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COMPARISON OF INFLUENCE FORGING AND EXTRUSION ON MICROSTRUCTURE OF HEUSLER ALLOYS

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Abstract. The results of investigation of the microstructure of two polycrystalline Ni-Mn-Ga alloys subjected to thermo-mechanical treatment by multiple isothermal forging and extrusion are presented. Alloy forging at a temperature of 680°C and 700°C leads to the formation of a bimodal structure which has large grains of several hundred micrometers surrounded by a layer of the fine-grained structure. As the result of the further treatment by extrusion at 710°C the volume fraction of the fine-grained structure is increased. At the same time, anisotropy of thermal expansion during the martensitic transformation is observed in the alloys in both states due to residual tensile stresses introduced in the last stages of treatment. The performed investigation shows high potential of the thermo-mechanical treatment for obtaining structurally modified Heusler alloys having a sufficient level of functional and service properties for practical applications

Keywords: Heusler alloys, martensitic transformation, thermo-mechanical treatment, multiple isothermal forging, extrusion, EBSD, texture, anisotropy

1. Introduction

In Heusler Ni-Mn-*X* (*X*=Ga, In, Sn, Fe, Co, etc.) alloys at the martensitic transformation the ferromagnetic shape memory effect and magnetocaloric effect are observed [1-7]. An irreversible change by 11% in the geometric dimensions of the sample in the magnetic field of 1 T [8] is observed in a single-crystal alloy. Significant values of the effects allow referring the alloys to promising functional materials. In the polycrystalline state, this value is an order of magnitude smaller, which is still enough to manufacture of controlled elements in various actuators and microelectronics. At the same time, a big disadvantage of polycrystalline samples is accumulation of the dislocations and other defects at the repeated martensitic transformation. It leads to a sharp embrittlement and destruction of the alloy. Accordingly, to increase the fatigue property of the alloy, it is required to obtain a structure with a lower frailty by formation of an impediment for origination and growth of cracks.

The thermo-mechanical treatment (TMT) is one of the most effective ways to influence the structure of metals and alloys. Especially it is achieved by using methods of large plastic deformations, such as high pressure torsion [9-12], rolling [13-17], and deformation by upsetting [18, 19]. However these methods allow obtaining the workpiece only of limited size, mainly in the form of thin tapes or plates.

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The authors successfully develop the method of the plastic deformation of Heusler alloys by the multiple isothermal forging (MIF) [20, 21]. The advantage of this method is obtaining a bulk billet of the processed material with sufficiently large internal stresses in the volume of the workpiece. It is known being necessary to form a crystallographic texture and tensile-compressive stresses at the treatment of the alloys by various methods. It enhances the anisotropy of a change in sample's dimensions during martensitic transformation. This increases the ferromagnetic effect of shape memory. For this reason, drawing is performed at the last stages of forging. It means that forging is carried out only in two directions and the workpiece is elongated in one direction.

The extrusion is another way to obtain a sharp texture in material. However, the treatment of Heusler alloys in the as-cast state does not lead to significant changes in the microstructure. Therefore, it is necessary to prepare the structure of the billet prior to extrusion. It was carried out by MIF. The paper presents the comparative analysis of the microstructure of the Heusler alloy subjected to two types of treatment: forging as-cast state and complex treatment by forging and follow extrusion. For investigation the Ni2MnGa alloys were chosen. It is a well-studied system and may be considered as a model object. In these alloys all kinds of physical properties at the martensitic transformation are studied and analyzed. The results of our investigation will allow us to correlate them with other Heusler Ni-Mn-*X* (*X*=Ga, In, Sn, Fe, Co, etc.) alloys.

2. Material and methods

Two Ni-Mn-Ga alloys for the investigation were prepared by the arc-melting method under argon atmosphere. The elemental composition was analyzed by the energy dispersive X-ray spectroscopy on a scanning electron microscope TESCAN Vega 3 SBH. The first alloy marked as Ga16/6 has the composition $Ni_{54.1}Mn_{19.6}Ga_{24.6}Si_{1.7}$, the second alloy marked as Ga17/3 - $Ni_{52.9}Mn_{21.1}Ga_{24.6}Si_{1.5}$. It should be noted that two alloys with different composition (~1%) were compared. However, it is known that such difference in composition leads only to a slight difference in the martensitic transformation temperature, and physical properties are not changed significantly.

