

Study of the effect of magnetic field on the structure of Ni-Mn-Ga shape memory alloys

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Abstract. In certain ferromagnetic shape memory Ni-Mn-Ga alloys, the shape changes of the material can be controlled by magnetic field. These changes occur in the martensitic phase and they are result of the martensitic twin boundary motion together with the change of the martensite variants. In the present study, the connection between the magnetisation of the alloys and their martensitic structure was observed by using different methods of investigation. The structure of non-stoichiometric Ni-Mn-Ga alloy was investigated by the optical metallography connected to the optical differential scanning calorimeter (DSC), scanning electron microscopy (SEM) and the X-ray diffraction θ - 2θ -analysis. Also, the straining of the materials in dynamic magnetic field was studied. The results of these different investigation methods were in good agreement with each other. However, with the methods in which electron beam was applied to the specimen, it was found that the structure was affected by the research method. The images of martensitic structure obtained with the different methods were compared with each other.

1. INTRODUCTION

Some of the Ni-Mn-Ga alloys having magnetic shape memory (MSM) effect - deformation controlled by applied magnetic field - tend to exhibit a great technological potential as actuators and sensors in electromechanical engineering [1]. The mechanism of magnetic shape memory effect was presented in [1,2] and this mechanism included the magnetically induced motion of the martensite twin boundaries. The large magnetic induced strains caused by the martensite twin boundary motion, were observed in [3-5]. Effect of the applied magnetic field has been displayed with X-ray study by change of the martensite variants [6,7]. The connection between the possible martensite variants observed with optical microscopy and magnetization in Ni-Mn-Ga alloys was shown in [7]. Since the magnetic field induced behavior of the alloy depends on the martensitic structure, it is interesting to study the effect of the various activities on the structure and strain caused by magnetic field. The aim of the present study was to investigate the structure of non-stoichiometric Ni-Mn-Ga alloy and its change caused by magnetic field using various methods.

2. EXPERIMENTAL

The non-stoichiometric Ni-Mn-Ga polycrystalline alloy was manufactured in Outokumpu Research Center (Finland) and the single crystal material was melted in induction furnace in argon atmosphere at HUT. Material was homogenized in evacuated quartz capsules for 72 h at 1273 K and then ordered at

1073 K for 48 h followed with cooling in air. The polycrystalline sample **A** and single crystalline **B** with close chemical composition used for all the studies were cut with slow speed diamond saw parallel to the planes {100} i.e. perpendicular to the direction of crystallization. The specimens were ground and then electropolished and etched in 25 % HNO₃ - ethanol electrolyte at ambient temperature.

For the magneto-thermo-mechanical deformation (MTM), the samples were heated above of M_s temperature and then cooled slowly under the magnetic field 0.25 T to ambient temperature. During cooling a compressive stress $\sigma=0.8\text{MPa}$ was applied parallel to the direction of crystallization for sample **A** and orthogonal for **B**. Direction of the magnetic field was the same as the direction of compression for sample **A** and **B**.

The reversible ac-magnetic-field-induced strains were measured by strain gages, in magnetic field up to $B = 1$ T both along $\langle 001 \rangle$ and perpendicular to it. After SEM observation and magneto-mechanical treatment the field-induced strain was measured in a load cell equipped with a capacity sensor. Compressive stress was applied to the sample by piston, which was driven by pressurized gas from the cylinder. The changes of these strains were investigated with the parallel or perpendicular magnetic field.

The SEM study was carried out with JEOL/JSM 840A type scanning electron microscopy connected to EDS analysing apparatus with Sun Voyager 4.0 software. A 10keV accelerated voltage was used and at working distance of 39mm.

The optical differential scanning calorimeter measurements (DSC) were carried out using the Linkam 600 equipment that consists of a heating/cooling table that is connected to an optical microscope.

The X-ray theta-two-theta analysis was carried out at 293 K with the Philips diffractometer using Cu_K radiation with plane graphite monochromator. The samples were clamped to the special specimen holder so that their position remained constant during measurements. Investigations were thus applied to the fixed spot of plane {100} without rotation of the sample.

