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ANNEALING INFLUENCE ON THE STRUCTURAL AND MAGNETIC PROPERTIES OF $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ POWDERS

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Abstract. Results concerning structural and magnetic properties of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ powders prepared by mechanical milling of amorphous ribbons are presented. The obtained powder was isothermally annealed in vacuum at temperatures between 400 °C and 500 °C. The nanocrystalline powder with the lower coercive magnetic field is obtained after annealing 1 hour at 475 °C.

Key words: amorphous magnetic materials, magnetic powders, nanocrystalline magnetic materials, structural properties, magnetic properties.

1. INTRODUCTION

Magnetic amorphous and nanocrystalline materials, inclusively in the shape of powders are intensely studied nowadays, both from the experimental an theoretical point of views [1–9]. Among them, FeCuNbSiB nanocrystalline alloys (ribbons, wires, thin films, and powders) exhibits ultra soft magnetic properties (low coercivity, high saturation magnetization, high permeability, low frequency losses, near zero magnetostriction) [1–7]. The excellent soft magnetic properties are attributed to the microstructure consisting of α-Fe(Si) grains embedded in a residual amorphous matrix. FeCuNbSiB nanocrystalline ribbons and powders present a major interest in fabrication of soft magnetic cores due their good soft magnetic properties in high frequency ranges [10, 11]. Using nanocrystalline FeCuNbSiB powders leads to an improvement of the high frequency soft magnetic properties of powder cores as compared to the conventional metallic powder cores. The powders can be obtained by chemical synthesis, mechanical alloying, atomization or mechanical milling, self-combustion route [7, 12–15]. Mechanically milled nanocrystalline ribbons as a way to obtain nanocrystalline powders have been intensely studied, especially in what concerns the effect of milling time on the structural and magnetic properties. It has been found that increasing the milling time, the size of the particles decreases and the nanocrystalline phase is formed which leads to better packing and good soft magnetic properties [6, 11, 13, 14].

Since the smaller the particle size the higher is the packing factor we aim to produce small sized nanocrystalline powders by mechanical milling of amorphous ribbons followed by subsequent annealing at various temperatures. The $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ powders were isothermally annealed in a vacuum furnace (10^{-6} Torr) at temperatures between 400 °C to 500 °C and the structure and magnetic properties were studied.

2. EXPERIMENTAL

$Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ amorphous powders were obtained by mechanical milling of ribbons previously prepared by rapid quenching from the melt, in an inert argon atmosphere using a tangential speed of 25 m/s of the rotating wheel. The as-quenched ribbons have been vacuum annealed for 1 h at 300 ºC, temperature which is well below the nanocrystallization temperature (520 °C–550 °C), in order to prevent grains formation. Small pieces of the annealed ribbons were introduced into the stainless steel vial of a SPEX Sample Prep 8000 M MIXER/MILL and milled for 1 to 4 hours in argon atmosphere. The volume ratio of stainless steel balls to amorphous ribbons was 7:1.

Powders particle size, the shape and surface morphology were examined by scanning electron microscopy (SEM) using a Jeol JSM-6390A microscope with a tungsten filament.

The powder microstructure was investigated by X-ray diffraction (XRD) in Bragg-Brentano configuration using a Bruker D8 Advance diffractometer (λ = 1.54056 Å). XRD patterns of powders were analyzed using the DIFFRAC plus Eva software by comparison with the XRD patterns of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ amorphous ribbons. The average crystalline size was estimated using Scherrer's formula, after subtracting the precursor material halo from the diffractograms and carrying out instrumental corrections [16].

The coercive magnetic field, H_c, was measured using a vibrating sample magnetometer (VSM) in a maximum applied field of 0.8 T, at room temperature. The Curie temperature and the crystallization temperature were determined from the magnetization, M, dependence on the temperature (thermomagnetic curve) using VSM [1, 4, 5].

3. RESULTS AND DISCUSSION

Figure 1 (a, b, c) shows the SEM images for $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ powders obtained after 1 h and 4 h milling time (Fig. 1a and Fig. 1b, respectively) as well as the dependence of powder average size on the milling time (Fig. 1c). The shape of the powder particles has irregular morphology, the size distribution being wide for

the same milling time. The average powder size decreases from about 90 μm to 30 μm when the milling time increases from 1 h to 4 h as can be observed in Fig. 1c.

