ARTICLE IN PRESS

Journal of Alloys and Compounds xxx (2014) xxx–xxx

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

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

As-cast nanocrystalline glass-coated microwires

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ARTICLE INFO

Article history:
Available online xxxx

Keywords:
Nanocrystalline materials
Rapid solidification
Magnetization processes
Magneto-impedance
Microstructure

ABSTRACT

As-cast nanocrystalline $Fe_{74.5-x}Si_9B_{13.5}Cu_xNb_3$ ($x = 1$ and 2.5 at.%) glass-coated microwires have been prepared by drawing from the melt in a single step process. The cooling conditions and Cu content were modified in order to facilitate the growth of α-(Fe,Si) nanograins directly from the wire formation process. By increasing the cooling distance, the microstructure evolves from a fully amorphous one at low cooling distances to a nanocrystalline central area and an amorphous shell towards the surface at higher cooling distances. Changes of the composition and cooling conditions allow an extensive tailoring of the magnetic properties and structure of the as-cast glass-coated microwires.

1. Introduction

Nanocrystalline materials have remarkable soft magnetic properties, which make them excellent candidates for various applications and devices such as sensors and actuators, inductors, transformer cores [1–4]. The first nanocrystalline alloy with the nominal composition $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ was proposed in 1988 by Yoshizawa et al. [5] and patented under the trade name FINEMET™. The classical fabrication of nanocrystalline materials entails the preparation of the material in amorphous state by direct quenching of the molten alloy (as ribbons, wires, glass-coated wires or bulk shapes – plates, bars, discs, etc.) followed by the subsequent annealing of the amorphous material at temperatures between 773 K and 873 K in order to induce the primary crystallization of α-(Fe,Si) nanograins [6,7], with typical grain sizes between 8 and 20 nm [5–7], randomly oriented and uniformly distributed in the Fe–Nb–B residual amorphous matrix. Because of the composition and very special microstructure, FINEMET-type materials are among the magnetically softest ones, with very high value of the initial relative permeability ($\sim 10^5$), very low coercivity (<1 A/m) and high saturation polarization (1.2–1.3 T) [6–8]. Given that the fabrication of the nanocrystalline materials implies more steps, new methods to produce them faster and with lower costs have been investigated. Even if the possibility to obtain nanocrystalline FINEMET-type alloys by controlling the quenching rate was mentioned since 1989 by Sawa and Okamura [8], the number of publications on this subject is limited and restricted to ribbon shaped materials obtained by single- or twin-roller melt-spinning technique [7–11].

Here, we report results on the preparation and characterization of as-quenched $Fe_{74.5-x}Si_9B_{13.5}Cu_xNb_3$ ($x = 1$ and 2.5 at.%) nanocrystalline glass-coated microwires obtained by direct drawing from the melt. The aim of this work is to present the effect of the cooling conditions on the crystalline structure and magnetic properties of FINEMET-type glass-coated microwires.

2. Experiment

$Fe_{74.5-x}Si_9B_{13.5}Cu_xNb_3$. ($x = 1$ and 2.5 at.%) glass-coated microwires, with the diameter of the metallic core of 19 μm and glass thickness of 15 μm have been prepared by drawing from the melt [12], using the distance between the inductor and cooling liquid (cooling distance, *D*) as a parameter. Fig. 1 shows a schematic of the equipment used to produce as-cast nanocrystalline glass-coated microwires. The cooling distance was determined each time accurately using a micrometer scale and was gradually increased, between 10 and 80 mm, in order to allow the partial crystallization of the alloy by slow cooling in air, followed by a fast cooling in water to "freeze" the material and to stop the growth of the crystalline grains. The copper content was increased within the alloy content in order to increase the number of nucleation centers [7] and to facilitate nanocrystallization in the wire preparation process. The drawing speed was 70 m/min for all wires.

All samples were characterized magnetically by using hysteresis loop and high frequency magneto-impedance (MI) measurements. The microstructure was investigated by X-ray diffraction (XRD) and ultra-high resolution transmission electron microscopy (UHR-TEM), and the thermal stability was studied by differential scanning calorimetry (DSC).

