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Magnetization Processes in Nanocrystalline Pr–(Fe,Co)–(Zr,Nb)–B Magnets

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In the present studies the magnetic interactions and magnetization reversal processes were analyzed based on the measurements of recoil curves. The investigated samples were the nanocrystalline 0.5 mm and 1 mm thick plates of the $Pr_9Fe_{50}Co_{13}Zr_1Nb_4B_{23}$ alloy, devitrified from the as-cast state at various temperatures from 923 K to 1033 K for 5 min. Phase structure analysis has shown similar phase constitution of the samples, however, the volume fraction of constituent phases and their crystallite sizes were different depending on the thickness of the samples annealed at particular temperature. Magnetic measurements have also shown the hard magnetic properties of the annealed specimens. However, their magnetic parameters were different for specimens of various thicknesses. In order to get better insight into the magnetization processes the δM plots and switching field distributions were analyzed for all investigated specimens.

topics: permanent magnets, rapid solidification, magnetization reversal

1. Introduction

Lot of scientific attention is payed to the metallic glasses with ferromagnetic properties due to their variety of applications in electromagnetic devices. Especially, Fe and Co based bulk metallic glasses can reveal both soft and hard magnetic properties [1–4]. Manufacturing of magnets through the devitrification annealing of bulk glassy precursors could be particularly useful method to obtain miniature magnets. Such products reveal very good corrosion resistance, avoiding further anticorrosion coating. However, use of such processing method requires rigorous technological window parameters concerning alloy compositiiton as well as devitrification annealing conditions. The objective of the present work was to determine the influence of the annealing temperature on the magnetic parameteres and on magnetization reversal of rapidly solidified plates of variuos thicknesses produced from the alloy of good glass forming ability [5].

2. Samples preparation and experimental methods

The 0.5 mm and 1 mm thick plates were produced by suction-casting technique from the ingots of the homogenized $Pr_9Fe_{50}Co_{13}Zr_1Nb_4B_{23}$ alloy. Subsequently the as-cast samples were heat treated at various annealing temperatures (T_a) from 953 K to 1033 K for 5 min. The annealing temperature of 953 K was chosen based on differential scanning calorimetry measurements carried out on fully amorphous ribbon of the $Pr_9Fe_{50}Co_{13}Zr_1Nb_4B_{23}$ alloy [5] and was close to the temperature of first crystallization peak. Both processing and annealing procedures were carried out under an Ar atmosphere. For all investigated samples the hysteresis loops were measured using LakeShore 7307 VSM magnetometer operating at room temperature in external magnetic fields up to 1600 kA/m. Furthermore, the recoil curves were measured as described in [6]. These measurements allowed to calculate the switching field distributions (SFD) and δM plots.

3. Results and discussion

The phase composition of the samples was analysed using the Rietveld refinement of X-ray diffraction (XRD) patterns. This analysis allowed not only to identify the phases but to calculate the crystallite sizes and weight fraction of constituent phases. It revealed the presence of the hard magnetic $Pr_2(Fe, Co)_{14}B$, paramagnetic $Pr_{1+x}Fe_4B_4$ and soft magnetic α-Fe crystalline phases in all annealed samples except for the 0.5 mm plates annealed at 953 and 983 K. For these samples the α-Fe was not detected. The weight fraction of α-Fe increased with T_a at the expense of the $Pr_2(Fe, Co)_{14}B$. The smallest crystallites of ≈ 10 nm were characteristic for α-Fe phase in every sample in which this phase was detected. The crystallites of $Pr_2(Fe,Co)_{14}B$ phase were much bigger with the mean diameters of ≈ 30–45 nm and increased with the annealing temperature. No remaining amorphous phase was detected after the heat treatment.

