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TEMPERATURE DEPENDENCE OF THE MAGNETIZATION OF THE $Ni_{52}Mn_{24}Ga_{24}$ ALLOY IN VARIOUS STRUCTURAL STATES

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Results are presented of a study of the temperature dependence of the magnetization σ(T) of the polycrystalline $Ni_{52}Mn_{24}Ga_{24}$ *alloy in various structural states: in the initial coarse-grained state, after severe plastic deformation by high pressure torsion, and after stepped annealing of the deformed specimen at temperatures from 200 to 700°C for 30 min. As a study of the σ(T) curve shows, in an alloy possessing a coarse-grained initial structure, a martensitic phase transition and a magnetic phase transition are observed in the room temperature interval. The martensitic transformation takes place in the ferromagnetic state of the alloy. This transformation is accompanied by an abrupt lowering of the magnetization of the material, associated with a lowering of the symmetry of the crystalline lattice and a high value of the magnetocrystalline anisotropy constant of the alloy in the martensitic phase. It is shown that as a result of plastic deformation there takes place a destruction of ferromagnetic order and a suppression of the martensitic transformation. Consecutive annealing after deformation leads to a gradual recovery of ferromagnetic order and growth of the magnetization of the material. Recovery of the martensitic transformation begins to be manifested only after annealing of the alloy at a temperature of 500°C, when the mean grain size in the recrystallized structure reaches a value around 1 μm.*

Keywords: ferromagnetic shape-memory alloys, Heusler alloys, martensitic transformation, severe plastic deformation.

INTRODUCTION

Heusler alloys of the Ni_2MnGa system possess unique properties, thanks to which they are considered to belong to the class of functional materials. In alloys of this system such effects as the ferromagnetic shape-memory effect (FSME) [1–4] and the magnetocaloric effect (MCE) [5–7] are observed, along with others. Their existence is due to a martensitic transformation taking place in the room temperature interval. The main attraction in alloys of the Ni_2MnGa system is the FSME, i.e., the possibility of controlling the shape and dimensions of a material with the help of an external magnetic field. In polycrystalline alloys the magnitude of this effect is about 1% [8]. This opens up wide perspectives of putting these alloys to use in various sorts of microelectronic devices and actuators. However, they have two important drawbacks: degraded mechanical properties and instability of the shape-memory effect. Upon cyclic heating and cooling of the specimen through the martensitic transition temperature, the structure gets destroyed. As is well known, applying thermomechanical processing to various metals and alloys leads to enhanced mechanical properties [9–14]. In the case of the examined alloys such processing increases the stability of the functional characteristics of the material. In other words, the alloy endures a large number of cycles of the martensitic transformation. To better understand how to enhance the functional characteristics of alloys of the given system by the

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method of deformational-thermal processing, a systematic study of the physical properties and structure of a material subjected to such treatment is needed. In this paper, we present results of a study of the magnetic properties and microstructure of the polycrystalline $Ni_{52}Mn_{24}Ga_{24}$ alloy in various structural states obtained by high pressure torsion with subsequent stepped annealing at temperatures in the range 200–700°C. We shall refer to this process in what follows as high pressure torsion (HPT). The present work is a continuation of previously published papers [15, 16].

MATERIALS AND METHODS

As the material to investigate we chose the polycrystalline $Ni_{52}Mn_{24}Ga_{24}$ alloy. The alloy was obtained by arc melting in an Ar atmosphere from high purity Ni, Mn, and Ga. To achieve homogenization, the specimens were annealed for 9 days at 827°C with subsequent quenching in water with melting ice. An ingot of the alloy has an elongated shape (in the form of a bar) since crystallization of the melt took place in an elongated copper crucible with a depression and dimensions 10×100 mm^2.

The specimens were subjected to HPT on Bridgman anvils. The pressure was about 5 GPa, torsion was performed for 5 revolutions at a rate of 2 rpm. After deformation, all of the specimens had the shape of disks with a diameter of about 10 mm and a thickness of 0.1–0.15 mm.

The martensitic transformation was investigated by recording the curves of the temperature dependence of the magnetization in a magnetic field around 240 kA/m. This was done by heating the specimen in the existence interval of the martensitic and magnetic phase transformations. The magnetization of the specimen was measured using an automated magnetic microbalance.

The grain structure of the alloy after deformation and annealings at various temperatures was examined with the help of a scanning electron microscope and a sensor that was sensitive to the orientational contrast of the material. This is the most convenient method for examining small-grain structure as it does not require the laborious preparation of thin foils and makes it possible to analyze a large area of the surface of the specimen. After polishing with sandpaper with different degrees of roughness, the examined surface was subjected to electrolytic polishing.

EXPERIMENTAL RESULTS

The temperature dependence of the magnetization of the polycrystalline $Ni_{52}Mn_{24}Ga_{24}$ alloy in its initial cast state, recorded during heating of the specimen in a magnetic field of 240 kA/m, is shown in Fig. 1 (curve *1*).