All the investigations for comparing of the microscopic structures and X-Ray results together with SEM were applied to the some fixed spots of the {100} planes of the samples.

3. RESULTS AND DISCUSSION

The optical differential scanning calorimeter reveals that the martensitic transformation occurs at ambient temperature, in sample **A** M_s is 306K and T_c 368K and in sample **B** those values are 305 K 370 K, respectively. Therefore, all the studies of the martensitic structure are carried out at the ambient temperature, i.e. below the M_s of the alloys.

The studied alloys have monoclinic lattice with $c>a$. This is in agreement with [5] in which the alloy with close chemical composition was determined to have tetragonal lattice with $c>a$. Fig. 1 shows orientation of the martensitic structure of the samples after MTM treatment. In Fig. 1, it is presented how the different direction of applied magnetic field and stress in MTM resulting different preferable martensite variants in sample **A** (Fig. 1a) and sample **B** (Fig. 1b curve 2). In the previous one, the preferable martensite variants were (400)+(040) and, in the latter one (004).

In Fig2, martensitic structures of samples **A** and **B** are presented. Fig. 2b and 2c show the martensitic structure change under static magnetic field (here strength and direction of the magnetic field are fixed, if either of them is changing, the field is dynamic field), which direction parallel easy magnetization direction, i.e. parallel studied surface. All existing martensitic structures change totally, it is verified by x-ray result that (001) variants change into (010) and (100) variants (Fig. 1b). In Fig. 1b, curve 2 shows the preferential (004) orientation of martensite (Fig. 1b, (1)) changes into the (400)+(040) orientation under magnetic field applied parallel to the studied surface (Fig. 1b, (2)).

It is discovered that both materials are sensitive to electron beam. In Fig. 3a, the martensitic structure is induced by magnetic cycling in sample **A**. While sample **A** is observed in SEM, the existing thin martensite lines grow up under electron beam (Fig. 3a and 3c). It is not clear whether this growth is caused by the magnetic field or by heat, because both of them have influence on the martensitic structure. In future more work need to study the effect of electron beam on martensitic structure of studied structure. According to the X-ray result in Fig. 1c, a new martensite variant occurs after SEM observation. For sample **B** so obvious structure change has not been observed, but the x-ray analysis result verified structure change by electron beam (Fig. 1d), It is noted that after two cycles the martensitic

structure becomes stable under electron beam. According to the magnetic induced strain curves, this structure that electron beam has induced, has quite small magnetic induced strain. In Fig. 4, the studied alloy **B** shows more than 5% field induced strain (Fig. 4a), as the value in the electron beam induced structure is less than 5%. Also, in polycrystalline alloy **A** where the field induced strain is 0.5% after MTM treatment (Fig. 4b), this value is decreased to 0.2% (Fig. 4c) for electron beam induced structure.