Presence of Si in the alloy is explained by its diffusion from the quartz glass during vacuum remelting. The detailed information about this procedure is described in the previous works [20, 21]. The Ga16/6 alloy in the as-cast state was subjected to TMT by MIF at 680°C, and Ga17/3 alloy in the as-cast state was subjected to complex TMT by MIF at 700°C with the following extrusion at 710°C. The temperature of the martensitic transformation was determined by measuring of temperature dependence of thermal expansion. The measurements were performed on the samples of 1 mm×1 mm×7 mm by the dilatometer based on differential transformer. The microstructure of the alloys was investigated by scanning electron microscope TESCAN Mira 3 LMH in the backscattered mode. EBSD analysis was carried out on this microscope with Channel 5 software. Accelerating voltage was 20 kV. Multiple isothermal forging was carried out on the machine of complex loading Schenck Trebel RMC 100. Forged alloy was also extruded on a special tool, in which the output circular section has a transition 10 mm→8 mm with the extrusion ratio of 1.6.

3. Results

3.1. Temperature dependence of the thermal expansion of Ga16/6 alloy after multiple isothermal forging. The temperature dependence of thermal expansion of Ga16/6 subjected the thermo-mechanical treatment by MIF at 680°C is presented in Fig. 1. The drawing of the workpiece at the latest stages of forging should form a crystallographic texture and tensile stresses, which results in the anisotropy of properties. Therefore the sample for measuring was cut along the drawing axis of forged workpiece. Heating and cooling of the sample was carried out in the temperature range -100°C ÷ -20°C. As it can be seen, at the martensitic transformation a sharp anisotropy of thermal expansion is observed. In the process of direct martensitic transformation, the sample is abruptly reduced by 0.13%. During the reverse martensitic transformation, this deformation is recovered. Typical martensitic transformation temperatures have the following values: M_S= –78°C; M_F= –89°C; A_S= –77°C; A_F = –65°C. The length changing during heating and cooling in other intervals occurs according to the anharmonic law.

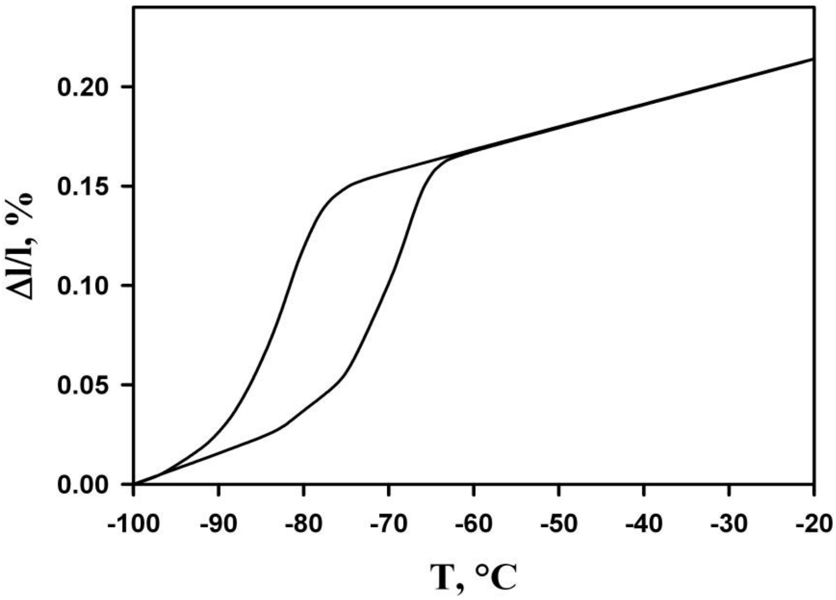

Fig. 1. Temperature dependence of the thermal expansion of Ga16/6 alloy along drawing axis after MIF at 680°C

Thus, the established anisotropy of thermal expansion at the martensitic transformation confirms the formation of deformation texture and tensile-compressive stresses during forging. In the process of drawing of the workpiece in the last stages of the MIF the residual compressive stresses are formed normal to the axis of the treatment. As shown earlier [22, 23] the anisotropy of the thermal expansion of the Heusler Ni_2MnGa alloy is subjected to the formation of a preferential orientation of the martensitic twins at the phase transformation. At the same time, for the formation of such structure, it is necessary to have both a crystallographic texture and residual compressive or tensile stresses in the crystal lattice.

