Journal of Magnetism and Magnetic Materials 322 (2010) 2099–2103

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

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

Characterization of nanocrystalline FeSiCr powders prepared by ball milling

Z. Bensebaa [a,*], B. Bouzabata [a], A. Otmani [b], A. Djekoun [a], A. Kihal [a], J.M. Grenèche [c]

[a] Laboratoire de Magnétisme et de Spectroscopie des Solides (LM2S), Département de Physique, Faculté des Sciences, Université de Annaba, B.P. 12, 23000 Annaba, Algérie
[b] Laboratoire de Recherche en Physico-chimie des Surfaces et Interfaces, Facultés des Sciences, Université de Skikda, B.P. 26, Elhadack, Skikda, Algérie
[c] Laboratoire de Physique de l'Etat Condensé, UMR CNRS 6087, Université du Maine, 72085 Le Mans, France

ARTICLE INFO

Article history:
Received 10 October 2009
Received in revised form
21 January 2010
Available online 1 February 2010

Keywords:
Nanocrystalline material
Mechanical alloying
FeCrSi alloy
X-ray diffraction
^{57}Fe Mössbauer spectrometry
Magnetic property

ABSTRACT

$FeSi_{10}Cr_{10}$ powder was mechanically alloyed by high energy planetary ball milling, starting from elemental powders. The microstructural and magnetic properties of the milled powders were characterized by scanning electron microscopy, X-ray diffraction, ^{57}Fe Mössbauer spectrometry and a vibratory sample magnetometer.

After 3 h of milling, the formation of two bcc solid solutions α-Fe_1 (Si, Cr) and α-Fe_2 (Si, Cr) is observed. Their grain sizes decrease with increase in milling time attaining, at 15 h of milling, 23 and 11 nm, respectively. Mössbauer spectra of the milled powder show the presence of two components. One is a ferromagnetic type with a broad sextuplet. Its distribution of hyperfine field is characterized by high and low hyperfine field's peaks and a mean value of 26.5 T. The other is a single paramagnetic peak. Its low concentration increases to ~4% at 15 h of milling. These results can be explained by different atomic environments affected by Si or/and Cr elements, as well as the increased disordered grain boundaries.

Magnetic measurements of the milled $FeSi_{10}Cr_{10}$ alloy powder exhibit a soft ferromagnetic character with a decrease of both magnetization at saturation (Ms) and coercive force (Hc) with milling time attaining values of Ms=151 emu/g and Hc=2500 A/m at 30 h of milling time.

1. Introduction

Nanocrystalline materials have been the subject of a great number of fundamental and applied research investigations due to their potential applications in a wide variety of technological areas such as magnetic data storage, electronics, catalysis, ceramics, structural components, etc. [1]. Recently, nanocrystalline magnetic materials have been intensively investigated because of their remarkable properties such as saturation magnetization, coercivity, magnetic ordering temperature and hyperfine magnetic field, which significantly differ from those of microcrystalline materials and are sensitive to the structure and microstructure [2]. These materials contain a large fraction of atoms located in grain boundaries. Moreover, such materials are of interest for magnetic research, since the reduced grain sizes approach the single magnetic domain size, thus offering the possibility of eliminating an influence from magnetic walls [3]. Numerous techniques have been developed to produce nanometer materials. Especially, mechanical alloying (MA) has been proved to be a competitive method for making nanocrystalline and amorphous alloys [4,5]. Mechanical alloying or high energy ball milling can be used conveniently to prepare nanostructured materials with often improved mechanical and magnetic properties [6,7]. The basic mechanism in milling elemental or pre-alloyed powders consists in a succession of fracture and welding that favours the formation of particles composed of nanometer grain size [6].The microstructure of alloyed powders prepared by MA is quite different from those prepared by other techniques. In the mechanical alloying process, the alloy phase is formed by solid-state interdiffusion reaction when the individual particles are mechanically deformed by impact. It leads to extended solubilities in the solid solution at low temperatures with respect to the thermodynamical equilibrium. Besides, the mechanical alloying process provides a more convenient, simple and economical way to synthesize nanocrystalline materials than the conventional processings. Therefore, we have chosen the FeSiCr system to investigate the influence of microstructural refinement on the soft magnetic properties. FeSiCr alloys are industrially used to fabricate electromagnetic wave absorber for mobile phone, local area network, radar systems and so on. FeSiCr alloys of large grain have been used as soft magnetic materials since a long time. But its magnetism under nanometer structure has been scarcely reported. It is interesting to examine the crystal structure and its corresponding magnetism after the mean grain size reduced down to a nanometer scale or the purpose of improving the magnetic properties of materials.

