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Microstructure and soft magnetic properties of Finemet-type ribbons obtained by twin-roller melt-spinning

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ABSTRACT

Soft magnetic ribbons of Finemet-type ($Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$) alloys are synthesized by the twin-roller melt-spinning technique directly from the melt, at tangential wheel speeds of 15, 18, 19 and 20 m/s. The microstructure and the magnetic properties are characterized by X-ray diffraction (XRD), transmission electron microscopy (TEM), differential thermal analysis (DTA), thermo-gravimetric analysis (TGA) and hysteresis loops measurements. Samples cooled at 20 m/s are amorphous, while those quenched at lower wheel speeds are partially crystalline. All samples studied present saturation magnetization values (150–160 A m^2/kg) higher than the commercial Finemet alloys ($\sim$135 A m^2/kg), obtained by controlled crystallization of amorphous single-roller melt-spun alloys. Optimal soft magnetic properties – $\sigma_S=(154\pm8)$ A m^2/kg and $H_C=(6.9\pm0.9)$ A/m – are found in samples quenched at 19 m/s, consisting of size-distributed bcc Fe–Si nanograins ($\sim$18 nm in average) embedded in an amorphous residual matrix. A minority nanocrystalline magnetic phase ($\leq$10 nm) is also detected.

1. Introduction

Over the past several decades, nanocrystalline alloys have attracted considerable scientific and technological attention because of their remarkable soft magnetic properties making them excellent materials for many applications and devices, such as transformers, inductors, sensors, common mode chokes, actuators, etc [1,2]. In 1988, Yoshizawa et al. [3] produced the first nanocrystalline alloy with nominal composition $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$. The alloy was patented under the trade name FINEMET™. This new alloy presented a particular microstructure, characterized by randomly oriented, ultrafine α-FeSi grains with a bcc structure [4] and typical grain sizes between 10 and 15 nm, uniformly dispersed in a residual Fe–Nb–B amorphous matrix. These features constitute the basis for the excellent soft magnetic properties observed, as a high value of initial permeability ($\sim10^5$), a very low coercivity (less than 1 A/m) and a high saturation polarization, of about 1.2–1.3 T. [5].

Finemet alloys are generally synthesized by controlled crystallization from an amorphous precursor. The material is first prepared as a ribbon by the single-roller melt spinning technique and then submitted to an annealing treatment at temperatures between 773 K and 873 K, which induces primary crystallization of α-FeSi nanograins [3,5].

The particular nanostructure is ascribed to the combined effect of copper and niobium and their low solubility in the Fe–Si phase. Copper enhances the nucleation of these nanograins while niobium impedes grain coarsening and inhibits the formation of boride compounds [1,4]. In addition, the boron content mainly determines the volume fraction of the Fe–Si phase [6]. Both the crystalline fraction and the average grain size decrease with increase in the boron content.

The magnetic behaviour of these nanocrystalline alloys can be explained in terms of the random anisotropy model [7,8]. Since the ferromagnetic exchange correlation length is greater than the structural correlation length (Fe–Si grain sizes), the local magnetocrystalline anisotropies are averaged out over several grains by exchange interactions and thus notably reduced in magnitude. Hence, the ultra-soft magnetic properties are determined by this average anisotropy, which is several orders of magnitude lower than the magnetocrystalline anisotropy of the individual grains [9,10] and strongly depends on the average nanograin size D. The random anisotropy model predicts a D^6-dependence of coercivity at small grain sizes, below about 40–50 nm [5].

The possibility of obtaining the nanocrystalline state of Finemet-type alloys directly from the melt by controlling the

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quenching rate was early mentioned by Sawa and Okamura [11] in 1989; however, the number of publications on this kind of process is limited and experiments are always performed in asymmetric heat extraction geometries, such as in a single-roller device. In this sense, the twin-roller provides uniform quenching conditions.

In the present article, we report the synthesis of Finemet-type alloys ($Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$) by twin-roller melt-spinning. The microstructure and magnetic properties of the alloys obtained at different quenching rates are investigated and compared with those reported for the commercial Finemet^{TM} alloy. It is possible to obtain the nanostructure leading to optimal soft magnetic properties directly from the melt, without any subsequent heat treatment for further devitrification.

2. Experimental

Ribbons with nominal composition $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ were prepared by the twin-roller melt-spinning technique. The precursor alloys were prepared from high purity elements by arc melting in a water-cooled copper crucible, under argon atmosphere. The ingots were turned over and re-melted several times to ensure homogeneity; the total mass of each precursor ingot was 5 g. These ingots were melted again under argon atmosphere in a quartz crucible with 1 mm diameter nozzle by induction heating; the molten alloy (~1573 K) was then evacuated by a positive argon pressure onto 5 cm diameter copper wheels, coated with a thick chromium layer, at different tangential speeds between 15 and 20 m/s. The wheel speed is controlled within ± 0.5 m/s. The ribbons so obtained were 1.5–1.8 mm width and 44–60 μm thick, as shown in Table 1.

The structure properties for each quenching rate were analyzed by X-ray diffraction (XRD) and transmission electron microscopy (TEM). XRD profiles were measured in a Philips PW 3830 diffractometer, with Cu Kα radiation ($\lambda = 1.5418$ Å). A profile fitting was made to each maximum in the spectra to determine the apparent peak width, after correcting for instrumental broadening. XRD results were used to determine the phases present in the samples, their lattice parameters and the average grain size, using the Scherrer formula.

