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Synthesis and structural characterization of nanocrystalline FeAlNbB alloy prepared by mechanical alloying

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ABSTRACT

The technique of mechanical alloying has been used to obtain FeAlNbB alloy with nanocrystalline structure. A mixture of high purity elemental powders of Fe, Al, Nb and B with a nominal composition $Fe_{65}Al_{15}Nb_{10}B_{10}$ was subjected to intense mechanical alloying in a high-energy ball mill under argon atmosphere. Morphological, microstructural, structural and thermal characterizations of the powders milled several times were investigated by scanning electron microscopy, X-ray diffraction and differential scanning calorimetry. Experimental results demonstrated that after an optimum milling time of 50 h, FeAlNbB (B2) phase with crystallite size less than 12 nm was achieved. Ferro-paramagnetic transition at Curie temperature, T_c, strains relaxation, recovery and recrystallization are revealed in the thermal analysis in the temperature range 35–700 °C.

1. Introduction

Nanocrystalline materials have attracted considerable attention due to their improved physical, mechanical, and magnetic properties in comparison to the coarse-grained polycrystalline materials. Those properties arise from the very small grain size and consequent high density of interfaces as well as the significant fraction of atoms residing at the grain boundaries [1]. Mechanical alloying (MA) is a process able to prepare powdered alloys. Solid state reactions occur when materials are processed in a ball mill. Moreover, it is easier to produce the nanocrystalline phase in a wider composition range by MA than by solidification methods [1–3]. Nanostructured Fe-based soft magnetic materials as a new class of engineering materials can be produced by mechanical driven forces by MA [4,5]. These materials have advantages in applications like: electric and magnetic measurements, transformer magnets, information storage and magnetic cores [6]. Boron has been found to facilitate the development of amorphous and nanocrystalline structures by being introduced in solid solution into the bcc-Fe phase [7]. On the other hand, nanocrystalline Fe–(Zr,Nb)–B alloys containing bcc-Fe nanocrystallites are of interest as soft magnetic materials [8,9]. Furthermore, addition of larger atoms such as Nb, that are rejected together with B atoms at the nanocrystallization interface, is found to generate diffusion double-layers with sharp concentration gradients. Additionally, the accumulation of Nb at nanograin/amorphous phase interfaces results in a diffusion controlled grain growth responsible for the limitation of the grain size [10]. Yao et al. [11] have found that the addition of Nb can help designing Fe-based bulk metallic glasses with adequate plasticity and ultrahigh strength. This is due to several considerations: (i) it is one of the elements with the highest Poisson's ratio in metal 0.40, potentially contributing to improve material plasticity; (ii) it can act as a glue to connect the distorted trigonal prisms consisting of Fe and B, facilitating the formation of the network like structure of $(Fe,M)_{23}B_6$ [12,13] leading to high strength; (iii) furthermore, it can stabilize the supercooled liquid and hence possibly improve the BMG Formation of Fe–B-based alloys [14]. In this work we have investigated the evolution of the alloy structure during the milling process and the magnetic properties of the $Fe_{65}Al_{15}Nb_{10}B_{10}$ alloy. It is the first time that this alloy has been developed by mechanical alloying. The Fe–Nb–B system has been shown that exhibits the smallest grain size in the Cu free Fe–M–B alloys (where M=Zr, Nb, Hf) [8].

2. Materials and methods

The starting materials were pure Fe (99.7%, 10 μm), Al (99.9%, 40 μm), Nb (99.85%, 74 μm), and amorphous B (99.6%, 10 μm). Initial powders with the nominal composition of $Fe_{65}Al_{15}Nb_{10}B_{10}$ (wt%) were milled using a laboratory planetary ball mill under argon atmosphere. Ball milling experiments were carried out in a hardened steel container. The ball to powder weight ratio used was maintained as 5:1 and the milling speed was adjusted to 700 rpm. To prevent sticking of the powder to container walls and the balls, and powder agglomeration during milling, the milling

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sequence was selected such as 10 min of milling followed by 5 min of idle period. After certain intervals of milling time (2, 10, 30, 40 and 50 h), small amount of powder was taken out for X-ray diffraction (XRD) studies. XRD measurements were done using a D-500 S equipment with Cu Kα radiation. The crystallite size and the lattice strains were calculated using the Rietveld refinement with the X'Pert High Score Plus program and the morphology of the milled powder was examined using a scanning electron microscopy (SEM) in secondary electron mode operating at a voltage of 15 kV. The SEM equipped was with a Vega© Tescan make energy dispersive X-ray spectrometry (EDS) analyzer. Differential scanning calorimetry (DSC) was performed in a DSC822 apparatus of Mettler–Toledo uder argon atmosphere, in the temperature range 35–700 °C, at heating rate of 10 °C/min.

3. Results and discussion

Fig. 1 shows the morphology of the powder mixture obtained before and after 20 and 50 h milling. The unmilled powders particles have spherical and flaky structure with an average size in the range of 5–20 μm (Fig. 1a). On further milling (20 h milling) and depending on the dominant forces, a particle may either become smaller in size through fracturing or agglomerate by welding (Fig. 1b). Also, the particle morphology and the particle size distribution become nearly homogenous. At final stage of milling (50 h milling), the coarse particles fracturing as well as fine particles cold welding occurred simultaneously the brittleness of the powders is increased. This leads to formation of smaller particles with semi-spherical shape (Fig. 1c). Fig. 1d shows the EDX microanalysis performed after milling for 50 h. The composition analysis of the individual particles was found to be in good agreement with the nominal composition of the powder mixture (63.85 at% Fe–14.55 at% Al–9.85 at% Nb–9.75 at% B).

