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Effect of magnetoelastic anisotropy on properties of Finemet-type microwires

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ABSTRACT

Magnetic properties and DSC peak near Curie temperature, $T_{\rm C}$, of amorphous and nanocrystalline microwires with different ratios $\rho = d/D$ were studied. The investigated compositions were close to Finemet-type: Fe_{70.8}Cu₁Nb_{3.1}Si_{14.5}B_{10.6}, Fe_{71.8}Cu₁Nb_{3.1}Si₁₅B_{9.1} and Fe_{73.8}Cu₁Nb_{3.1}Si₁₃B_{9.1}.

The effects of magnetoelastic energy, stored during the Finemet-type microwires fabrication, on hysteresis loops, T_c and heat capacity of Finemet-type microwires were investigated. Hysteresis loops of all as-prepared microwires showed rectangular shape, typical for Fe-rich microwires. As expected, coercivity, H_c , increases with the decrease of the ratio ρ . On the other hand, the change of heat capacity at T_c , ΔC_p , exhibits linear increase with the ratio ρ . This relationship holds for microwires in the initial state as well as after annealing. Glass removal results in considerable change of both H_c and ΔC_p , which reveals the effect of internal stresses. Structural relaxation of microwires results in a shift of T_c calorimetric peak of amorphous phase to higher temperatures, while crystallization leads to peak disappearance. This effect was attributed to the dependency of T_c calorimetric peak on the value of magnetostriction of magnetic phase, which declines to zero with Finemet alloys crystallization.

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1. Introduction

Amorphous and nanocrystalline magnetically soft glass coated microwires (typically of $10-30 \,\mu$ m in diameter) attracted growing attention within the last few years owing to their outstanding hysteretic magnetic properties (magnetic bistability, enhanced magnetic softness, fast domain wall propagation) and GMI effect suitable for technological applications [1,2]. Particularly, recent studies have demonstrated that considerable improvement of soft magnetic properties and GMI effect of glass coated microwires is possible selecting appropriate chemical composition of metallic nucleus and adequate annealing conditions [1]. In some cases, nanocrystallization allows achieving good magnetic softness and enhanced GMI effect in ferromagnetic microwires [3,4]. Consequently, it is quite important to study the effect of magnetoelastic anisotropy on magnetic properties and Curie temperature of Finemet-type glass-coated microwires.

It is worth mentioning that the simultaneous solidification of composite microwire consisting of ferromagnetic nucleus surrounded by glass coating introduces considerable internal stresses inside the ferromagnetic nucleus during simultaneous solidification of composite microwire due to the difference in the thermal expansion coefficients of the glass and the metal [1,5–7]. Generally magnetic properties and overall shape of hysteresis loops of amorphous microwires depend on the composition of the metallic nucleus as well as on the composition and thickness of the glass coating. As discovered before in [1], shape of hysteresis loops changes from rectangular, typical for amorphous Fe-rich compositions, to inclined, typical for Co-rich compositions. Microwires with vanishing magnetostriction exhibit quite soft magnetic properties.

Such strong dependence of the hysteresis loops on these parameters should be attributed to the magnetoelastic energy given by:

$$K_{\rm me} \approx \frac{3}{2} \lambda_{\rm s} \sigma_{\rm i},$$
 (1)

where λ_s is the saturation magnetostriction and σ_i is the internal stress. The magnetostriction constant depends mostly on the chemical composition and is vanishing in amorphous Fe–Co based alloys with Co/Fe \approx 70/5 [1,8]. On the other hand, the estimated values of the internal stresses in these glass coated microwires arising from the difference in the thermal expansion coefficients of metallic nucleus and glass coating are of the order of 100–1000 MPa, depending strongly on the ρ -ratio ($\rho = d/D$ – ratio between the metallic core diameter, d, and total microwire diameter, D) [5–7]. The internal stresses increase with rising of the glass coating thickness (i.e. with ρ -ratio decreasing). Such large internal stresses give rise to a drastic change of the magnetoelastic energy, K_{me} , even for small changes of the glass-coating thickness at fixed metallic core diameter. Additionally, such a change of the ρ -ratio should be

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related to the change of the magnetostriction constant with applied stress [1]:

$$\lambda_{\rm s} = \left(\frac{\mu_{\rm o}M_{\rm s}}{3}\right) \left(\frac{{\rm d}H_{\rm k}}{{\rm d}\sigma}\right),\tag{2}$$

where $\mu_{o} M_{s}$ is the saturation magnetization.