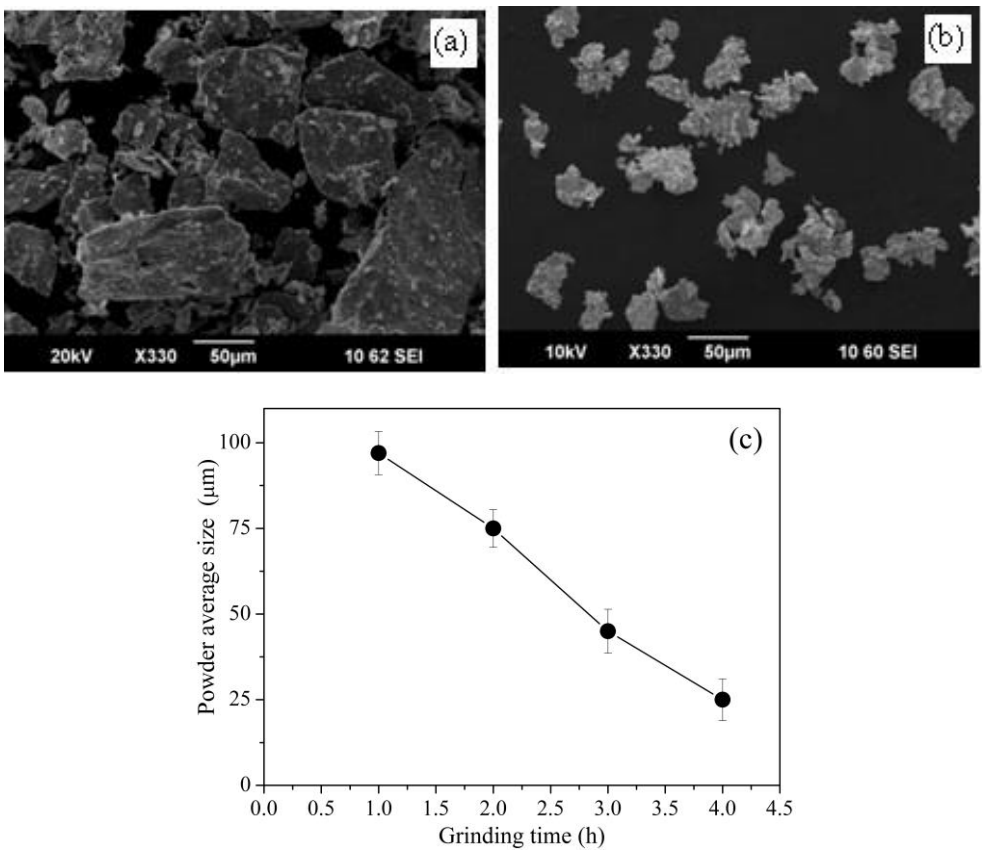

Fig. 1 – SEM images of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ powders after 1h (a), 4h (b) milling time and the dependence of powder average size on the milling time (c).

The results presented in the followings are focussing on the powders with powder average size of about 30 μm (obtained after 4 h milling time).

Figure 2 presents the normalized thermomagnetic curves for the annealed amorphous ribbon at 300°C (precursor material), unannealed powders obtained after milling the amorphous ribbons, and for isothermally annealed powders for 1 h at 450°C, 475°C, 485°C and 500°C, as well as the dependence of the Curie temperature of the amorphous phase, T_{ca} on the annealing temperature, T_{ann} (Fig. 2 inset). The thermomagnetic curve of the precursor material present a typical behaviour for a $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ ribbon with a Curie temperature of the amorphous phase of about 320°C and about 620°C Curie temperature of the nanocrystalline phase. The thermomagnetic curves for the unannealed and annealed

samples indicate an increase of the amount of nanocrystalline phase with the increasing the temperature. For both unannealed powders and powders annealed at temperatures lower than about 485°C, the magnetic moment decreases down to T_{ca}. For the unannealed powders T_{ca} is approximately 340°C, and the first crystallization temperature, T_{x1}, is about 500°C. For the annealed powders T_{ca} increases when the annealing temperature increases (as can be observed in the inset of Fig. 2) while T_{x1} remains approximately constant. In the case of the unannealed powders the magnetic moment is different from zero, between T_{ca} and T_{x1}, indicating the presence of small quantities of crystalline phase generated by the milling process [13, 16].

