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Original Research Paper

Magnetic and structural properties of amorphous/nanocrystalline $Fe_{42}Ni_{28}Zr_8Ta_2B_{10}C_{10}$ soft magnetic alloy produced by mechanical alloying

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Magnetic behavior, microstructural evolution, and amorphization studies of $Fe_{42}Ni_{28}Zr_8Ta_2B_{10}C_{10}$ alloy, synthetized by mechanical alloying, were investigated. The non-equilibrium microstructure originated from a grain size reduction to about 2.5 nm indicated by X-ray diffraction and the introduction of internal strain up to 3.8%. The results showed that as the milling time increased the amorphous phase became dominant and reached about 92 wt.% at 176 h. The magnetic measurements which were obtained by vibrating sample magnetometer, showed an increase in saturation magnetization up to 12 h and then a decrease until 66 h followed by a slow increase. Simultaneously, the coercivity increased, decreased and finally reached a constant level of about 24 Oe. The value of coercivity obtained in the present study is less than the values reported for the widely investigated mechanically alloyed Fe–Ni–Zr–B alloys, which shows this alloy is a very good soft magnet.

1. Introduction

Although a large number of substances are being used as magnetic materials at the present time, only three pure metals, Fe, Ni and Co, exhibit ferromagnetism at room temperature. Almost all magnetic alloys contain at least one of these ferromagnetic metals [1,2]. By alloying these metals with other elements with a suitable synthesis method the magnetic substances with various magnetic and structural properties can be prepared for different applications. In recent years, a number of magnetic alloys and special structures, like magnetic multi-layers, amorphous metals, granular alloys and others have attracted attention because of the new and striking properties they display [3,4]. Among them, soft magnetic amorphous alloys have exhibited unique soft magnetic behavior and properties such as low coercivity, large saturation magnetizations, and low hysteresis loss [5–7].

Nanocrystalline soft magnetic alloys can be described in general as $TL_{1-x}(TE,M,NM)_x$ where TL denotes a late (ferromagnetic) transition metal element, TE is an early transition metal element, M is a metalloid, and NM is a noble metal [7]. In this research, the alloying elements have been chosen based on the described formula (iron and nickel were chosen as Tl, zirconium and tantalum as Te, boron and carbon as M and noble metal was not used because usually, in solid state processes, there is no need to use noble metals which are utilized as nucleating agents [7]). Soft magnetic materials have many applications, such as transformer cores, inductive devices and high frequency antennas [8–10].

The rapid progress in research on amorphous materials over roughly the past decade has led to an understanding of many important aspects of the physical properties of amorphous materials like soft magnetic properties, high corrosion resistance and good glass-forming ability.

In this regard, many researchers have focused on the magnetic studies of Fe-based amorphous soft magnets. Inoue et al. [11], Kim et al. [12], Kopcewicz and Grabias [13], Makino et al. [14], O'Handley [15], Herzer and Warlimont [16], Miracley and Sanders [17], Chakk et al. [18] and Sharma and Suryanarayana [19] and their co-workers developed and studied many amorphous Fe-based alloys and their properties. In this work a hybrid composition of elements which was not investigated by other researchers, was selected to form $Fe_{42}Ni_{28}Zr_8Ta_2B_{10}C_{10}$ by ball milling with the objective to understand the influence of milling time on structural evolution as well as magnetic properties.

2. Experimental procedure and calculations

High-purity elemental powders, including Fe (~10 μm), Ni (~10 μm), Zr (<63 μm), Ta (~42 μm), B (~2 μm) supplied by

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Merck, and graphite, were mixed for composition of $Fe_{42}Ni_{28}Zr_8Ta_2B_{10}C_{10}$ (at.%). Mechanical alloying was carried out in a planetary ball mill having steel balls and cups, at a speed of 320 rpm, while the ball to powder weight ratio was 20:1. A total number of 10 balls were used, 5 of which had a diameter of 20 mm and 5 with a diameter of 10 mm. The vial was cylindrical with an inner diameter of 10 cm and a capacity of 80 ml. The cup and balls were made of carbon steel, and both iron and carbon were used as alloying elements in the system. The process control agents were not used during any operation to reduce the contamination. The cups were filled with high purity (>99.95%) argon gas, before any process to reduce one of the sources of contamination. To prevent probable air entrance, the cups were well-sealed. Thus contamination was unexpected during mechanical operation. The powders were milled at different times of 0.5, 12, 22, 66, 110 and 176 h and the products from the milling were analyzed by a D8 advance Bruker X-ray diffractometere using CuKα radiation ($\lambda = 0.154$ nm), with an angle range of 20–90°. The step time and size were 10 s and 0.01° respectively. The size of coherent diffraction domains of α-Fe phase and micro-strains were calculated by analyzing the XRD peak broadening using X'pert software. Three main XRD peaks were considered for low milling times and the main peak at higher milling times.

In order to determine the composition of the alloy after blending the elements, quantitative analysis was performed by XRD pattern and MAUD software for the sample milled for 0.5 h. Fig. 1 shows the X-ray spectrum adjusted with MAUD software for the analysis.