Fig. 2a illustrates the XRD patterns of milled $Fe_{65}Al_{15}Nb_{10}B_{10}$ alloy powder mixture after different milling times. In the starting powder all the XRD peaks expected of the constituent metallic elements (Fe, Al and Nb) are presented in the mixture. The diffraction peaks of B are not seen due to its low atomic scattering factor and its amorphous nature. According to both the large and negative enthalpy of mixing of the Al–B (−40 kJ/mol), Fe–B (ΔH=−45 kJ/mol), Nb–B (ΔH=−39 kJ/mol) diffusion couples [15,16], and to the fact that smaller B atom is faster diffusing than the larger Al and Nb atoms, one expects that the first reactions could be of Al–B, Fe–B and Nb–B types. Indeed, the small shift of the main diffraction peak (110) of α-Fe towards higher 2θ angles, after 10 h of milling, can be related to the lattice compression and/or the diffusion of B into Fe lattice and consequently, to the formation of bcc substitutional Fe(B) metastable solid solution (Fig. 2a). The appearance of new peak at about 2θ=37° on the left side of the main Nb diffraction peak, after 30 h of milling, can be attributed to the formation of bcc-Nb(B) solid solution with a lattice parameter close to a=3.4244 (1) Å (Fig. 2). The same result has been observed by Iizumi et al. [17] and Souilah et al. [18]. In other Nb containing nanocrystalline alloys, the metastable Fe(B) solid solution corresponds to fcc $Fe_{23}B_6$ phase which adopts $Cr_{23}C_6$ prototype structure with Fm3m space group [12]. They reported that the formation of this phase can be favored by the Nb alloying element. The peaks related to Al disappeared after 40 h of milling. In fact, Al dissolved completely in the Fe lattice to form Fe(Al) solid solution. With the increase in milling time to 50 h, only broadened peaks of FeAlNbB with bcc structure were found present. During milling, one can see the increase of the width of peaks as a result of crystallite

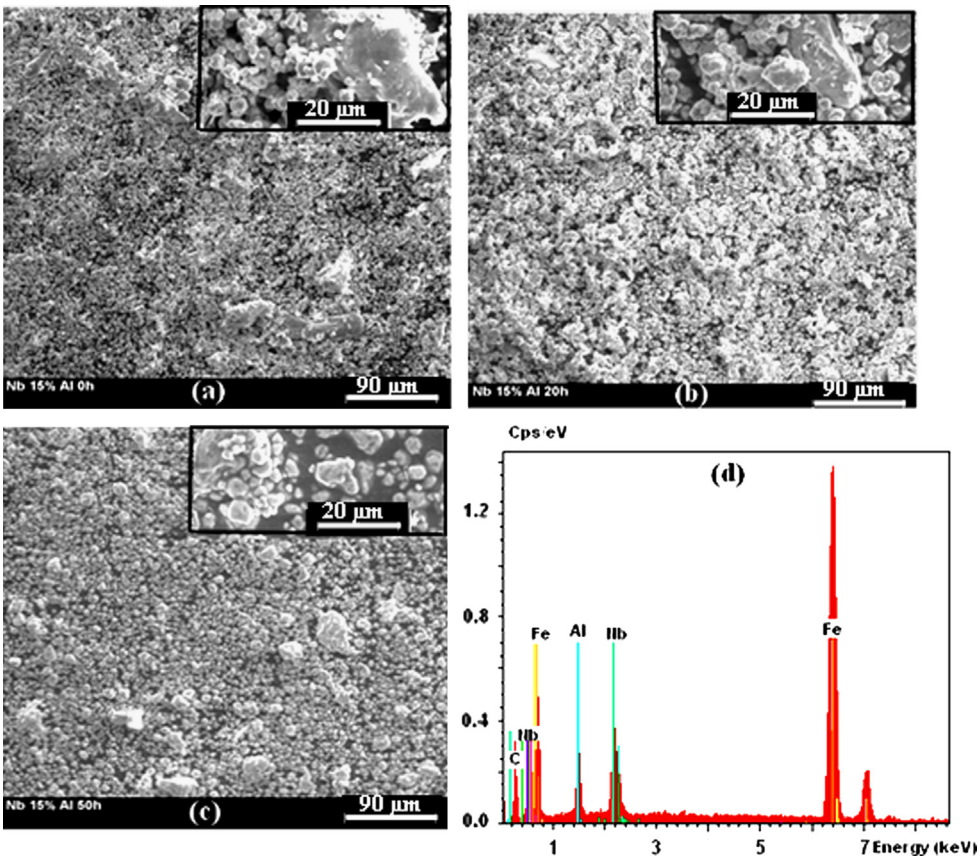

Fig. 1. Scanning electron micrographs (secondary electron image mode) corresponding to mechanically milled powders: (a) 0 h, (b) 20 h, (c) 50 h and (d) EDX microanalysis of localized zones obtained (c) after 50 h of milling.