Specimens for TEM observation were prepared by electropolishing, in a Struers Tenupol double jet operating at 10 V, with an electrolyte of 5 vol% perchloric acid in acetic acid, at room temperature. Transmission electron microscopy observations and selected area diffraction patterns were performed in a Philips CM 200UT microscope, operating at 200 kV.

The thermal analysis was performed in a SDT Q600 DTA/TGA (TA Instruments) up to 1073 K, under argon stream, at a heating rate of 10 K/min. The crystallization process in the ribbons was monitored by the differential thermal analysis (DTA) and the Curie temperatures of the magnetic phases determined by thermo-gravimetric analysis (TGA) in the presence of a weak magnetic field (1.25 mT).

Room temperature saturation magnetization was measured in a vibrating sample magnetometer (VSM) Lakeshore 7300, with a maximum field of 1.5 T. The magnetic field was applied parallel to the ribbon length (~4 mm) leading to value of geometric demagnetizing factor of $N \sim 0.005$; then, in the present alloys demagnetizing effects cannot be neglected. On the other hand, coercivity was estimated from the hysteresis loops measured for ribbons with different lengths, from 30 to 12 mm, using a quasi-static fluxmetric method, by applying to the sample an axial field and collecting the induced signal in a secondary compensated pick-up coil.

Table 1
Mean thickness t and width w of the ribbons obtained at different tangential speeds between 15 and 20 m/s. Lattice constant a, silicon content x_α from Refs. [6,14,15] and mean grain size of the α-Fe(Si) nanograins, estimated from Scherrer equation (D) and from TEM observations (D_{TEM}), are also given.

Sample	V15	V18	V19	V20
t [μm]	60 ± 5	44 ± 6	46 ± 7	51 ± 6
w [mm]	1.5 ± 0.1	1.8 ± 0.3	1.6 ± 0.3	1.6 ± 0.2
a [Å]	2.843 ± 0.001	2.841 ± 0.001	2.845 ± 0.001	–
x_α [at%]	18.0 ± 0.5	19.0 ± 0.5	17.0 ± 0.5	–
D [nm]	28 ± 10	28 ± 10	27 ± 10	–
D_{TEM} [nm]	22 ± 6	19 ± 6	18 ± 7	–

3. Results and discussion

3.1. XRD analysis

XRD patterns of the as-quenched samples, synthesized at tangential wheel speeds of 15, 18, 19 and 20 m/s (samples V15, V18, V19 and V20, respectively) are shown in Fig. 1a. Sample V20

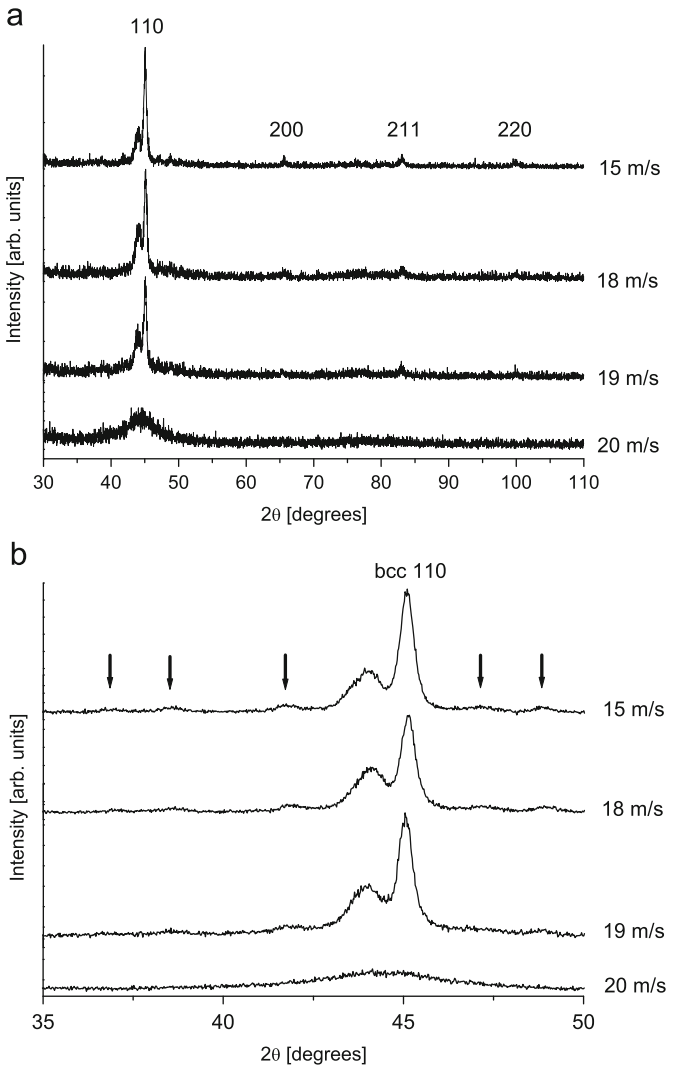

Fig. 1. (a) X-ray diffraction patterns of the as-quenched samples synthesized at different wheel speeds between 15 and 20 m/s. The principal diffraction lines of the bcc α-Fe(Si) phase are also shown. (b) Low step X-ray diffraction patterns of the samples studied. The principal diffraction peak (1 1 0) of the bcc α-FeSi phase and other peaks corresponding to an unknown phase are also presented.