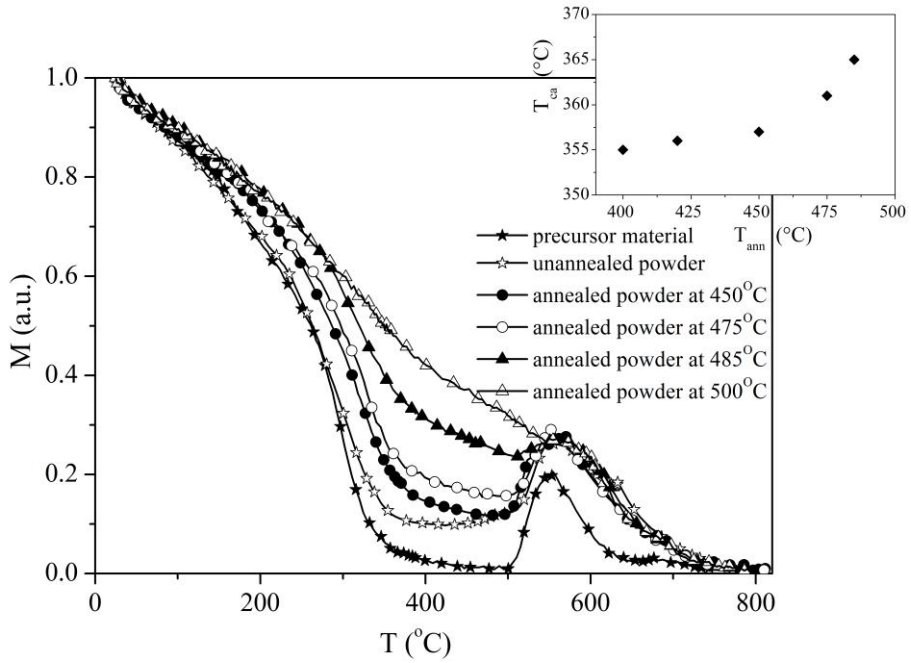

Fig. 2 – Normalized thermomagnetic curves for the precursor material, unannealed powders and isothermally annealed powders and T_{ca} dependence on T_{ann} (inset).

The evolution of the nanocrystalline phases indicated by the thermomagnetic curves was confirmed by XRD analysis. Figure 3 (a, b, c) presents the XRD patterns for the precursor material and milled powders (unannealed and isothermally annealed at 400°C, 450°C, 475°C, 485°C, 500°C) (Fig. 3a) and also the deconvolution curves for annealed powders at 475°C (Fig. 3b) and 500°C (Fig. 3c). For the unannealed powders a halo, typical for the amorphous state ($2\theta = 45°$), can be observed. The X-ray diffraction of the powders annealed at temperatures of up to about 450°C present a specific diffraction peak at $2\theta = 45°$ corresponding to the

(110) reflection of α-Fe(Si) phase, whose height increases with the increase of the T_{ann}.