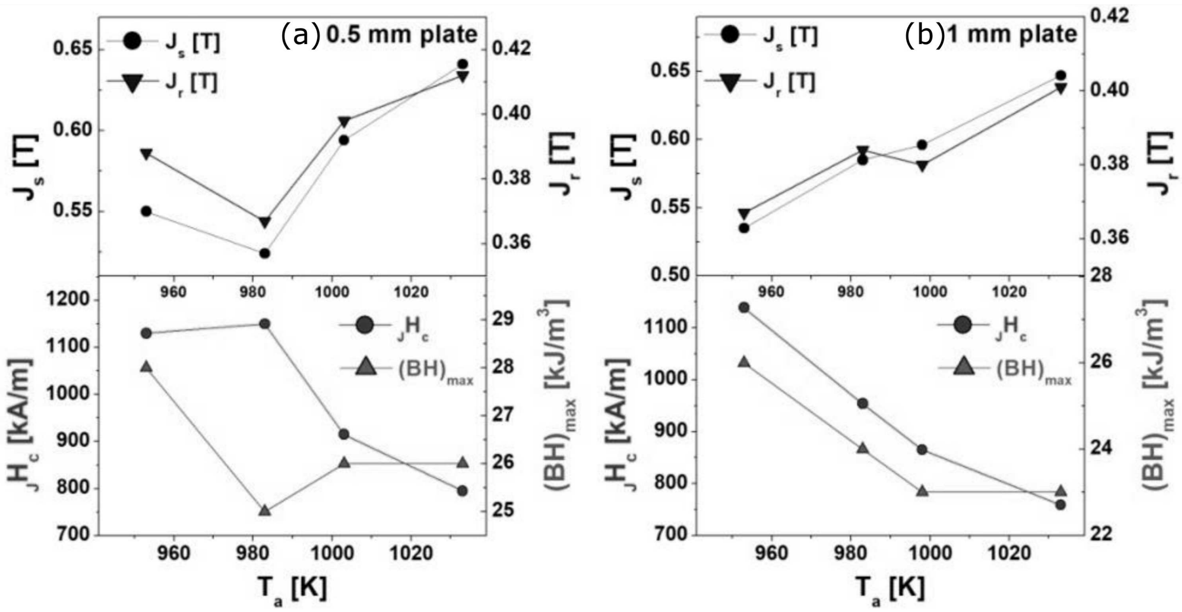

Fig. 1. The dependences of magnetic parameters (saturation polarization J_s, polarization remanence J_r, coercivity field ${}_JH_c$ and maximum magnetic energy product $(BH)_{\mathrm{max}}$) on the annealing temperature (T_a) for 0.5 mm (a) and 1 mm (b) thick plates.

The dependences of magnetic parameters on annealing temperature (Fig. 1) were constructed based on the values taken from the hysteresis loops. The increase in the annealing temperature above 983 K resulted in the rise of the saturation polarization (J_s) and the remanence (J_r) for both types of plates. The highest values of J_s and J_r were achieved for samples annealed at 1033 K and J_s was equal for both 0.5 mm and 1 mm thick plates reaching 0.64 T. This growth in J_s with T_a is related to the increase in the weight fraction of α-Fe phase. Things look very different in the case of coercivity changes (${}_JH_c$), where ${}_JH_c$ decreases significantly for samples annealed above 983 K. For the 0.5 mm plate, the ${}_JH_c$ reaches 1150 kA/m when annealed at 983 K and decreases to 795 kA/m (for $T_a = 1033$ K). This behaviour is mainly related to the growth of the crystallite sizes of the hard magnetic phase and slight decrease of its weight fraction with increasing T_a.

The series of recoil curves were measured for all samples at initially saturated and initially thermally demagnetized state. From these data the switching field distribution (SFD) curves were constructed based on the procedure described in [7]. The profile of SFD peak reflects the rate of magnetization reversal with an increase in applied recoil field and is related to the microstructural homogeneity of the sample [7]. For example if the microstructure of the sample is homogeneous (only one magnetic phase with narrow grain size distribution) one can expect a swift reversal of magnetization in narrow range of reversed magnetic field. The SFD curve for such case has the form of a narrow peak [8]. If the SFD curve broadening occurs, the material has the heterogeneous microstructure and/or the dipolar and magnetic interactions between grains play significant role in magnetization reversal [9]. Furthermore, in the case of highly heterogeneous microstructure the characteristic tail at SFD curves in low or high fields is present [10].

The SFD curves calculated for all samples are presented in Fig. 2. It was possible to fit each of obtained curve with only one Gaussian function. The values of the fields corresponding to the maxima of the Gaussian distributions (H_{max}) and their half widths (W_H) are collected in Table I. All curves are characterized by significant values of W_H which indicates that the magnetization reversal does not proceed as an abrupt and swift process but in a wide range of reverse fields. The maxima of the SFD are slightly higher than the values of coercivities of the samples. For the 0.5 mm plate annealed at 953 K and 985 K the H_{max} of SFD curves take higher values, which means that the most of the sample's magnetization reverse in higher fields. For higher T_a the SFD maxima are shifted towards the lower fields. This behaviour could be related to the presence of the α-Fe phase in the samples annealed at higher T_a.

The interactions between grains can be studied base on the δM plots, which represent deviation in the system from the Stoner–Wohlfarth model of magnetization reversal process for non-interacting particles. According to the interpretation given by Kelly et al. [11], the positive values of the δM corresponds to exchange interactions between grains, while negative are related to dipolar interactions. In Fig. 3 the δM plots are presented for investigated specimens.

