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$Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ powders prepared by mechanical grinding: structural and magnetic properties

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$Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ powders with powder average size from about 110 μm to 30 μm were obtained by mechanical grinding of ribbons thermally treated for 1 hour at 450°C. Morphological, structural and magnetic analyses of the powders were carried out. Increasing the milling time, mechanically induced crystallization of the powders was observed. The mechanical grinding process leads in the decrease of the saturation magnetization and the increase in the coercive magnetic field. The increase of the coercive magnetic field becomes significant for powders' average size smaller than about 40 μm.

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1. Introduction

Over the last decades, the FeCuNbSiB magnetic alloys (ribbons, wires, thin films, powders) have been extensively studied due to their remarkable soft magnetic properties [1-10]. The interest in such systems displaying soft magnetic behaviour has been raised by their potential applications in a large variety of magnetic devices. The microstructure optimization, performed mostly through suitable thermal treatments which allow the amorphous-nanocrystalline transformation, is the key to improve the soft magnetic properties [1, 3, 8]. After adequate heat treatments, a fine nanocrystalline structure consisting of bcc Fe(Si) nanograins surrounded by a residual amorphous matrix is obtained. As the temperature increases above a critical value, boride phases are formed and the soft magnetic properties are deteriorated.

FeCuNbSiB powders are good candidates for magnetic cores, improving the efficiency of these devices when operating in high-frequencies regime [1, 11-13]. These powders can be obtained by mechanical grinding, mechanical alloying, atomization [9-15]. There are studies concerning the mechanical grinding-induced crystallization of some Fe-based amorphous alloys [8, 10].

The aim of this work was to investigate the grinding time influence on the morphology, structural and magnetic properties of $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ powders. The powders were prepared by mechanical grinding from ribbons with the same composition (provided by Vacuumschmelze GmbH).

2. Experimental details

In order to establish the optimum annealing temperature corresponding to the lowest coercive magnetic field / highest saturation magnetization, $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ ribbons were annealed for 1 hour in a vacuum furnace (10^{-6} Torr), at temperatures between 350°C and 550°C. The ribbons have been cut into small pieces and then sealed in a hard stainless steel jar together with stainless steel balls (the mass of ribbon to the mass of balls ratio being 1:7). The grinding was done using a SPEX Sample Prep 8000M MIXER/MILL under argon atmosphere.

The powders' morphology has been investigated using the scanning electron microscopy (SEM) technique (microscope Jeol JSM – 6390A, with W filament). The microstructure was investigated by X-ray diffraction (XRD) using a Bruker AXS D8 Advance diffractometer with Cu-K_α radiation (1.5418 Å), in Bragg-Brentano geometry. The hysteresis loops were recorded with a vibrating sample magnetometer (VSM) (LakeShore 7410 VSM). The Curie and the crystallization temperatures were determined from the magnetization dependence on the temperature (thermomagnetic curve).

3. Results and discussion

Fig. 1 presents the normalized thermomagnetic curves for ribbons in the as-cast state and after annealing at 450°C, 500°C and 550°C. The ribbon annealed at 450°C presents nanocrystalline behaviour, while the ribbons treated at 500°C and 550°C becomes crystalline. Ribbons annealed for 1 hour at 450°C have been used as precursor material in the grinding process. For the precursor material, the coercive magnetic field, H_c, and saturation magnetization, M_s, are 1.66 kA/m and 148 emu/g, respectively (about 25 % smaller and 7 % higher, respectively, than in the as-cast state).

Fig. 2 shows the $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ powder morphology after different grinding times, as well as the powder average size dependence on the grinding time. The shape of the powder particles is irregular, the size

distribution being wide for the same grinding time. The powders average size decreases when the grinding time increases (from about 110 μm to 30 μm when the grinding time increases from 10 minutes to 60 minutes).