3.2. Temperature dependence of the thermal expansion of Ga17/3 alloy after complex treatment by forging and extrusion. The temperature dependence of thermal expansion of Ga17/3 subjected the complex thermo-mechanical treatment by MIF at 700°C and the following extrusion at 710°C is presented in Fig. 2. The sample for measuring, as well as the forged alloy, was cut along the treatment axis from the central part of the workpiece. Measurement was carried out in the field of martensitic transformation. An abrupt change of the geometric dimensions of the sample is observed at the phase transformation. A reduction of the length by 0.05% is observed at the direct transformation. The sample of alloy subjected only to forging has a similar nature. The strain is recovered at the reverse transformation. The length changing during heating and cooling in other intervals occurs according to the anharmonic law. Typical martensitic transformation temperatures have the following values: M_S= –85°C; M_F= –109°C; A_S = –99°C; A_F = –74°C. There are breaks of the heating and cooling curves at the martensitic transformatiom. The reason is transformation in different phases: the big grains phase and the small grains phase. And it is known than thermo-mechanical treatment of Heusler alloy leads to a shift of the martensitic transformation to low temperatures [24].

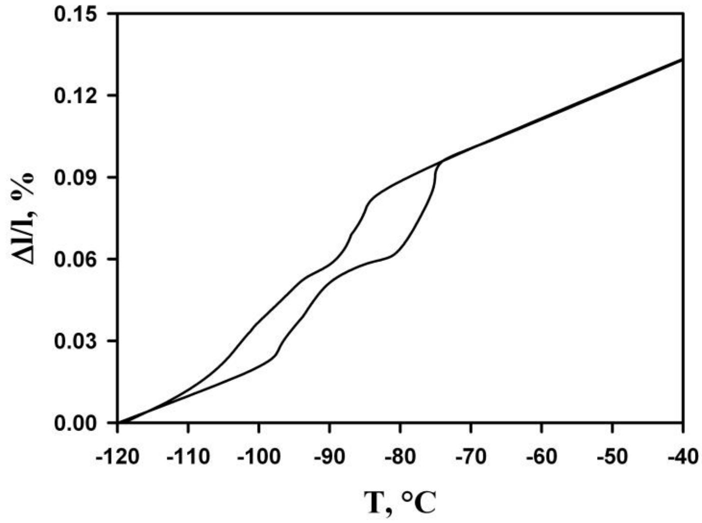

Fig. 2. Temperature dependence of the thermal expansion of Ga17/3 alloy along the extrusion axis after MIF at 700°C and extrusion at 710°C

3.3. Microstructure of Ga16/6 alloy after forging. The microstructure of the forged alloy in the parallel section to treatment axis is shown in Fig. 3 a.