* Corresponding author.
E-mail address: z_bensebaa@yahoo.fr (Z. Bensebaa).

0304-8853/$ - see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.jmmm.2010.01.040

In this paper, the effect of milling time on the morphology, microstructure and magnetic properties was investigated for the nanocrystalline $FeSi_{10}Cr_{10}$ alloy prepared by mechanical alloying.

2. Experimental procedure

The mechanical alloying of Fe–10Si–10Cr (at%) powder was carried out in a planetary ball mill (Fritsch P 7) under argon atmosphere. The starting materials were elemental powders of Fe, Si and Cr. The ball to powder weight ratio was set to 17:1 and the milling was performed at 400 rpm. Morphology evolution of particles was followed by scanning electron microscopy (SEM). Structural changes were studied by X-ray diffraction (XRD) in a Philips diffractometer using CoKα radiation (λ_{Co}=1.7889 Å). The structural and microstructural characteristics, namely lattice parameters, grain size and microstrain, were derived from the refinement of X-ray patterns using Maud program [8] based on the Rietveld method [9]. The ^{57}Fe Mössbauer spectra were performed at room temperature in a transmission geometry using a conventional constant acceleration spectrometer and fitted using the MOSFIT program [10]. The magnetic characterization was performed in dc magnetic field up to 10 T at 300 K.

3. Results and discussions

3.1. Milled particle morphology

Fig. 1 illustrates the morphological changes of the $FeSi_{10}Cr_{10}$ powder particles unmilled and milled for 5 and 30 h. As it is observed in Fig. 1 the initial elemental powders have different particle sizes and shapes: the Fe particles are spherical with a mean size of 6 μm while the Si and Cr particles exhibit an irregular ellipsoidal shape with mean sizes of about 150 and 200 μm, respectively. The milling process introduces plastic deformations of the particle constituents. At the early stages of milling, Si particles are probably trapped in the ductile particles of Fe and Cr. After 5 h of milling time, the small ductile particles of Fe are welded to larger particles, then repeatedly flattened, cold welded and fractured, with the tendency to refine with the formation of rounded particles with a mean size ranging between 15 and 40 μm. At this stage, a lamellar structure occurs in some particles. Continuous deformation up to 30 h of milling yields a steady state equilibrium between the two processes of fracture and welding. The particles that contain substantially all the starting elements in the expected proportion become finer with a more homogenous size distribution and with a mean particle size of 3 μm. This behaviour is usually observed in the milled Fe-based alloys [11].

3.2. Microstructure and structure observations

The structural evolution and phase transformation during mechanical alloying of the $FeSi_{10}Cr_{10}$ powder were followed by XRD. Fig. 2 shows the Rietveld refinement of the XRD patterns of $FeSi_{10}Cr_{10}$ powder milled for different times. At 3 h of milling, the peak of Si and Cr disappears. The best Rietveld refinement of the XRD pattern cannot be obtained by means of a single α-Fe (Si, Cr) structure with spherical grains as shown in Fig. 3a. Therefore, to describe the non-homogenous broadening of the Bragg peaks, two bcc phases assigned to the disordered solid solutions α-Fe_1 (Si, Cr) and α-Fe_2 (Si, Cr) were considered (Fig.3b). Table 1 lists the values of parameters determined using the Rietveld refinement with two phases and the obtained relative concentrations of α-Fe_1 (Si, Cr) and α-Fe_2 (Si, Cr) are about 91% and 9%, respectively.

