ZFC mode, samples were initially cooled down to 5K in zero magnetic field and after applying 100 Oe field magnetization was measured on warming the sample up to 390K followed by cooling back to 5 K in the presence of the field (FCC) and again during the warming (FCW). With lowering of temperature, initially magnetization increases and falls rapidly around 198, 234, 296 and 326K for I-W, LN_2, FC and SC samples, respectively. The large drop in the magnetization indicates the formation of the new structural phase which can also be associated to the large magnetocrystalline anisotropy.

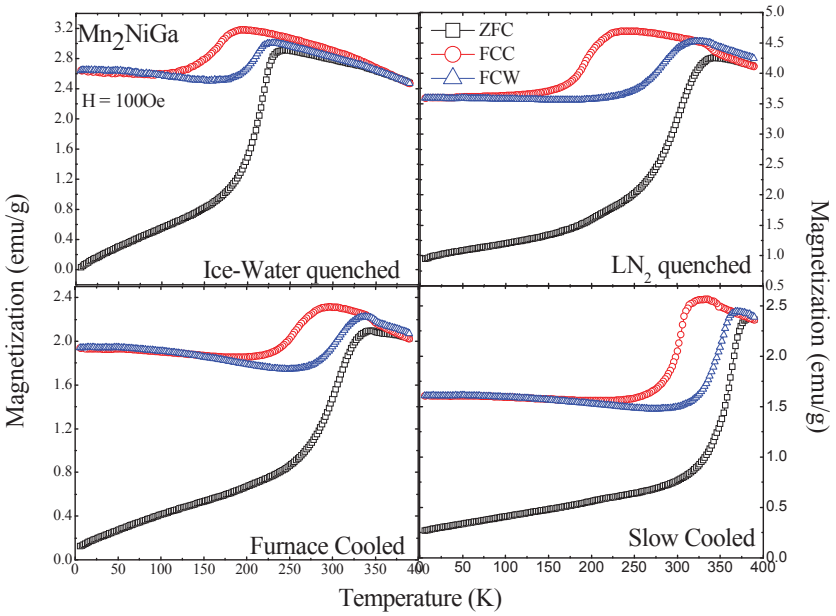

Figure 1: Temperature dependence of magnetization measured under ZFC, FCC and FCW protocols at H = 100Oe for Mn_2NiGa alloys with cooling condition as: (a) ice-water quenched (b) liquid nitrogen quenched (c) furnace cooled and (d) Slow/controlled furnace cooled.

It is clearly seen that magnetization of all Mn_2NiGa alloys exhibit large hysteresis between FCC and FCW cycles. The sharp decrease in the magnetization and thermal hysteresis indicates the existence of a first order transformation from high temperature austenite phase to martensite phase. Below martensite transition, splitting between ZFC and FC curves is observed which can be attributed to the frustrated spin arrangements arising from the ferromagnetic coupling between Mn atoms from the same site and antiferromagnetic coupling of Mn at two different crystallographic sites. The martensite start (M_s), martensite finish (M_f), austenite start (A_s) and austenite finish (A_f) temperatures related to austenite to martensite and martensite to austenite transitions during cooling and warming are given in Table 1. High temperature magnetization measurements suggest that all the samples under study are ferromagnetic at room temperature as the T_C is above 600K for all the samples (values are given in table 1). It can be observed that starting and finish temperature associated with martensite and austenite phase transformation depends strongly on the cooling rate corresponding to different cooling techniques. Slow furnace cooled sample possesses the highest martensite start temperature above room temperature ~ 326K. The width of the thermal hysteresis was determined by A_f-M_s as given in Table 1. Highest thermal hysteresis of ~ 90K is determined for LN_2 sample which is indicative of the high energy dissipation during the transformation arising from the frictional stress associated with the movement of austenite-martensite interface. Another striking feature in the M (T) plot is the large magnetization difference (i.e. ΔM) between the martensite and austenite phase. Variation in the drop in the magnetization around M_S in (%) obtained upon warming from austenite to martensite phase under ZFC cycle suggests that change in the cooling condition strongly affects the magnetization in the low temperature martensite phase.

Table 1: Values of the martensite start (Ms), martensite finish (Mf), austenite start (As) and austenite finish (Af) temperatures, Curie temperature (TCA), transformation hysteresis and ΔM for Mn2NiGa prepared under different cooling conditions.

	M_S	M_F	A_S	A_F	T_C^A	Transformation Hysteresis (A_f-M_s)	ΔM (%)
	(K)						
SC	326	265	308	368	617	42	66
FC	296	224	269	339	631	43	57
N_2	234	153	221	324	635	90	87
I-W	198	109	171	230	614	32	56

Figure 2 shows the Rietveld refinement of the neutron diffraction pattern collected at 300K for I-W quenched Mn_2NiGa sample using tetragonal structure with I4/mmm space group. Parameters obtained from the refinement of the neutron diffraction pattern are given in Table 2. In Mn_2NiGa, half of the Mn occupies Ni site with atomic position 4d (0, 0.5, 0.25) and other half occupies 2a (0, 0, 0) site with Ga atom residing at 2b (0, 0, 0.5) site. Occupancies obtained from the refinement suggest the antisite disorder between Mn at Ni site and Ga atoms. The net moments obtained are -0.661, 2.135 and 0.87 (μ_B) for 4d, 2a and 2b atomic site, respectively. Magnetic moment at 2b site can be attributed to the replacement of Ga atoms by Mn atoms from Ni site. The opposite sign of the moment at 4d and 2a site suggest the antiferromagnetic interaction between Mn spins. These results are in good agreement with the results published by Singh et al.[8]