3. Results and discussion

Previous studies regarding magnetic behavior of the nanocrystalline glass-coated microwires obtained by annealing from amorphous precursors [13] have associated nanocrystalline state with a coercivity decrease at about half of the value of the amorphous state. For this reason, at the beginning, we used the coercivity as an indicator of the nanocrystalline state formation.

Fig. 2 shows the axial hysteresis loop of the as-quenched $Fe_{74.5-x}Si_{13.5}B_9Cu_xNb_3$ microwires ($x = 1$ and 2.5 at.%) obtained at different cooling distances, *D*.

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0925-8388/$ - see front matter © 2014 Published by Elsevier B.V.
http://dx.doi.org/10.1016/j.jallcom.2013.12.165

One observes that the magnetic properties are strongly influenced by the cooling distance for both copper concentrations. For a copper concentration of 1 at.%, the coercivity does not show a significant variation by increasing D to 40–45 mm. However, the coercivity increases abruptly for distances over 50 mm due to the crystallization of the metallic core. For the samples with 2.5 at.% Cu the hysteresis measurements show a decrease in coercivity when the cooling distance (D) increases – from 133 A/m at D = 10 mm down to a minimum of 74 A/m at D = 35 mm – followed by an increase for larger cooling distances (Fig. 3). This behavior indicates that the samples with 2.5 at.% Cu content obtained for a cooling distance of 35 mm exhibits a nanocrystalline structure, as it will be confirmed by the structural investigations discussed below.

Due to the stronger dependence of coercivity on cooling distance for the samples with 2.5 at.% copper, we focused the following studies on this alloy.

To study the influence of the cooling distance to the surface magnetic properties we performed magneto-impedance (MI) measurements. The MI ratio was calculated relative to zero field – $\Delta Z_{(H=0)}=[Z-Z_{(H=0)}]/Z_{(H=0)}{}^{*}100$ – and to the maximum field (5.5 kA/m) – $\Delta Z_{(H=\max)}=[Z-Z_{(H=\max)}]/Z_{(H=\max)}{}^{*}100$.

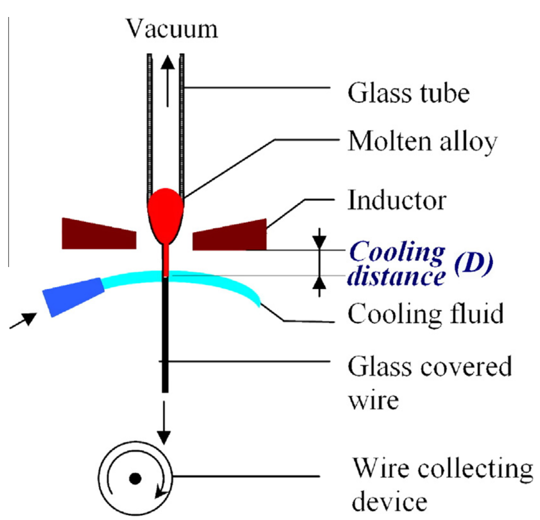

Fig. 1. Schematic of the glass-coated wire equipment.

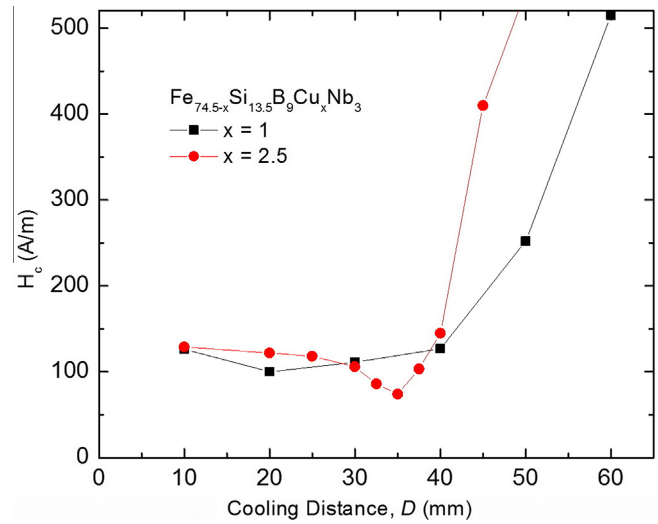

Fig. 3. Coercivity vs. cooling distance, D, for $Fe_{74.5-x}Si_{13.5}B_9Cu_xNb_3$ glass-coated wires having a 19 μm metallic core diameter and a 15 μm glass coating thickness.

