Journal of Alloys and Compounds 674 (2016) 266-271

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom





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ALLOYS AND COMPOUNDS

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A R T I C L E I N F O

Article history: Received 12 January 2016 Received in revised form 4 March 2016 Accepted 7 March 2016 Available online 10 March 2016

Keywords:

Magnetic glass-coated microwires Giant magnetoresistance effect Taylor- Ulitvosky technique Heat treatment Magnetoresistance Granular structure Internal stresses Taylor-Ulitovsky method Glass-coated microwire Annealing

ABSTRACT

We studied the effect of the annealing on the structure, transport properties and the magnetoresistance of $Cu_{95}Co_{95}$ glass-coated microwires prepared by Taylor-Ulitovsky technique. We observed a significant enhancement of the magnetoresistance, MR, effect in the samples annealed at 400 °C. On the other hand low temperature annealing (150–200 °C) allowed stress relaxation and elimination of the texture observed in as-prepared samples, although only slightly affects the MR effect. Annealing considerably affects the temperature dependence of resistivity. We observed resistivity minimum in both as-prepared and annealed samples associated with the Kondo effect. This minimum persists even under magnetic field in as-prepared samples. In annealed sample minimum disappears under applied magnetic field. Observed enhancement of the MR effect therefore must be attributed to the structural changes of the studied samples.

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1. Introduction

Granular materials consisting of nano-sized ferromagnetic grains embedded into a nonmagnetic metallic matrix attracted continuous attention mostly owing to giant magnetoresistance (GMR) effect firstly reported in thin films [1] and slightly later reported for various granular materials [2–4]. Usually granular materials can be prepared from the immiscible elements, typically ferromagnetic elements such as Co or Fe, embedded into conductive metallic matrix (typically Cu, Ag, Au or Pt). The advantage of these granular materials is that they can be prepared using different rapid quenching [5–7] or mechanical alloying [8] techniques allowing fast and massive preparation of the samples.

The origin of the GMR effect in granular alloys has been

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explained considering the same nature as in the case of multilayered thin films, i.e. the spin-dependent scattering of conduction electrons within the magnetic entities as well as at the interfaces between the magnetic and nonmagnetic regions [1–8]. On the other hand there are still open questions as regarding the structure responsible for GMR effect in granular materials. Thus Co particles embedded in Cu matrix [5], small Co clusters within a Cu matrix [9], homogeneous spinodal decomposition characterized by long parallel Co-excess stripes [4] are proposed by various authors.

Rapid quenching techniques, allowing fabrication of metastable metallic alloys using the melt quenching, are quite common for preparation of the granular materials [4-6,9]. Usually, after appropriate recrystallization of the obtained alloys using annealing a more stable structures consisting of nano-sized inclusions into conductive metallic matrix can be obtained. Aforementioned precipitation of the small grains into conductive matrix is related to the phase diagram of the granular materials. Usually quite low solubility of the ferromagnetic metals in the metallic matrix metals at

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room temperature takes place in phase diagrams of these metals. The source of the metastability is that in most of cases (i.e. Co–Cu) the solubility of ferromagnetic metal increases with temperature. Consequently, at room temperature a supersaturated solid solution can be obtained after the rapid quenching from high temperature. The appropriate annealing of such material may lead to the small particles precipitation form the matrix and formation of the materials consisting of fine grains of ferromagnetic elements embedded into the metallic non-magnetic matrix [5,7].

The most traditional well established rapid quenching technique allowing preparation of ribbons with either amorphous of metastable structure is planar melt spinning [4-6,10]. Quite different rapid quenching technique allowing fabrication of composite (glass-coated microwires) has been widely used for preparation of amorphous and nanocrystalline microwires [11,12]. This technique allows fabrication of long (up to few km) and continuous (up to 10 km continuous) microwires coated by glass and therefore recognized as quite promising from the view point of applications. The presence of the glass-coating with rather different thermal and mechanical properties can change considerable quenching conditions during the rapid quenching process. For instance, huge difference in thermal expansion coefficients of metallic nucleus and glass-coating induce strong internal stresses inside the metallic nucleus that can be tailored varying the glass-coating thickness and metallic nucleus diameter [11,13].

Consequently one can expect that the structure of the microwires with granular structure as well as GMR behaviour can be different and must be affected by the preparation parameters (quenching rate, internal stresses, annealing conditions ...).