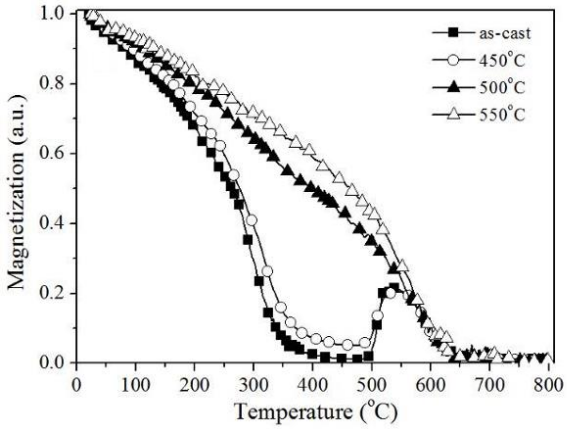

Fig. 1. Normalized thermomagnetic curves for as-cast and annealed $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ ribbons

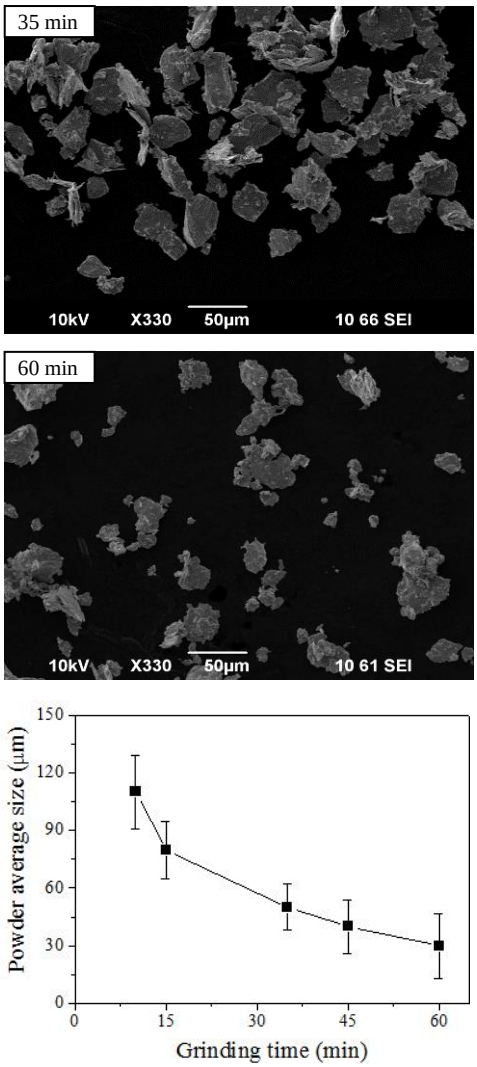

Fig. 2. $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ powders morphology and the powder average size dependence on the grinding time

Fig. 3 presents normalized thermomagnetic curves for powders obtained after 10, 35, 45 and 60 minutes grinding times, as well as the Curie temperature of the amorphous phase, T_{Ca}, dependence on the grinding time. All the powders exhibited the typical magnetic behaviour of nanocrystalline FeCuNbSiB alloys. As it can be observed, increasing the grinding time increases the mechanically induced crystallization degree. When the grinding time increases from 10 minutes to 60 minutes, the T_{Ca} value increases from 351°C to 367°C. The first crystallization temperature remains approximately constant.

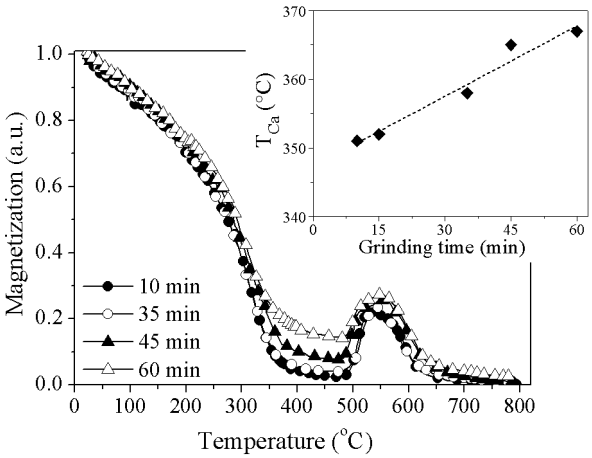

Fig. 3. Normalized thermomagnetic curves for $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ powders; T_{Ca} dependence on the grinding time (inset)