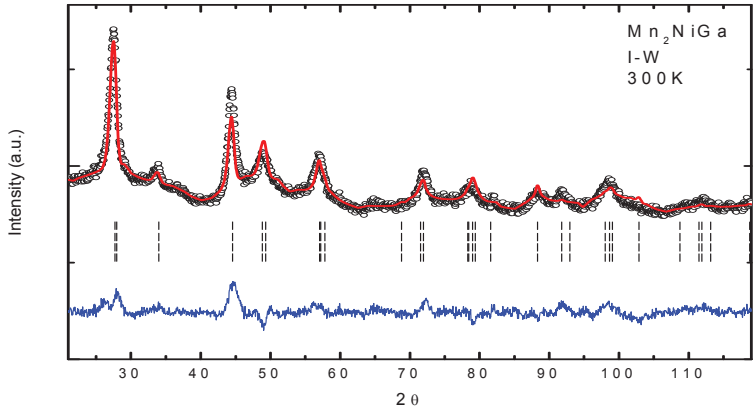

Figure 2: Room temperature powder neutron diffraction pattern of as prepared bulk I-W quenched Mn_2NiGa. The observed and calculated patterns are presented by black circle and red line, respectively. Blue line shows the difference between the observed and the calculated patterns whereas vertical tick presents nuclear and magnetic Bragg positions.

Table 2: Parameters obtained from the refinement of the neutron diffraction pattern for as prepared bulk I-W quenched Mn_2NiGa alloy.

Space group	***I4/mmm***		
Unit cell(Å)	**a = b = 3.9127 (4) c = 6.7909 (5)**		
Cell Volume (Å)^3	**103.96**		
Site	Atom	Moment (μ_B)	Occupancy
4d	Ni Mn Ga	-0.661	0.06250 0.04286 0.01964
2a	Mn	2.135	0.06250
2b	Ga Mn	0.87	0.05542 0.00708

Figure 3 (a) shows the room temperature neutron diffraction patterns obtained for LN_2 , FC and SC alloys, note that, here ND pattern of I-W quenched sample is also included for comparison purpose. It is evident from the figure that, the reflection corresponding to martensite phase splits into two peaks. Additional peaks could not be indexed using same model that was used to refine the I-W sample which suggests the presence of the chemically modulated super-period martensite structure. Figure 3 (b) shows Le Bail fitting trials performed on room temperature neutron diffraction pattern collected on LN_2 quenched Mn_2NiGa alloy. As it can be seen that the splitting observed for the

peak at ~ 27.4° cannot be indexed using combination of 3M or 5M tetragonal and cubic phase. Considering 7M monoclinic structure splitting of peak at 27.4°can be indexed but reflection at 35.7° cannot be indexed. A good fit is obtained only when we consider 7M monoclinic structure with cubic phase where all the peaks are indexed. Obtained lattice parameters from Le Bail refinement are a = 4.26 Å, b = 28.6 Å and c = 5.35 Å, β = 93.1° suggesting a sevenfold increase in the unit cell along b direction.

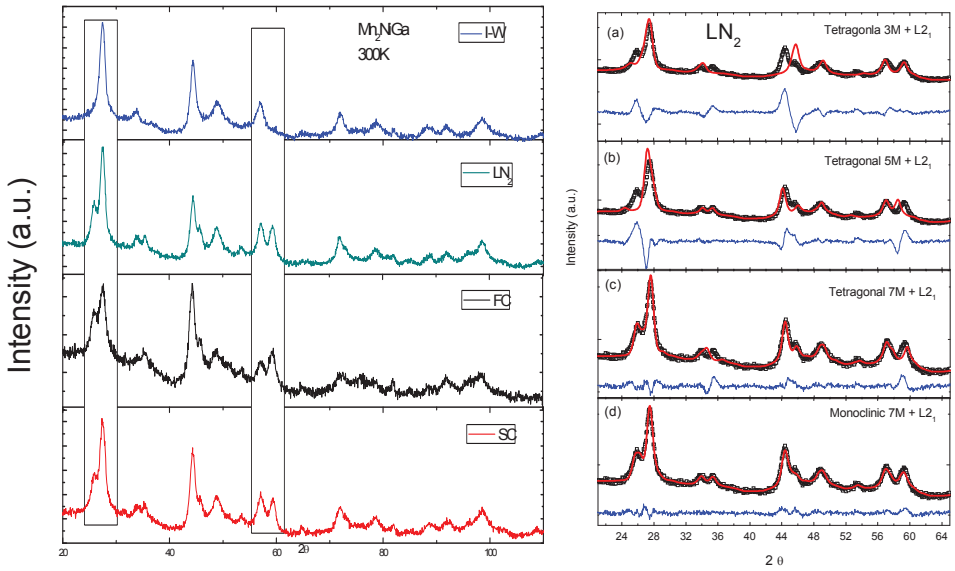

Figure 3: (Left) Comparison of the room temperature neutron diffraction patterns for I-W, LN_2, FC and SC samples. (Right) Trials of the Le Bail fittings performed on the LN_2 quenched Mn_2NiGa sample (a) Tetragonal 3M + $L2_1$ (b) Tetragonal 5M + $L2_1$ (C) Tetragonal 7M + $L2_1$ and (d) Monoclinic 7M + $L2_1$.

CONCLUSIONS

Present results suggest that by varying the cooling rate, martensite transformation as well as the martensite structure can be tuned.

ACKNOWLEDGMENTS

Authors are thankful to Prof. K.R. Priolkar for fruitful discussion. Authors thank the Department of Science and Technology, India (SR/NM/Z-07/2015) for the financial support to carry out neutron diffraction measurements and Jawaharlal Nehru Centre for Advanced Scientific Research (JNCASR) for managing the project.

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