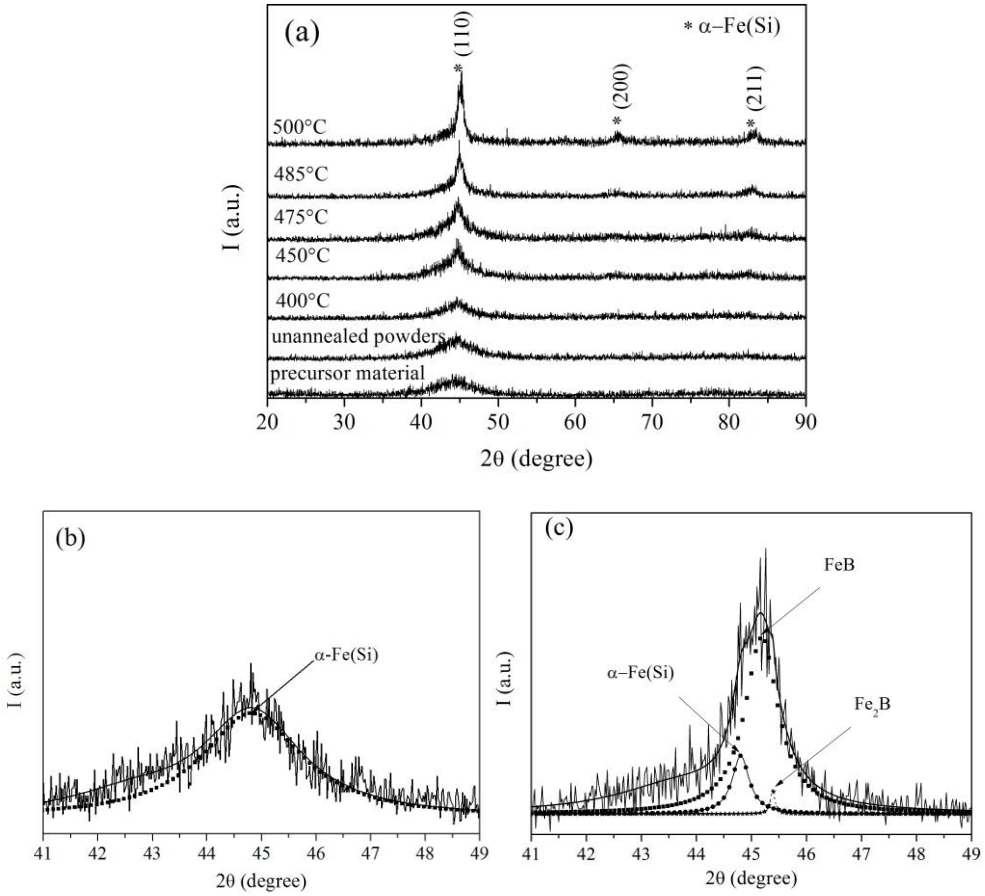

Fig. 3 – XRD patterns for precursor material, unannealed, and annealed powders (a); deconvolution for diffraction peak at 2θ = 45° for annealed powders at 475°C (b) and 500°C (c).

For powders annealed at 475°C, α-Fe(Si) grains (110) with average size about 8 nm were obtained. After annealing at temperatures above 475°C the formation of boride phases is observed. For the powders annealed at 485°C the average grain size of boride and α-Fe(Si) is 10 nm and 9 nm, respectively. By increasing the annealing temperature, the crystalline volume fraction increases. For 500°C annealing temperature the grain size of borides and α-Fe(Si) are around 25 nm and 16 nm, respectively.

Figure 4 presents the dependence of the ratio $H_{c\ \text{annealed powder}}$ / $H_{c\ \text{unannealed powder}}$ on the annealing temperature. The coercive field of unannealed powders

(H_c = 2.96kA/m) is about two times higher than that of precursor material, due to the stresses induced during the milling process. The nanocrystallization process induced by the annealing, leads to the decrease in the coercive field value. The lowest value of H_c was obtained for the powders annealed at 475°C (35% smaller than that of the unannealed powders).

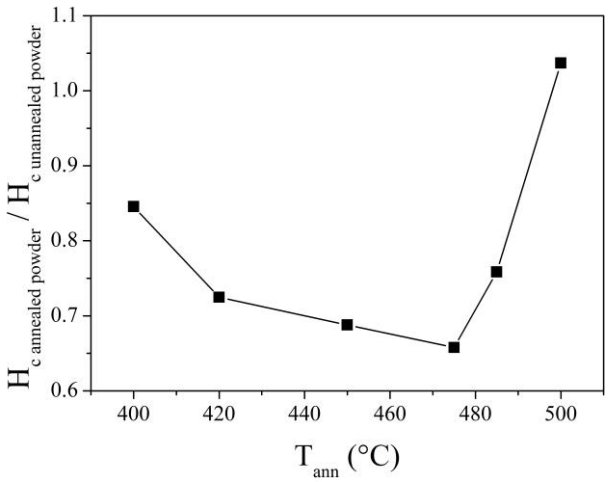

Fig. 4 – The dependence ratio $H_{c\ \text{annealed powder}}$ / $H_{c\ \text{unannealed powder}}$ on the on annealing temperature.

By increasing the annealing temperature boride phases are formed which lead to the deterioration of the soft magnetic properties [1]. These results are in good agreement with the results obtained by thermomagnetic and XRD analysis. No significant changes have been observed in the value of the saturation magnetization of the annealed powders compared with the unannealed powders.

4. CONCLUSIONS

The effect of annealing on the structural and magnetic properties of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ powders (with average size of about 30 μm) obtained after milling in argon atmosphere of amorphous ribbons were studied. The obtained powders were isothermally annealed in a furnace, in vacuum, at temperatures between 400°C and 500°C.

Diffraction patterns for annealed powders before 400°C show a typical halo of an amorphous phase (2θ = 45°). An increase of the annealing temperature above 400°C leads to the formation of a sharp diffraction peak at (2θ = 45°) corresponding to the (110) α-Fe(Si) phase, whose height increases with the increase of the annealing temperature. Powders in the nanocrystalline state with the

lowest coercive magnetic field were obtained after annealing for 1 hour at 475°C. For annealing temperatures higher than about 485°C the boride phases are present and the nanocrystalline properties are deteriorating.

These results are the basis for further studies where powders embedded in Zn-phosphate will be compacted to produce powder cores with good magnetic properties and good response in high frequencies.

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