The existence of the two bcc α-Fe (Si, Cr) phases can be explained by the different diffusion process of Si and Cr into the bcc Fe matrix induced by high energy ball milling. This effect does originate two types of atomic environments of Fe with different lattice parameters or/and grain size. A similar result has been proposed in the nanostructured $Fe_{92}P_8$ alloy [12,13]. In addition, all the diffraction peaks have a fairly large line broadening, due to internal microstrains and the formation of nanocrystalline grains [14].

The reduction of grain size with the increase of microstrains is due to the localization of plastic deformation in the form of shear bands containing a high density of dislocations that generate cells and sub-grains by self-annihilation [14]. The grain size at 15 h of milling is about 23 and 11 nm for α-Fe_1 (Si, Cr) and α-Fe_2 (Si, Cr) phase, respectively (see Table 1). Smaller grain sizes are obtained for the α-Fe_2 (Si, Cr) phase with higher deformed nanograins and high microstrains (0.56% at 15 h of milling). The variations of the

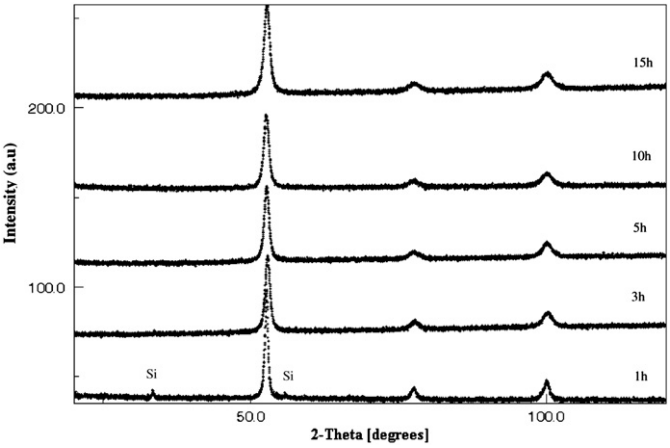

Fig. 2. XRD patterns of $FeSi_{10}Cr_{10}$ powder milled for different times.

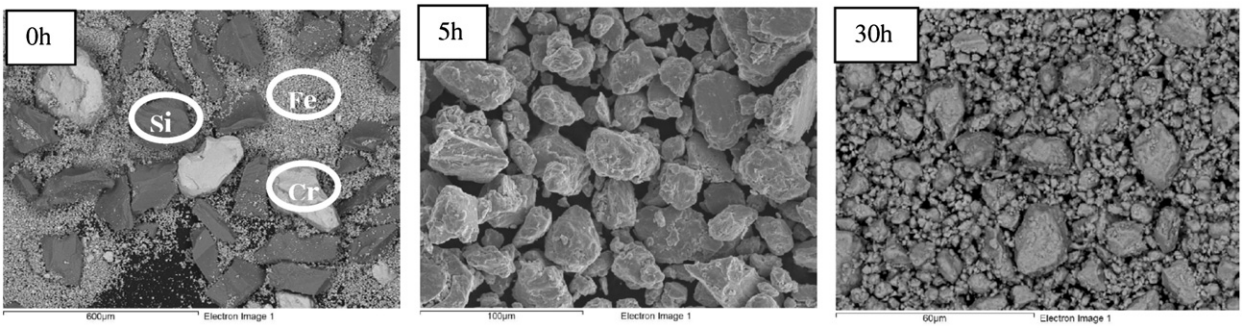

Fig. 1. Scanning electronic micrographs of the $FeSi_{10}Cr_{10}$ powder for different milling times.