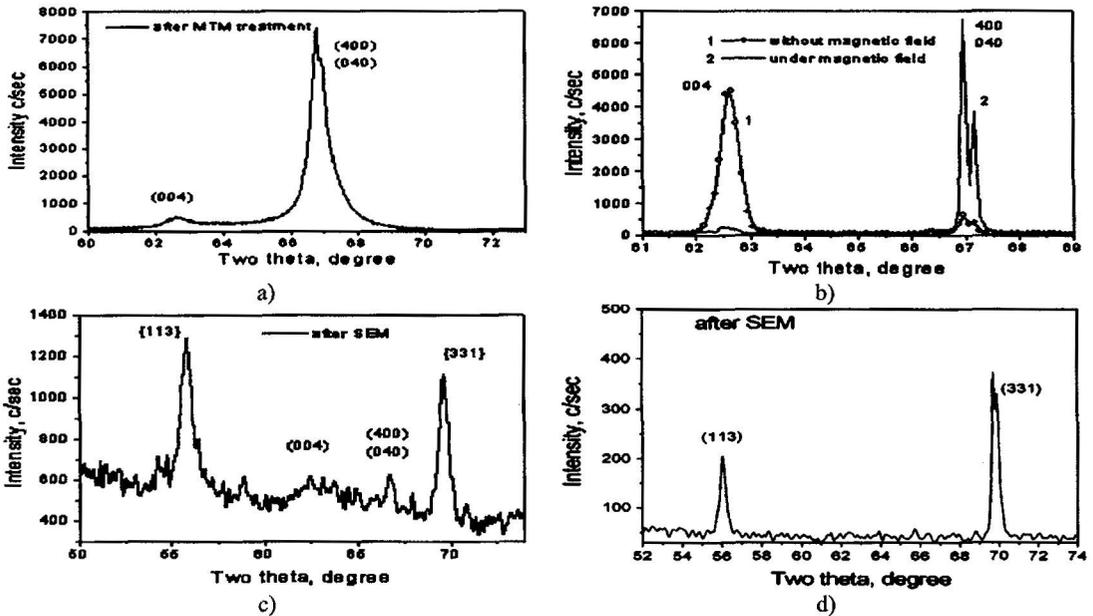


Fig. 1. X-ray diffraction patterns of the sample surface perpendicular to the direction of crystallization: (a) -sample A after MTM, (b) -sample B after MTM (1) and under applied magnetic field, (c) - sample A after SEM study; (d) -sample B after SEM study

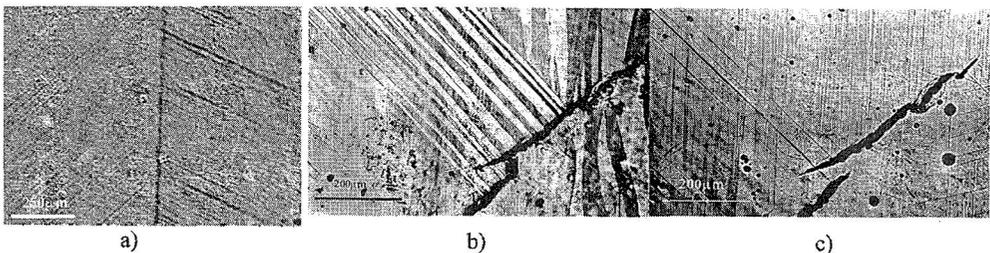


Fig. 2 Optic studies of the martensitic structure: a) sample A after MTM treatments; b) sample B after MTM treatment; c) sample B under static magnetic field parallel studied surface.

In Fig. 3d, 3e and 3f, the backscattered electron images (BEI) (the incident electron after elastic scattering with the nucleus of sample comes back out of sample is backscattered electron) of sample **B** are presented. Here in Fig. 3e, the easy magnetization direction is normal to the studied surface i.e. the same as it was in the magnetic field cycling. In Fig. 3f, the easy magnetization direction is parallel to the studied surface after magnetic cycling with compress, both of them parallel to the surface. It is noted that after magnetic cycling martensitic structures change only between two variant but in static field the change of martensitic structures depends on the previous structure.