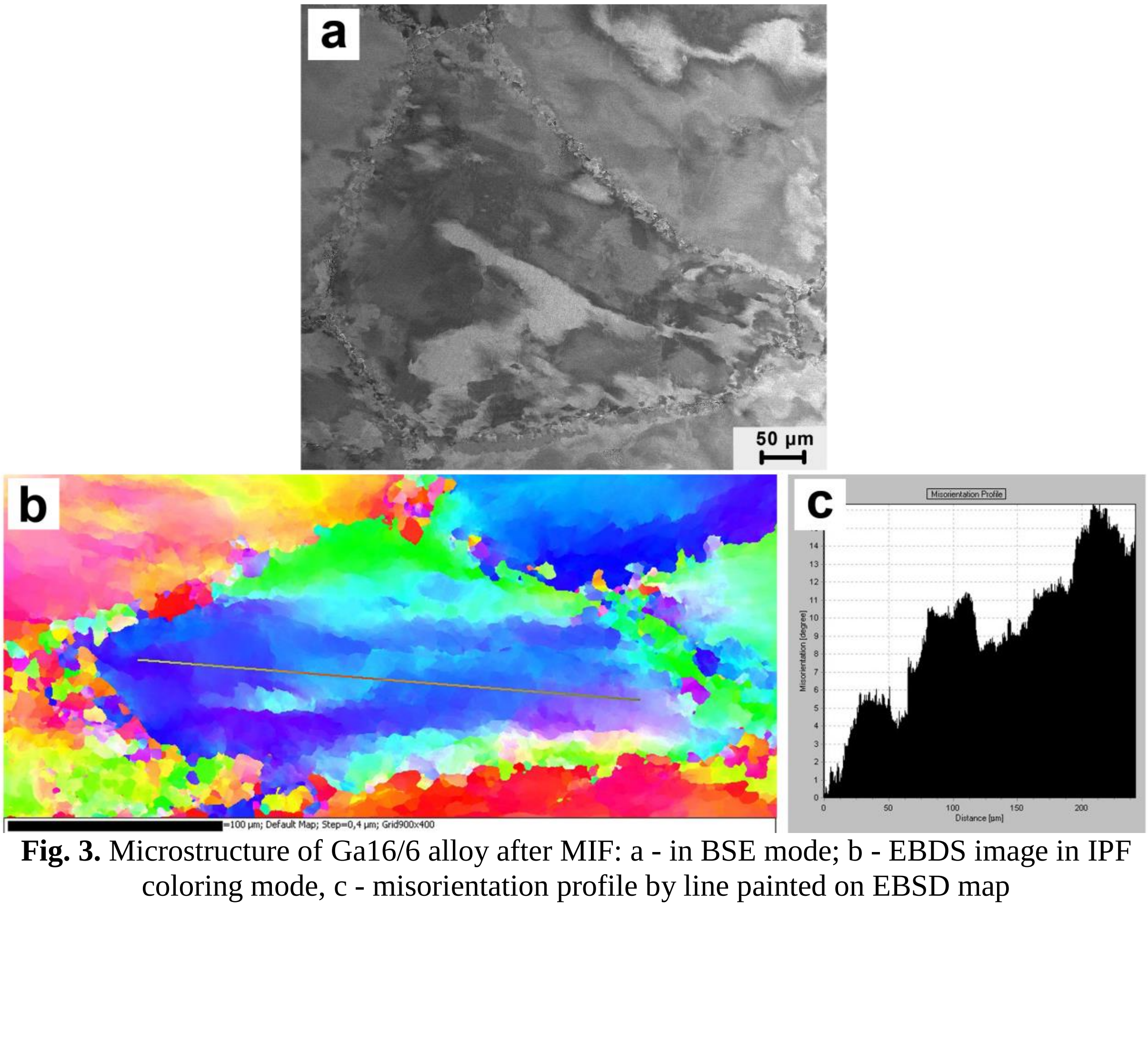

Fig. 3. Microstructure of Ga16/6 alloy after MIF: a - in BSE mode; b - EBDS image in IPF coloring mode, c - misorientation profile by line painted on EBSD map

The image is obtained by scanning electron microscope in BSE mode. As a result of the treatment by MIF the as-cast structure with a grain size of 200-400 μm is transformed into a bimodal structure in which the large grains of about 100-200 μm are surrounded by a fine-grained structure. The formation of new grains on the boundaries is occurred in the process of treatment by the mechanism of discontinuous dynamic recrystallization. In the result the areas with fine-grained structure in the border zones are formed. A clear contrast between the grains of the fine-grained structure indicates high-angle misorientations between the grains. There is a characteristic contrast in the body of large grains, which indicates presence of large residual stresses or substructure. Thus, the necessary stresses are concentrated in large grains, which must perform the functional assignment (changing the size of the alloy sample).

The EBSD analysis was carried out for the purpose of more detailed analysis of orientations, texture and residual stresses in the treatment alloy. A local area of 0.36 mm×0.16 mm with the step size of 0.4 μm is presented in Fig. 3 b. The map of orientations is shown in IPF coloring mode. The nature of the results corresponds to the data obtained in the study in the BSE mode. There is a clear color grain contrast in the fine-grained structure. In the large grain the elongated curved shape with different contrast is observed. The Misorientation Profile was used for the detailed analysis changes in orientation along a line in the body of the large grains. It is indicated in Fig. 3 b by a straight line. The Profile is shown in Fig. 3 c. The histogram shows the orientation of points relative to the first point rather than the previous one. It can be seen that the last points of the profile are disoriented relatively to the first at angles of about 15°. Thus, the EBSD analysis data correlates with the results of the microstructure analysis in the BSE mode.