The evolution of the nanocrystalline phases indicated by the thermomagnetic curves was confirmed by XRD analysis. X-ray diffraction patterns for the precursor material, as well as for the powders are presented in Fig. 4. Increasing the milling time, the mechanically induced crystallization of the powders is observed. A (110) diffraction peak corresponding to the α-Fe(Si) phase with a bcc structure appears for the samples grinded more than 35 minutes. The intensity of (110) peak increases with increasing the grinding time, indicating that the amount of residual amorphous phase decreases. Other two diffraction peaks (200) and (211) of α-Fe(Si) appear in the diffractograms after 45 minutes of grinding. The average grain size, D, was evaluated using the Scherrer's formula, $D = 0.9\lambda / \beta \cos\theta$, where λ is the wavelength of the X-ray, β is the width at half maximum peak (in radians) and θ is the Bragg angle [8]. After 35 minutes of grinding the average crystallites size of (100) α-Fe(Si) phase was about 7 nm.

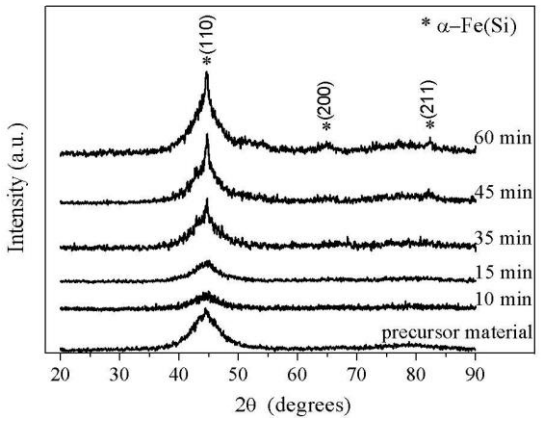

Fig. 4. XRD patterns for the precursor material and powders

Fig. 5 presents the hysteresis loops for the precursor material and for powders grinded for 35 minutes and 60 minutes. The mechanical grinding process leads to the decrease in the saturation magnetization and to the increase in the coercive magnetic field due to the anisotropy induced by stress, demagnetization effect, defects, and surface oxides.

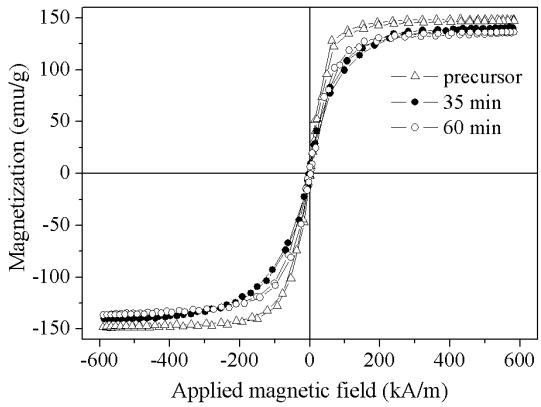

Fig. 5. Hysteresis loops of the precursor material and powders grinded for 35 and 60 minutes

The increase in the coercive magnetic field is more significant for powders' average size smaller than about 40 μm (about 9 % and 65 % for powder obtained after 35 minutes and 60 minutes grinding times, respectively, compared with the precursor material). The saturation magnetization of the powders is smaller than that of the precursor material (up to about 8 %).

4. Conclusions

The influence of grinding time (between 10 to 60 minutes) on the structural and magnetic properties of $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ powders has been investigated.

After the grinding process, mechanically-induced crystallization of the powders appears, the evolution of the nanocrystalline phases being confirmed by the thermomagnetic and XRD analysis.

As the particle size of grinded powder decreases, the coercive magnetic field increases, the increase being more significant for powders' average size smaller than about 40 μm.

In a future work, the annealing influence on the structural and magnetic properties of the powders will be analyzed.

The magnetic behaviour of cores fabricated using $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$ powders will be investigated as well.

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