For TEM characterization the as-milled powders were immersed in pure ethanol and dispersed using an ultrasonic vibrator for about 5 min. The powders were examined by the Philips CM120 at 120 kV. Scanning electron microscopy (SEM) was employed for microstructural observation of milled powders, using a Cambridge Stereoscan scanning electron microscope.

The volumetric strain ε_V^A, due to solute addition was calculated based on Egami's model [20]. The alloying elements and compositions were designed to satisfy Egami's model to have a very high potential toward amorphization. The governing equation is:

$$\varepsilon_V^A = \frac{2}{3}\left(\frac{1-2v}{1-v}\right)\lambda_1 \qquad (1)$$

where ε_V^A is the volumetric strain, v is the Poisson's ratio (¼ for central force system [21]), and λ_1 is the mismatch between solute atoms and coordination hole of the nearest neighbor atoms.

$$\lambda_1 = \frac{\sum y_i C_i}{1+\sum C_i(y-1)}\left[\left(\frac{2}{\sum(1+x_i)}\right)^3 - 1\right] \qquad (2)$$

where x_i is the $R_{\text{solute}}/R_{\text{solvent}}$, y is the ratio of deformed volume of solute and solvent as given below, and C_i is the atomic concentration.

$$y = X^3\left[\frac{2}{3}\left(\frac{1-2v}{1-v}\right)\left(\frac{2}{1+x}\right)^3 + \frac{1+v}{3(1-v)}\right] \qquad (3)$$

In order to determine the proportion of the amorphous phase in the as-milled powder samples, the Rietveld quantitative analysis was used [22]. The method derives the amorphous phase content from the overestimation of an internal crystalline standard in the Rietveld refinement of an appropriate mixture of a standard and the sample powders [22]. The 2θ range for Rietveld method was from 30° to 135° with step size and time of 0.01° and 10 s respectively. The nanocrystalline W powder with an average crystallite size of 18.2 nm, prepared by ball milling pure W under the argon atmosphere for 1 h, was used as the standard. New powder mixtures were made slowly in pure ethanol, by mixing 75 wt.% of the as-milled alloy powders with 25 wt.% of the standard. De La Torre et al. [23] proposed Eq. (4) for the weight percentage (A) of the amorphous phase in a sample that contains a mixture of amorphous and a known crystalline phase (the standard, in our case).

$$A = (1 - (W_S/R_S)/(100 - W_S) \times 10^4 \qquad (4)$$

where W_S (%) is the weight percent of the internal standard (in this study $W_S = 25\%$) and R_S (%) is its Rietveld analyzed concentration (RAC) calculated by MAUD software [22].

The saturation magnetization (M_S), coercive field (H_C) and hysteresis loss were measured as a function of the applied magnetic field in a vibrating sample magnetometer (VSM, Dexing, Model 250) at room temperature between 0 and 10 KOe with an instrumental error of 1%.

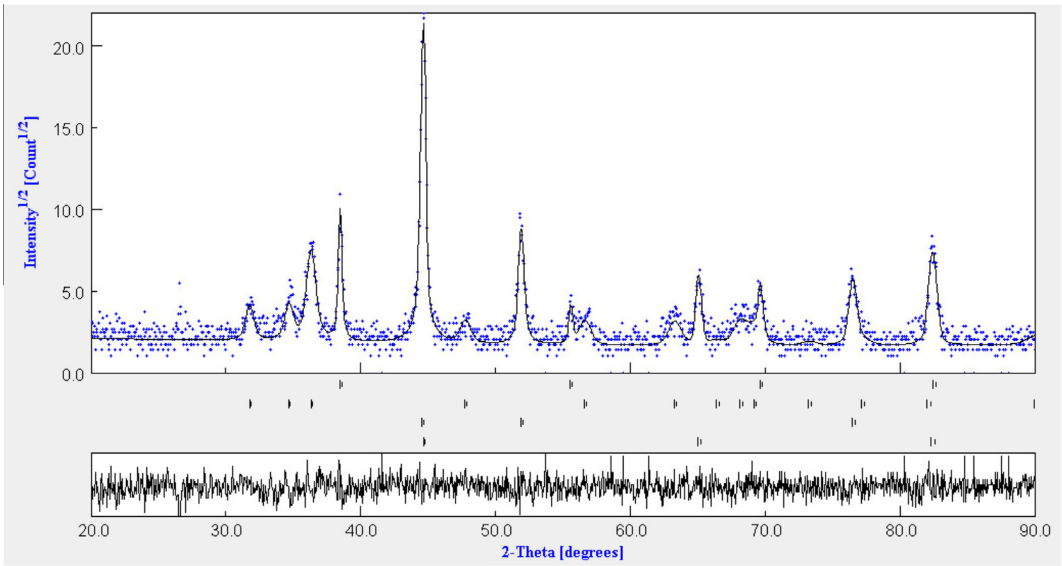

Fig. 1. X-ray spectrum adjusted with MAUD software for sample milled for 0.5 h ($R_W < 5\%$).