A EBSD map of the entire section of the workpiece with the area of 2.2 mm×9.8 mm and the step size of 3 μm is made for the analysis of the crystallographic texture. It allows evaluating the structure of the entire volume of the workpiece. A map of crystallographic orientations in IPF coloring mode is shown in Fig. 4 a. It is seen that the nature of the structure is the same throughout the section of the workpiece. In general, the structure with bimodal grain distribution is homogeneous for the workpiece. The Pole Figures (PF) are calculated from the EBSD data for the analysis of the crystallographic texture. At the same time, the large and the small grains are divided into separate subsets by software for separation of their textures. The PF for large grains is shown in Fig. 4 b, the PF of fine grain structure is shown in Fig. 4 c. The PF are presented for the {100} and {111} planes, since it most fully reflects the nature of the texture of the cubic lattice. Of course, the standard analysis of the crystallographic texture of the large grains is not sufficient for the statistical sampling. However, despite the presence of some localized maxima it is still clear that a significant texture is absent. In case of fine-grained structure, at least three localized maxima are observed in the central part, the upper and lower poles of the stereographic plane. These maxima correspond to the same orientation of the cubic unit cell of the crystal. At the same time, there are no localizations at the points of these maxima for the PF of the coarse grains. This nature of the orientation allows us to conclude that there is the crystallographic texture of the fine-grained phase. Thus, in the MIF process, the orientation of the large grains has not changed. As a result of intermittent dynamic recrystallization, new fine grains have received a slight texture.

a)