It is clear from the curve of $\sigma(T)$ that an abrupt increase in the magnetization is observed in the room temperature interval due to occurrence of the reverse martensitic transformation. Such behavior of the curve in magnetic fields below 500 kA/m is characteristic of alloys of the given system. This is explained by the low symmetry of the crystalline structure and the high value of the magnetocrystalline anisotropy constant of the martensitic phase. With further increase of the temperature, a smooth decrease of the magnetization is observed in the curve, and at 125°C the transition to the paramagnetic state takes place.

The microstructure of the alloy in the initial state at room temperature is shown in Fig. 2. Microcracks are observed in the structure, passing along the grain boundaries. They are about 0.5 μm wide. As a result of repeated cycles of the martensitic transformation, a piling up of defects takes place in them. In the final count, this leads to growth of the cracks and destruction of the material. In the body of a grain there are also observed bands of varying thickness. These are martensitic twins.

Temperature dependences of the magnetization of the alloy after HPT and subsequent stepped annealings at temperatures in the interval 300–700°C over the course of 0.5 h are plotted in Fig. 1 (curves *2–6*). As can be seen, after severe plastic deformation magnetization of the specimen in the measured temperature interval is practically absent (curve *2*). An analogous phenomenon was also observed in [17, 18]. This effect is explained by fragmentation of the grain structure of the material and by the high density of defects observed after severe plastic deformation. It is well known that a highly defective crystalline structure hinders ferromagnetic ordering [19, 20]. Along with destruction of ferromagnetic ordering, the martensitic transformation in the alloy is also suppressed. Curve *3* corresponds to the state

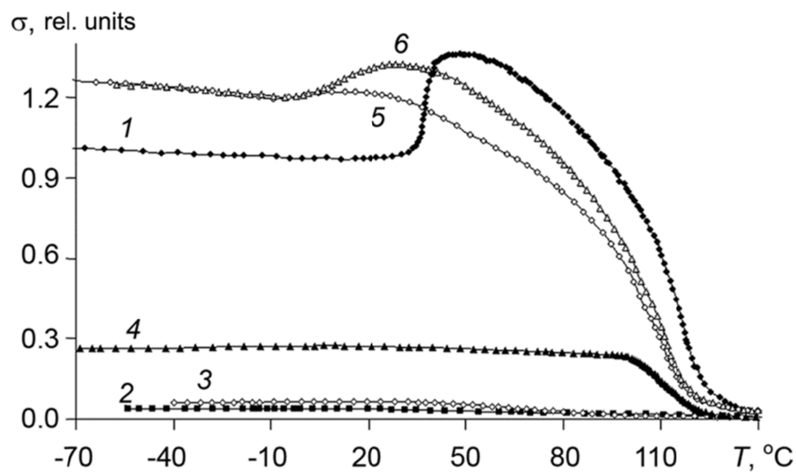

Fig. 1. Temperature dependences of the specific magnetization of the polycrystalline $Ni_{52}Mn_{24}Ga_{24}$ alloy in various structural states: curve *1* corresponds to the initial state, curve *2* is for the alloy after HPT, curve *3* corresponds to HPT and annealing at 300°C, curve *4* corresponds to HPT and annealing at 400°C, curve *5* corresponds to HPT and annealing at 500°C, and curve *6* corresponds to HPT and annealing at 700°C.

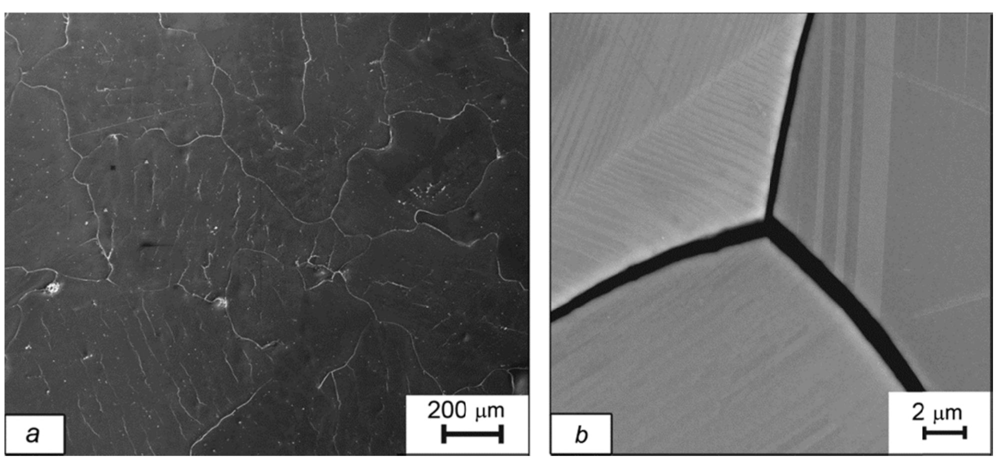

Fig. 2. Microstructure of the $Ni_{52}Mn_{24}Ga_{24}$ alloy in the initial state.

after annealing at a temperature of 300°C. A small value of the magnetization is observed, corresponding to about 5% of the initial state. This change indicates that at this annealing temperature, recovery processes start to take place in the material, to some extent lowering the number of defects of the structure. Recovery is accompanied by a lowering of the density of highly mobile point defects and dislocations.