Recently we already reported on the influence of the internal stresses on crystalline structure, crystallization temperature and magnetic properties of microwires with nanocrystalline structure obtained after the recrystallization from the amorphous precursor [14–16]. Additionally we already reported on preparation of Co-Cu glass-coated microwires using Taylor-Ulitovsky exhibiting considerable GMR effect (below 10%) [17–19].

It is well-known that the structure of granular alloys can be considerably changed and the GMR effect can be significantly enhanced using adequate thermal treatment [2-6,19]. Moreover recently we observed electrical resistivity minimum in Co₅Cu₉₅ glass-coated microwires [17].

Therefore in this paper we present our last experimental results on the influence of annealing on magnetic and transport properties of Co_5Cu_{95} glass-coated microwires.

2. Materials and methods

 Co_5Cu_{95} glass-coated microwires (total diameters, D $\approx 16.9 \,\mu$ m) consisted of metallic nucleus (with diameters, d $\approx 13.3 \,\mu$ m) covered by outer glass shell (Durand glass) have been prepared by the Taylor-Ulitovsky technique [18].

X-ray diffraction (XRD) measurements were carried out by means of a D8-Advance (BRUKER) diffractometer provided with automatic divergence and receiving slits as well as graphite monochromator. CuK α ($\lambda = 1.54$ Å) radiation was used in all the patterns. Magnetic and magneto-transport properties have been measured at various temperatures within 5–300 K using suitable options of the Quantum Design magnetometer (PPMS). Magnetoresistance has been defined as:

$$\Delta R/R(\%) = (R(H) - R(0)) \times 100/R(0)$$
(1)

Whenever the other was not stated the MR was measured for the field directed along the axis of the microwire, parallel to the current. Samples have been annealed in conventional furnace at various annealing temperatures, T_{ann} , (150–400 °C) varying the annealing time, t_{ann} .

MR in all samples were tested under electric current I = 10 μ A.

3. Experimental results and discussion

After annealing at 400 °C generally we observed considerable enhancement of the MR effect. For example we observed increasing of maximum $\Delta R/R$ from 1 to 13% after annealing at 400 °C for 10 h (Fig. 1). On the other hand annealing at lower temperature (T_{ann} = 150° C) did not affect the MR effect: $\Delta R/R(H)$ curves of Co₅Cu₉₅ sample after annealing at 150 °C: for 10 h is almost the same as in as-prepared samples (Fig. 1).

Consequently we systematically studied the influence of annealing time on magnetoresistance, MR, of Co_5Cu_{95} microwires annealed at 400 °C. The evolution of the maximum MR ratio on annealing time measured is shown in Fig. 2. Considerable enhancement of MR ratio after annealing at 400 °C (from 1% up to 17%) can be appreciated.

Low-temperature annealing has been performed in order to relax the internal stresses. From the previous knowledge on stress relaxation in glass-coated microwires we can assume that the relaxation processes run starting from 100 °C [20]. From Fig. 1 we can assume that the stress relaxation is not the key factor that affects the MR increasing (see Figs. 1 and 2) of studied samples.

Recently we reported that as-prepared Co_5Cu_{95} microwires exhibit a well pronounced minimum in temperature dependence of resistance at about 40 K, attributed by us to Kondo-like behaviour [17].

Consequently we measured temperature dependence of resistivity, R(T), in Cu₉₅Co₅ sample annealed at 400 °C. As can be seen from Fig. 3, the temperature of resistivity minimum, T_m, decreases after annealing (from 47 to 22 K). Additionally, the temperature coefficient of resistivity of annealed samples is much higher than of as-prepared Cu₉₅Co₅ sample.

These considerable differences must be associated with structural changes after annealing. Using XRD technique we observed the preferred grain orientation (texture) probably related to the fabrication method of microwires.

We observed presence of two phases, fcc α -Co and fcc Cu phase. Annealing affects the XRD of Cu₉₅Co₅ sample (Fig. 4a and b). After annealing of the sample Cu₉₅Co₅ we observed decreasing of the preferred orientation. The first peak is appreciably increased



Fig. 1. Δ R/R(H) dependences measured in as-prepared, annealed at 150 °C for 10 h and at 400 C for 15 h Co₅Cu₉₅ microwires measured at 5 K.

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