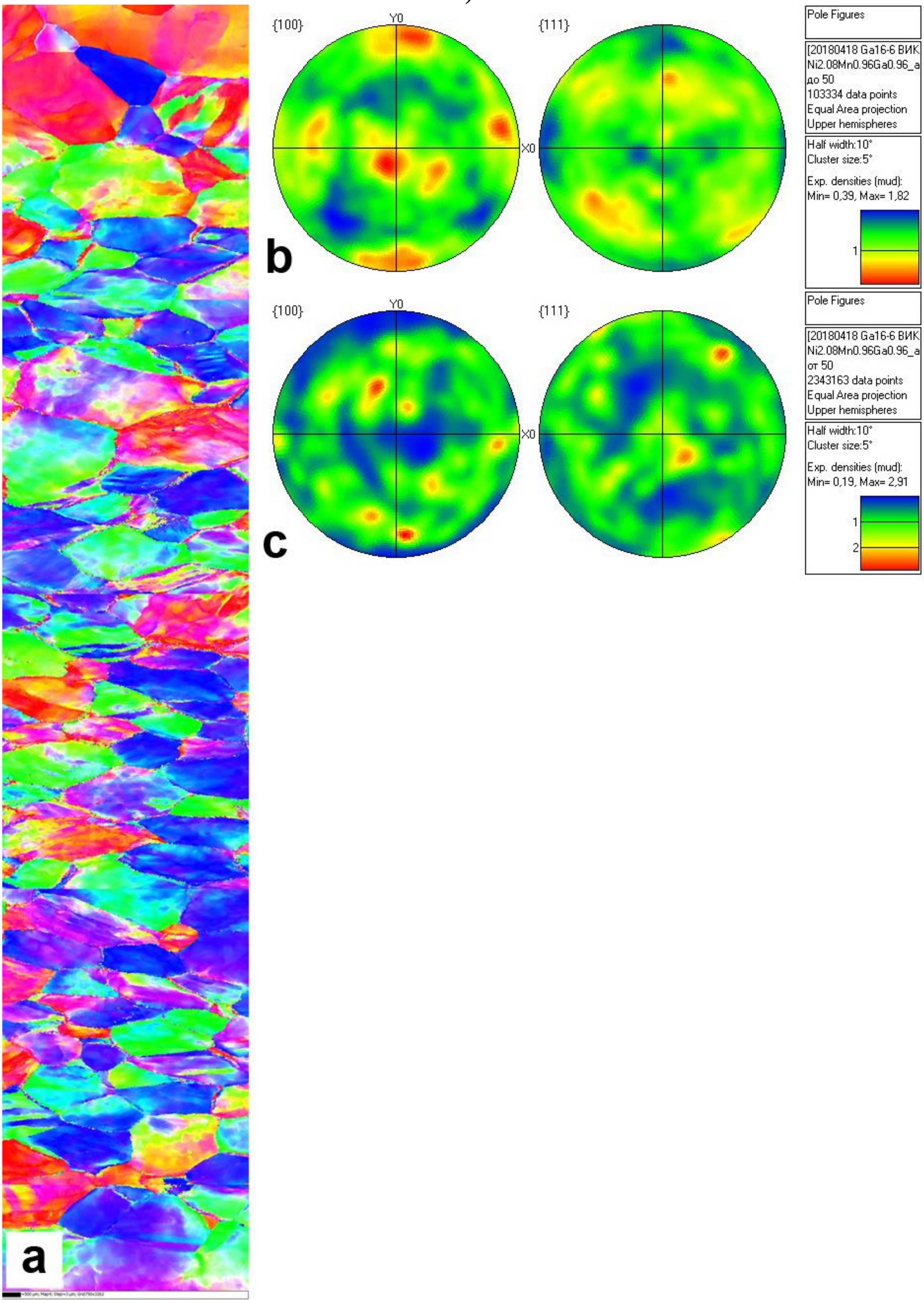

Fig. 4. EBDS analysis of Ga16/6 alloy after MIF: a - EBDS image in IPF mode, b - Pole Figures for small grains, c - Pole Figures for big grains

3.4. Microstructure of Ga17/3 alloy after forging and extrusion. The microstructure study after complex treatment (MIF + Extrusion) was performed in the plane along the extrusion axis. The image of the microstructure in BSE mode is presented in Fig. 5 a. It shows that the microstructure after the complex treatment is similar to the microstructure after forging. The large grains with a size of 100-200 μm are surrounded by the fine-grained structure. The fine-grained structure is characterized by a fairly clear contrast between neighboring grains. The boundaries are straight and thin. This indicates a high-angle misorientation of the grains. In the body of large grains there are areas without clear boundaries and having weak diffuse contrast. It indicates the presence of residual stresses and substructures. The main difference of the structure after the complex treatment is the increase of the volume fraction of the fine-grained structure. The layer of small grains has the width about 10 grains. Thus, the extrusion leads to an increase in the volume fraction of the fine-grained structure.