The analysis of the magneto-impedance results (Fig. 4) shows relatively low MI ratio (<10%) for cooling distances between 10 and 40 mm and relatively high values of the MI ratio (up to 50%) for microwires obtained using a cooling distance between 45 and 60 mm. Further increase of the cooling distance leads to a decrease in the amplitude of the MI ratio. The amplitude of the MI ratio is related to the surface magnetic properties due to the skin effect of the high frequency current used for sample excitation. The thickness of the external shell of the wire which influences the MI response is given by the penetration depth and decreases with the increase of the frequency. The amplitude of the MI ratio shows that the surface of the material is magnetically softer, due to nanocrystallization, for a cooling distance between 45 and 60 mm (Fig. 4a).

The optimum nanocrystalline structure is characterized by a larger permeability as compared to the amorphous state, or to a structure with larger grains, which are obtained for smaller, respectively larger cooling distances (as shown in Fig. 5a for D = 10 mm and D = 80 mm, respectively). The main contribution to the permeability increase is the vanishing magnetostriction in the nanocrystalline state, which significantly reduces the effect of the residual stress.

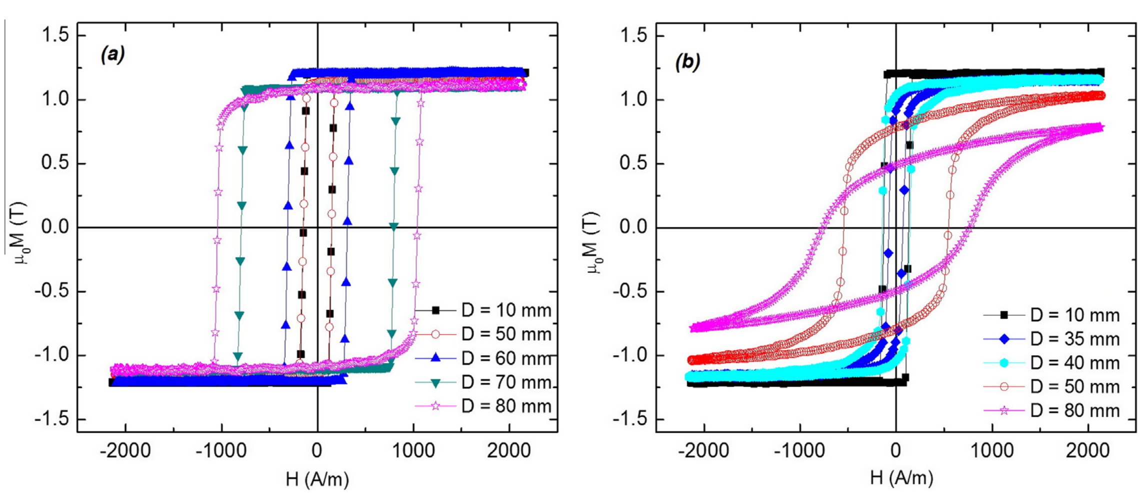

Fig. 2. Hysteresis loops of the $Fe_{74.5-x}Si_{13.5}B_9Cu_xNb_3$ glass-coated wires with the diameter of the metallic core of 19 μm and the glass coating thickness of 15 μm, obtained at different cooling distances (D): (a) x = 1 at.%; (b) x = 2.5 at.%.

Please cite this article in press as: S. Corodeanu et al., J. Alloys Comp. (2014), http://dx.doi.org/10.1016/j.jallcom.2013.12.165