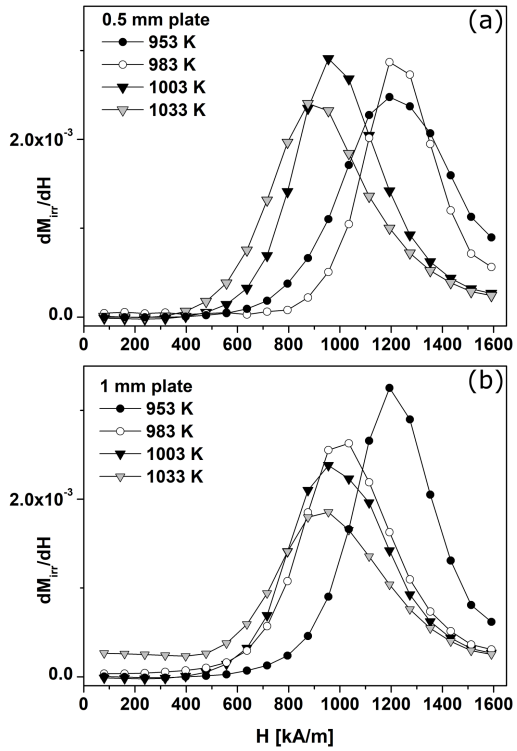

Fig. 2. The SFD for the nanocrystalline 0.5 mm (a) and 1 mm (b) thick plates of the $Pr_9Fe_{50}Co_{13}Zr_1Nb_4B_{23}$ alloy annealed at various temperatures.

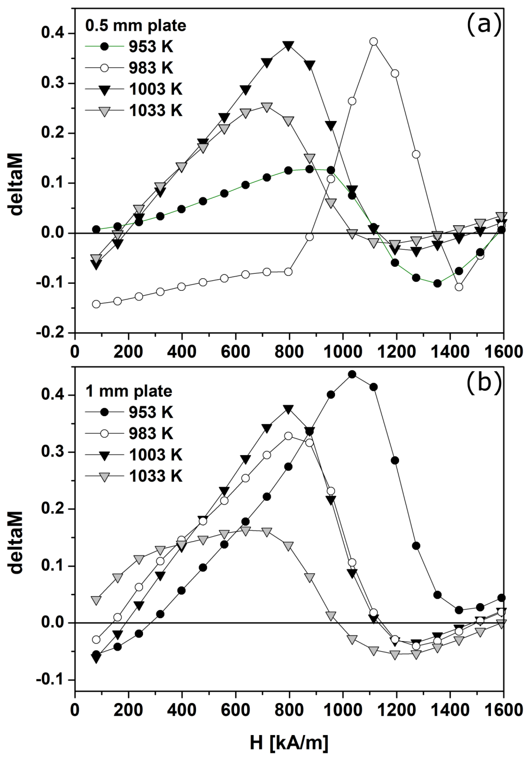

Fig. 3. The δM plots for the nanocrystalline 0.5 mm (a) and 1 mm (b) thick plate of the $Pr_9Fe_{50}Co_{13}Zr_1Nb_4B_{23}$ alloy annealed at various temperatures.

TABLE I

The parameters of the Gaussian fitting of SFD curves: $H_{\max}$ — the value of magnetic field at which the maximum in $\mathrm{d}M_{\mathrm{irr}}/\mathrm{d}H$ occurs, W_H — the half width of Gaussian curve.

T_a [K]	$H_{\max}$ [kA/m]	W_H [kA/m]
	0.5 mm plate	
953	1193	410
983	1190	385
1003	954	369
1033	875	362
	1 mm plate	
953	1193	286
983	1034	338
1003	964	375
1033	954	385

For all samples, the parameter takes both positive and negative values. The magnetic field at which δM reaches maximum values is the highest for the specimens having the highest coercivities (for 0.5 mm plate annealed at 983 K and for 1 mm plate — 953 K). Additionally, in case of 1 mm plate annealed at 953 K the strongest interactions, related to positive δM values are present in almost whole range of magnetic fields. For the samples annealed at temperatures higher than 983 K, the δM parameter takes negative values in higher fields. For these samples the magnetostatic interactions dominate over the intergranular exchange coupling, which is related to the larger weight fraction of the soft magnetic phase (α-Fe), than in the samples annealed at lower temperatures.

4. Conclusions

It was shown that for the rapidly solidified plates of the $Pr_9Fe_{50}Co_{13}Zr_1Nb_4B_{23}$ alloy the increase of the annealing temperature resulted in the precipitation of the nanocrystals of the α-Fe phase. However, in the 0.5 mm thick plate annealed at 953 K it is possible that some remnant amorphous phase is present, which results in relatively high J_s. On the other hand, in 0.5 mm thick plate the growth of α-Fe occurs at T_a higher than 983 K. It was shown that nucleation of α-Fe and the growth of $Pr_2(Fe, Co)_{14}B$ grains leads to the shift of SFD maxima towards the lower H. Furthermore, a presence of α-Fe is the source of dipolar interactions between grains, that was indicated by the shapes of δM plots.

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