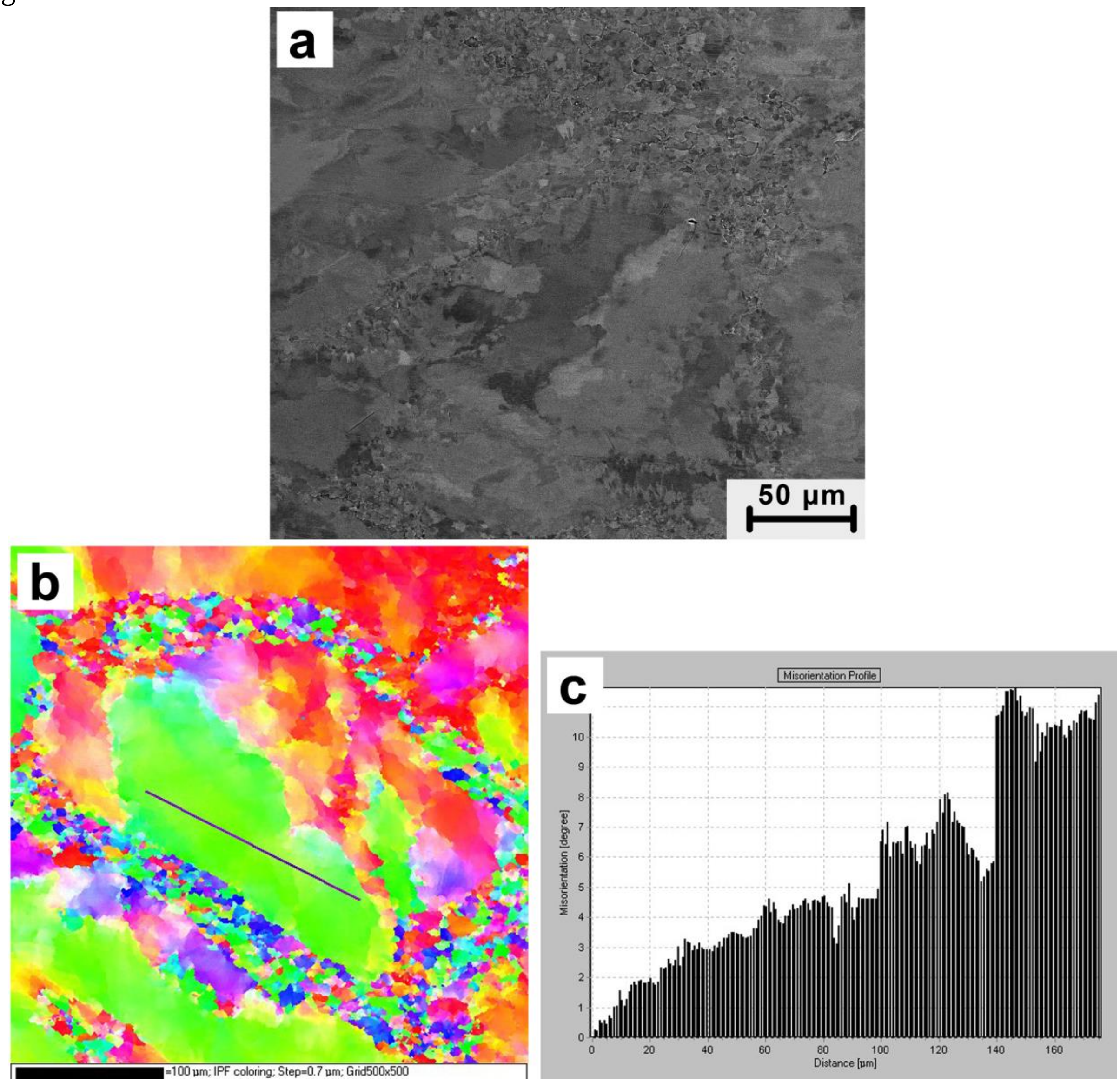

Fig. 5. Microstructure of Ga17/3 alloy after MIF and extrusion: a - in BSE mode; b - EBDS image in IPF mode, c - misorientation profile by line painted on EBSD map

An EBSD analysis was performed for the purpose of a more detailed analysis of the orientations, texture and internal stresses in the treatment alloy. The map for a section of 0.35 mm×0.35 mm with the step size of 0.7 μm is shown in Fig. 5 b. The data are presented in

IPF coloring mode. The EBSD data confirmed the results of the analysis of the structure in BSE mode. The fine-grained structure also has high-angle misorientations. The substructure of coarse grain is confirmed by the misorientation profile marked in the grain body and indicated in Fig. 5 b. The histogram is presented in Fig. 5 c. The histogram shows the orientation of points relative to the first point rather than a previous one. The extreme points of the profile are misoriented by 11°.

The EBSD analysis along the extrusion axis was performed at the entire section of the workpiece to evaluate the crystallographic texture in the material. The data for area of 2.1 mm×8.4 mm with the step size of 3 μm are shown in Fig. 6 a.