Subsequent annealing of the specimen at 400°C (curve *4*) leads to a marked increase in the magnetization of the material; however, here also the magnetization of the specimen at temperatures below the Curie point amounts to only 20% of its value in the martensitic phase in the initial state. Growth of the magnetization of the specimen takes place as a consequence of a significant lowering of the density of defects and microstresses in the specimen as a result of annealing. There takes place a lowering of the defect density of the structure, as a result of which ferromagnetic ordering becomes possible. However, in the σ(*T*) curve in the existence region of the martensitic transformation no significant change in the magnetization of the specimen is visible, which would tend to indicate that the martensitic transformation is still suppressed.

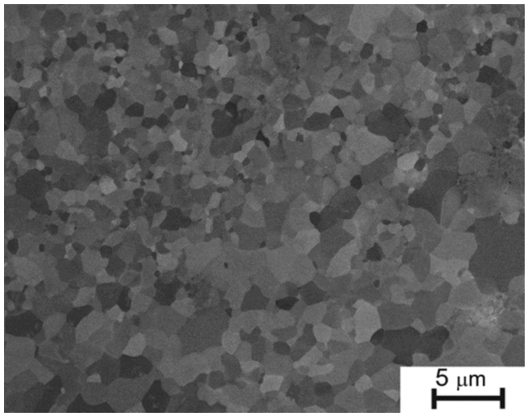

Fig. 3

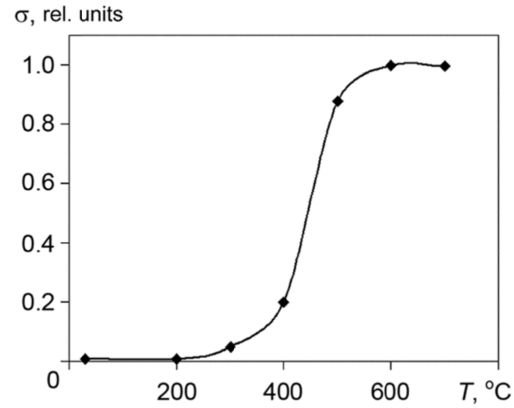

Fig. 4

Fig. 3. SEM image of the microstructure of the $Ni_{52}Mn_{24}Ga_{24}$ alloy after HPT and annealing at 500°C.

Fig. 4. Magnetization of the alloy at 30°C after annealings at different temperatures.

Annealing of a deformed specimen at 500°C for 30 min almost completely recovers the ferromagnetic properties of the material (curve *5*). In the temperature interval from –10°C to +10 °C there is observed a small growth, which indicates that the martensitic transformation begins to be partially realized. Microstructural studies show that in the structure of a specimen after annealing at such a temperature there is observed a recrystallized structure with average grain size around 1 μm. A corresponding SEM image is shown in Fig. 3. The spread in the grain sizes speaks to the fact that recrystallization of the structure proceeds in a nonuniform fashion.

The curve of the temperature dependence of the magnetization of the specimen annealed at 700°C for 30 min shows that growth in the interval from –10°C to +10°C is manifested progressively more distinctly (curve *6*). This speaks of a still more significant recovery of the martensitic transformation.

The combined data on changes in the magnetic properties of the alloy as a result of heat treatment after HPT are shown in Fig. 4, which presents the dependence of the magnetization at 30°C on the annealing temperature of the specimen. This graph allows us to identify the dynamics of variation of the magnetization during annealing. It can be seen that the greatest changes in the magnetic properties occur after annealing at 500°C. Such changes are obviously due to the process of recrystallization, i.e., to generation and growth of new grains with small defect density of their structure. As a consequence of replacement of deformed grains by new grains and lowering of the number of defects, the ferromagnetic properties of the alloy are recovered throughout the entire bulk of the specimen.

CONCLUSIONS

A study of the magnetic properties and microstructure of the polycrystalline $Ni_{52}Mn_{24}Ga_{24}$ alloy in its initial cast state and in states obtained by severe high pressure torsion followed by stepped annealings at temperatures of 300–700°C has allowed us to trace out the passage of the martensite–austenite phase transition responsible for the shape-memory effect, to trace out changes in the structural state of the material during annealing, and to determine the temperature at which recrystallization commences.

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