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Structure and hyperfine interactions in Al-doped FINEMET

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Abstract

Mössbauer spectrometry, TEM and VSM were used to investigate microstructure and properties of amorphous and nanocrystalline $Fe_{73.5-x}Al_xNb_3Cu_1Si_{13.5}B_9$ ($0 \leqslant x \leqslant 7$) alloys. It was found that aluminum substitution caused changes in many characteristics: Curie temperature of the crystalline phase as well as of the amorphous one, magnetization, mean magnetic moment, mean hyperfine field, isomer shift and crystallization temperature. Character of the reported changes points to heterogeneity of as-quenched alloys and also of crystalline grains in the annealed samples, particularly for higher Al concentration. This is confirmed by Mössbauer temperature measurements performed for the nanocrystalline alloy with maximal Al content. At $x = 5$ the ordering into DO_3 structure is observed.

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Keywords: Nanocrystalline alloy; Mössbauer spectrometry; Microstructure; Thermomagnetic properties

1. Introduction

The family of novel soft magnetic materials called FINEMET is known for very high effective permeability, high saturation magnetization, small magnetostriction and coercivity [1–6]. The excellent soft properties comparable to those of permalloys and Co-based amorphous alloys originate from specific structure, composed of crystalline grains with typical sizes of several nanometers enclosed by an amorphous matrix. Such an ultrafine structure is produced by a proper heat treatment of a melt-spun precursor; the most common is 1 h isothermal annealing at temperature between 500 and 600 °C. The alloy composition originally proposed by Yoshisawa et al. [7] to obtain the nanocrystalline material was $Fe_{73.5}Nb_3Cu_1Si_{13.5}B_9$. Since 1988, when FINEMET was discovered, various methods have been proposed in order to improve its magnetic and mechanical properties, e.g. modification of the heat treatment

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procedure (isochronal annealing [8], two-step annealing [9], flash annealing [10], nitriding [11]) or changes of the alloy composition [12–16]. In the last-mentioned case, good results were achieved when replacing a certain number of iron atoms by aluminum. That is why the Al-doped FINEMET alloys are the objects of intensive studies, reported in many papers [17–25]. It has been found that some magnetic characteristics, for instance, saturation magnetization M_s [19,20,23], coercivity H_c [19,22], saturation magnetostriction λ_s [21], Curie temperature [23,25], magnetic moment at 0 K [27], spin-wave stiffness constant D [26–28] as well as crystallization temperature T_x [23] and electrical resistivity [21] are changed to a more or less extent as an effect of aluminum substitution.

In this work results of some structural and magnetic studies are reviewed, but first of all Mössbauer investigations of these materials are reported.

2. Experimental methods

Series of samples of composition $Fe_{73.5-x}Al_x Nb_3Cu_1Si_{13.5}B_9$, $x = 0, 1, 2, 3, 5, 7$ (called below Al0, Al1, Al2, Al3, Al5, Al7, respectively) were prepared by the rapid quenching method, in a form of 30-μm-thick and 5-mm-wide ribbons. To generate a creation of crystallites, isothermal heat treatments were performed at 490, 550 and 650 °C for 1 h in a vacuum furnace.

The microstructure of samples was studied by transmission electron microscopy (TEM) [21]. To investigate magnetic properties, vibrating sample magnetometer (VSM) was employed [23]. Thermomagnetic curves were taken in an external field of 0.5 T, 0.24 T, 10 mT and 2 mT at a heating rate of 10 °C/min from room temperature up to 800 °C. The low-temperature magnetization (over the range from liquid helium temperature to room temperature) was measured in an external magnetic field of 1 T applied in the plane of the ribbon.

Mössbauer investigations were carried out by using a transmission spectrometer arranged in vertical geometry, $^{57}Co(Rh)$ source of gamma radiation and a drive system working in a constant acceleration mode. The velocity scale (divided into 256 channels) was calibrated with a pure iron foil. For temperature measurements a special vacuum high-temperature furnace was employed. Each sample comprised of few pieces of the ribbon placed in a row, with an active surface of about 2 cm^2. To obtain a good resolution of Mössbauer spectra, they were collected during long periods (usually a few days), especially at temperature studies, because of reducing the γ-ray intensity by the furnace.

For samples Al1–Al7, both in as-quenched state and annealed at $T_a = 490$, 550 and 650 °C, Mössbauer measurements at room temperature were performed, which makes a complement of previous studies relating to the samples annealed at $T_a = 550$ °C [21,23]. Additionally, the sample Al7 subjected to annealing at $T_a = 550$ °C was investigated at elevated temperatures, $T = 100$, 150, 200, 250, 275, 300, 325 and 350 °C.

3. Structural and magnetic investigations

TEM micrographs reveal [21] that after annealing at 490 and 550 °C all alloys were nanocrystalline with typical grain size around 10–12 nm (Fig. 1), almost independently on aluminum content. Both thermoresistivity curves [23] and TEM micrographs indicate a structural transition to polycrystalline state in the temperature range 615–670 °C. Thorough examination of the electron diffraction patterns [21] (changes of the lattice constant and signs of inhomogeneity) suggests the presence of Al atoms inside the grains that compose a more or less ordered α-FeSiAl phase. As seen later, thermomagnetic investigations performed in an external magnetic field of 0.24 T also evidence the incorporation of aluminum into the grains.

Thermomagnetic curves taken for as-quenched samples (Fig. 2) show a monotonic decrease in magnetization in the temperature range up to the Curie temperature of the amorphous phase T_c^{am}. There are no magnetic transitions between T_c^{am} and the crystallization temperature T_x at which the magnetization increases rapidly due to the major precipitation of ferromagnetic BCC Fe(SiAl) nanograins from the amorphous matrix.