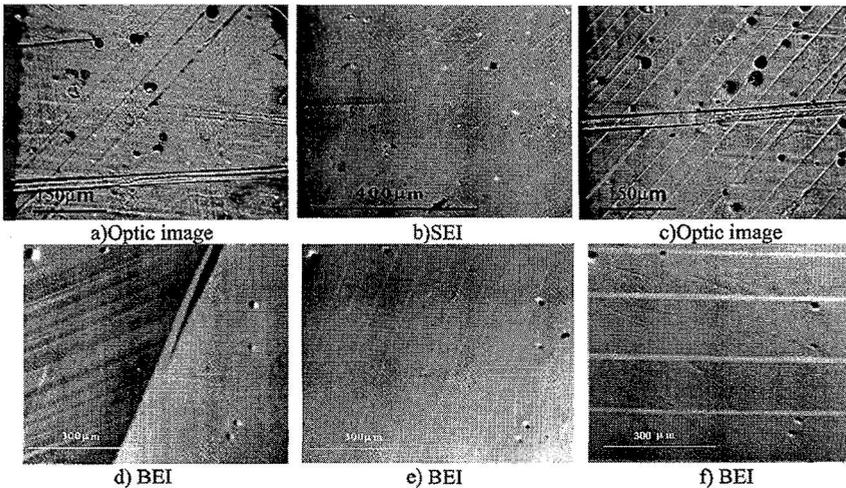


Fig. 3 martensitic structures a) optical study of sample A after magnetic cycling; b) sample A observed by SEM with secondary electron image (SEI); c) optical study of sample A after SEM studied; d) sample B after MTM treatment observed by SEM with BEI; e) sample B martensitic structure after magnetic cycling observed by SEM with BEI; f) sample B after magnetic and compress treatment observed by SEM with BEI.

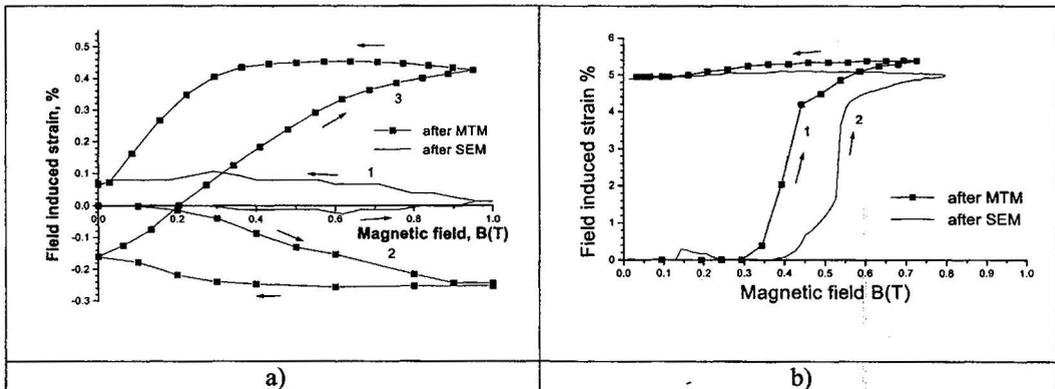


Fig. 4 Field induced strain. (a) **Sample A**: 1, 3 field direction parallel to strain direction; 2 field direction normal to strain direction. (b) **Sample B** field direction normal to strain direction

Fig. 5 shows the change of martensitic structure under static magnetic field. The studied surface of sample A has a big twin boundary (from Fig. 5. a1-a4), where big twin band is composed of thin martensite stripes. Under normal applied magnetic field thin martensite stripes in both bands change, but their behaviours are different. Some martensite stripes in left band change direction (the lower-left in Fig. 5 a2). Martensite stripes in right band combine, distance between stripe increases, and the length decreases. Big twin boundary moves to left. Strong relief make left band very dark and some martensite stripes in right band go through twin boundary into left band, these phenomena suggest martensitic structures change accompany with big strain. When applied magnetic field direction is parallel to the horizontal direction, big twin boundary continues moving to left. Martensite stripes in right band nearly disappear and strong relief makes left band difficult to see. When applied magnetic field direction is parallel to vertical direction, twin boundary continues moving to left. A new martensite stripe appears (Fig. 5. a4 lower-right).

In Fig. 5 b, the behaviour of big twins of sample B under static magnetic field are presented. It is very clear that twin boundaries are moving with magnetic field direction. The high magnification reveals that

those big twin bands are composed of thin martensites. Good contrast of big twins also suggest big strain in accompany with twin boundary motion. Fig. 5 c shows the behaviour of thin martensite stripes in sample B under static magnetic field. This behaviour seems to be independent of applied magnetic direction. In the process of study (c1-c4), thin martensite stripes continue to combine. The average of distance between martensite stripes is from c1 to c4, 5 μ m, 10 μ m, 15 μ m, 20 μ m respectively.