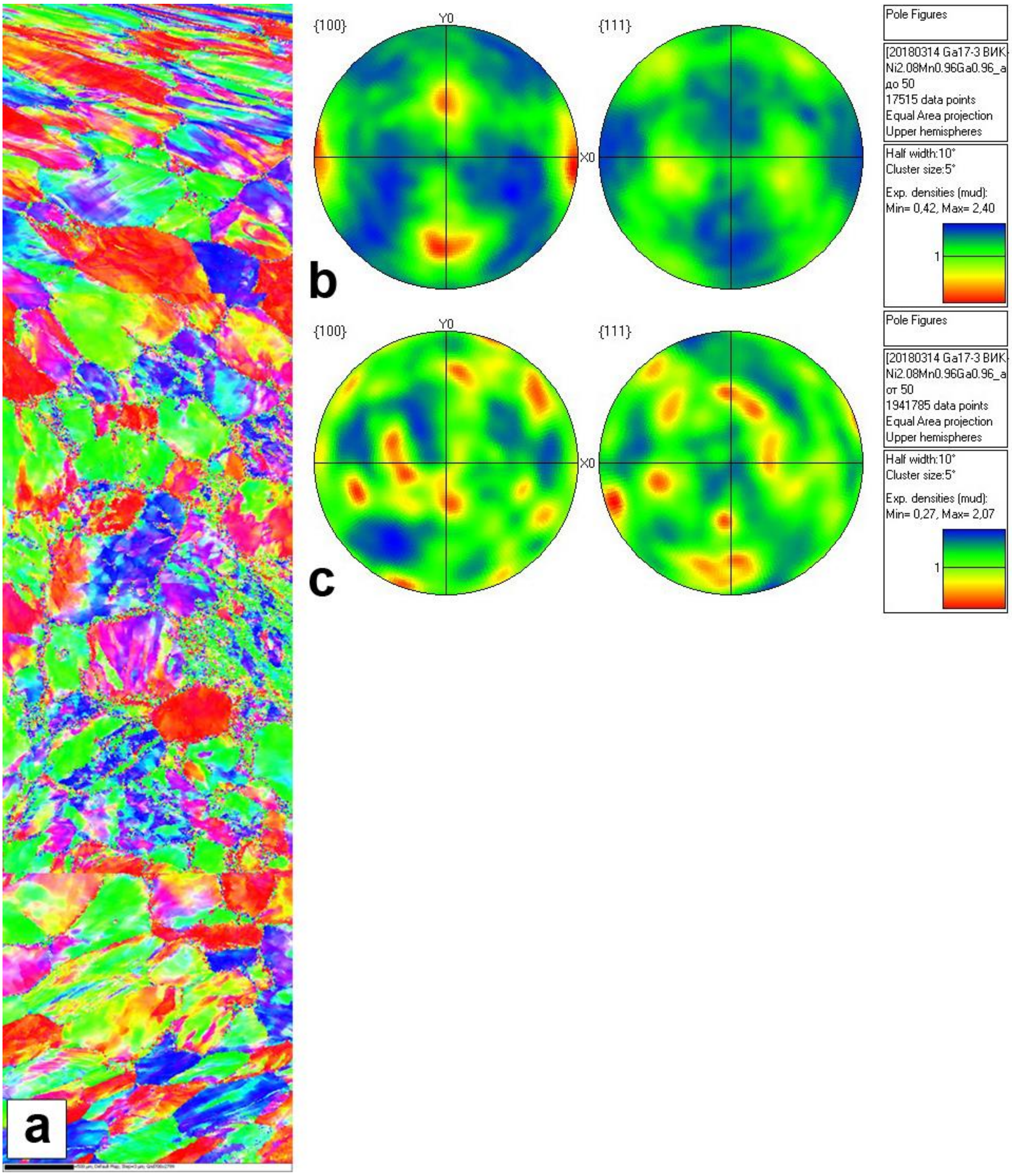

Fig. 6. EBDS analysis of Ga17/3 alloy after MIF and extrusion: a - EBDS image in IPF mode, b - Pole Figures for small grains, c - Pole Figures for big grains

The metallographic texture is absent. There is a slight stretching of the grains along the treatment axis only in the edge areas. The large grains are equiaxial in the central part of the workpiece. The structure has a slight heterogeneity over the cross section of the workpiece. The Pole Figures of {100} and {111} planes were constructed throughout the orientation map for the analysis of the crystallographic texture. As in case of the forged state, the data for large grains and for the fine-grained structure were separated by the software. The Pole Figures for large grains are shown in Fig. 6 b, for the fine-grained structure in Fig. 6 c. Despite some localized maxima for the family of {100} planes located on the stereographic plane the texture is not evident because the statistical data for such a number of large grains is not enough. The localized maxima of one crystallographic direction are observed for the fine-grained structure. Thus, the fine-grained structure has a single-component crystallographic texture.

4. Conclusions

After the thermo-mechanical treatment by multiple isothermal forging and the complex treatment by forging and subsequent extrusion the studied Heusler alloys show the anisotropy of the properties at the martensitic transformation. An abrupt length changing of about 0.05-0.13% was observed at the phase transformation in the samples cut along the deformation axes. The anisotropy of the thermal expansion is resulted due to both the treatment texture and the residual tensile stresses in the treated samples. The jump at the phase transformation in treatment states is lower than in as-cast state, in which it may reach 0.35%. It is suggested that the low jump occurs due to the low texture after the treatment and insufficient strain at both forging and extrusion. Therefore, in order to enhance the texture sharpness, it is necessary to increase the number of canting at the forging and the strain at the extrusion. The undoubted advantage of the proposed approach for Ni2MnGa alloys is forming of bimodal structure after the thermo-mechanical treatment by forging and forging with subsequent extrusion. In such structure, the original large grains of 100–200 μm are surrounded by the fine-grained structure. The stability of the functional properties of Heusler alloys with multiple cycles of the martensitic transformation should be higher due to the phase stresses relaxation and retardation of defects accumulation and microcracks initiation. Moreover, the additional treatment by extrusion provides volume fraction increase of the small grains. It enhances the relaxation ability of the alloy. Correspondingly, further increase of the strain should enhance both the anisotropy of properties and the cyclic strength of the alloy more by increasing the structure bimodality.

To sum up, the studies have shown a high potential of the thermo-mechanical treatment for obtaining the structurally modified Heusler alloy having a sufficient level of functional and service properties for the practical application.

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