It is worth mentioning, that residual stresses of glass-coated microwires arising during simultaneous solidification of metallic nucleus and glass coating, mostly have been estimated from the simulations of the process of simultaneous solidification of metallic nucleus inside the glass tube [5-7] and experimental determination of such residual stresses is rather complex. One of the experimental evidence of existence of such stresses is the dependence of hysteresis loops and particularly magnetic properties (coercivity, remanent magnetization) on ρ -ratio [1,9].

Consequently, tailoring of the magnetoelastic energy, K_{me} , is essentially important for achieving optimal magnetic properties of glass-coated microwires [1,10–12]

Accordingly, any method allowing estimation of internal stresses in glass-coated microwires is quite suitable for soft magnetic properties optimization.

Lately a few publications on utilisation of differential scanning calorimetry, DSC, method for the studies of the properties of amorphous alloys in vicinity of Curie temperature have been released [13,14]. This method allows determination of $T_{\rm C}$ and activation energy of relaxation and crystallization processes. Besides, DSC method allows precise determination of $T_{\rm C}$ (with reproducibility ±0.2 K).

In this paper we studied effect of magnetoelastic energy stored during the Finemet-type microwires fabrication on hysteresis loops, Curie temperature and heat capacity.

2. Experimental

Four Finemet-type compositions were investigated: $Fe_{70.8}Cu_1Nb_{3.1}Si_{14.5}B_{10.6}$ (A), $Fe_{71.8}Cu_1Nb_{3.1}Si_{15}B_{9.1}$ (B), $Fe_{73.8}Cu_1Nb_{3.1}Si_{13}B_{9.1}$ (C) and $Fe_{70.8}Cu_1Nb_{3.1}Si_{16}B_{9.1}$ (D). Glass-coated microwires with different metallic nucleus diameter, *d*, and total microwire diameter, *D*, were produced by modified Taylor–Ulitovsky method [1].

Hysteresis loops have been determined by flux metric method, as described elsewhere [1,3]. DSC measurements were performed using DSC 204 F1 Netzsch calorimeter in

DSC measurements were performed using DSC 204 F1 Netzsch calorimeter in Ar atmosphere at a heating rate of 10 K/min. In order to increase the sensibility of DSC measurements the mass of the samples was increased up to 30–50 mg (details see in [8]) that was possible due to special high-volume aluminum containers.

The Curie temperature, the peak area and the change of the specific heat near the Curie point were estimated from DSC curves using standard IT application.

X-ray diffraction (XRD) analysis was carried out on Rigaku ULTIMA-4 diffractometer in $\text{CuK}\alpha$ radiation.

3. Results

Hysteresis loops of as-prepared microwires showed rectangular shape, typical for Fe-rich amorphous microwires (Fig. 1) [1].

As expected, coercivity, H_c , of as-prepared microwires depends on ratio $\rho = d/D$ (Fig. 2).

Conventional (without magnetic field or applied stress) annealing at temperatures below the nanocsrystallization did not affected both coercivity value and overall shape of hysteresis loops, as previously observed for amorphous Fe-rich microwires [13].

On the other hand, glass removal leads to decrease in coercivity of microwires for as-prepared samples and after annealing.

DSC curve reflects the thermal properties of microwires and depends on many parameters of production and subsequent heat treatment [14]. Typical DSC curves of as-prepared Finemet-type microwires are shown in Fig. 3.

Magnetic transition at the temperature near 300 °C (T_C) and two peaks of crystallization (T_1 and T_2) can be seen. T_1 corresponds to the release of α -Fe nanocrystals, and T_2 – boride phase [15].



Fig. 1. Hysteresis loops of as-prepared $Fe_{73,8}Cu_1Nb_{3,1}Si_{13}B_{9,1}$ microwires with different ρ -ratios: (a) ρ = 0.87, (b) ρ = 0.38.

Curie temperature is very sensitive to the composition of the Finemet amorphous alloys: increase of Fe and B content results in the rise of T_c , while Cu and Nb reduce its value [16]. This is also true for the studied microwires: increase of Fe content from 70.8% (alloy A) to 73.8% (B) leads to T_c decrease from 310.4 °C to 299.3 °C.

Heat treatment of microwires leads to a change of the position and the shape of DSC peak in the vicinity of the Curie point (Fig. 4). The shape of T_C calorimetric peak was estimated as the difference of heat capacities ΔC_p at T_C and at 20 °C above this point.



Fig. 2. Coercivity dependence on ρ -ratio for as-prepared Fe_{73.8}Cu₁Nb_{3.1}Si₁₃B_{9.1} microwires.

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