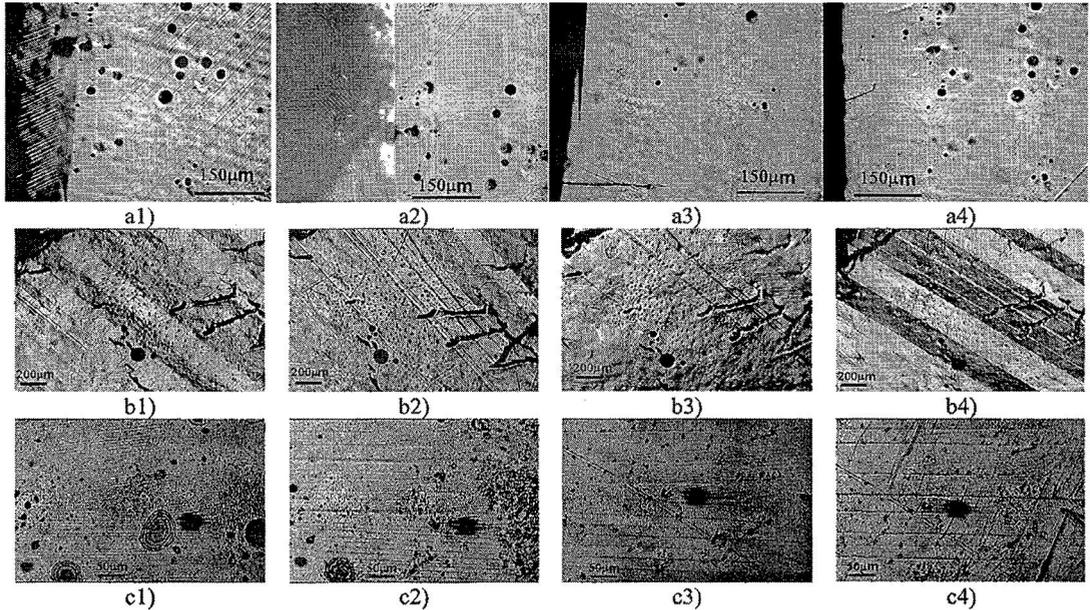


Fig. 5 The change of the martensitic structure in sample A (one spot: a1-a4) and B (two spots: b1-b4 and c1-c4) caused by fixed magnetic field. Magnetic field directions 1: without magnetic field; 2: magnetic field direction normal to studied surface; 3: magnetic field parallel to studied surface to the horizontal direction; 4: magnetic field direction parallel to studied surface to the vertical direction.

According to those results, it is supposed that the big twin boundary motion is driven by strain accompany with thin martensitic structure changes. The big twin boundary is interface of two martensite variants, where is stress balance area between two variants. When the change of martensitic structure is caused by magnetic field, the stress balance near twin boundary area is disturbed and the motion of twin boundary is needed to re-establish this balance.

4. CONCLUSION

In the present study, the changes of martensite in magnetic field are studied for non-stoichiometric Ni-Mn-Ga alloy both with polycrystalline and single crystal structure. With the single crystal alloy a shape change above 5 % was obtained in the magnetic field, while with the polycrystalline specimen this was only 0.5 %. It is found that in the electron beam induced structure these field induced strains decrease further.

In the studied materials, the shape change is obtained by changing of the martensite variants. These variants are separated from each other with a big twin that moves in the applied magnetic field. This motion of the big twin boundary is connected to the change of very thin martensitic structure.

The effects of dynamic and static magnetic field on martensitic structure are different from each other. For dynamic magnetic field martensitic structures only change between two variants but in static magnetic field the change depends on the previous martensitic structure.

ACKNOWLEDGEMENTS

The funding support of National Technology Agency (Tekes), Finland, and the consortium of Finnish companies (Outokumpu Oyj, ABB Oyj, Metso Oyj, AdaptaMat Oyj) as well as the Academy of Finland for this work is gratefully acknowledged. For the material the authors would like to thank Lic.Tech. Tuija Suortti and M.Sc. Juha Järvi, Outokumpu Research Center, and Mr. Olli Mattila and Mr Tarmo Viren from HUT. Authors would like to thank also Mr. I Glavatsky and Mr. Ari Jääskeläinen together with Mrs. Pirjo Kirpiala for their help.

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