Magnetic and Thermal Properties of Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ Amorphous Ribbon and Ball-Milled Powders

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Received: October 11, 2014 / Accepted: October 28, 2014

Abstract

This paper reports magnetic and thermal properties of Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ amorphous ribbon and its powders prepared by ball milling. Differential thermal analysis of the amorphous ribbon showed its Curie temperature at 400 °C, and two crystalline peaks at 500 and 550 °C, respectively. X-ray diffraction analysis demonstrated that a secondary phase FeB appeared at 500 °C, causing a maximum strain of 0.195 % in the ribbon at 600 °C. The optimized soft magnetic properties of Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ ribbon were found in the sample annealed at 450 °C, with saturation magnetization $B_m = 1.84 \times 10^4$ G and coercivity $H_c = 17.95$ Oe. After ball milling, the Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ powders reached a mean particle size of 10 μm, with partial crystallization during the ball milling process. The ball milling decreased $B_m$ while increased $H_c$, due to anisotropy induced by stress.

Keywords: Amorphous ribbon, Fe-Si-B-Cu-Nb alloy, Magnetism, Phase transformation, Powder.

1. Introduction

The excellent soft magnetic properties of amorphous ribbons have been widely developed since late 1980’s (Otsuka et al., 2009). It is known that iron-based amorphous alloys have high tenacity and strength, good corrosion resistance, and can be made as thin ribbons while at a low cost. Silicon steel sheets are the earliest applications of such soft magnetic materials (Akhter et al., 2009), and their soft magnetic properties could be enhanced by annealing the ribbon in vacuum under magnetic saturation (Muhammad et al., 2014), allowing applications in cores for electrical equipment such as transformers or pulse power devices (Francoeur et al., 2012).

In order to retain the excellent magnetic and mechanical properties, these materials are usually restricted to low dimensions, such as ribbons, wires or powders. However, the working parts in devices usually require specific shapes. However, the functions of amorphous alloys are significantly influenced by shapes of sheets, wires, powders or films, because of their glass forming ability (Yoshida et al., 2000). The amorphous alloys can be fabricated by conventional casting but limited to the mold shape. Because the coercively of these amorphous alloys is lower than other types of oxide magnetic materials, many efforts are still being devoted to obtain amorphous alloy cores with desired shapes for applications in high-frequency range. The amorphous alloys offer excellent properties in coercively, but it is difficult make them into certain sharps.

Powder metallurgy provides a prospective solution to make compact products based on low-dimensional resources, enabling practical applications of metallic amorphous alloys. For this reason, how to prepare amorphous alloy powders for
the pressure molding process becomes a main research topic (Zaluska et al. 1983; Makino et al., 2009; Perigo et al., 2011; Makino, 2012; Oleksáková et al., 2013; Li et al., 2012; Inoue et al., 2001). In this study, Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ powders were prepared by a ball milling process, and their thermal and magnetic properties were investigated to optimize the process.

2. Experimental Procedures

In this research, the experimental Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ Fine-ment ribbons were provided by Hitachi Corp. The powders were prepared from ribbons by the ball milling process. Because finer particles could improve the electric property, decrease the core loss and raise the coercivity (Hsieh et al., 2009), a two-step milling process was used to check the milling effect.

The Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$, ribbons were annealing at 400, 450, 500, 550, 600, 650 and 700 °C for 15 minutes, respectively, in a vacuum to investigate their thermal effects on magnetic properties. To prepare for the ball milling, the ribbons were firstly cut into approximately square flakes and then crushed to smaller debris by a crushing machine. Two-step milling at 250 and 300 rpm for 24 and 20 hours, respectively, were sequentially preceded. For the second milling, a higher milling speed was required to produce finer powders. Finally, the smaller milling balls were used to separate the agglomerates generated from the second milling. All milling processes were done under argon atmosphere.

The morphology was observed by a Scanning Electron Microscope (SEM, Hitachi-S4700), and the average particle size was measured by a particle size distribution analyzer (Horiba La950). The phase transformations after annealing were investigated by X-ray diffraction (XRD, Rigaku DMX-2200). An LCR (L: inductance; C: capacitance; R: resistance) Meter (Agilent E4980A) was utilized to measure the permeability and quality factor at different frequency. A Vibrating Sample Magnetometer (VSM, LakeShore 7400) was used to examine the magnetic properties of ribbon or powders.

3. Results and Discussion

Figure 1 shows XRD patterns of the Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ ribbons after annealing at different temperatures. It is found that an α-Fe(Si) phase appears clearly without any other phase annealed at 450 °C. At a higher temperature, an Fe$_B$ phase appears at 500 °C. The peaks of α-Fe(Si) and Fe$_B$ become evident when the annealing temperature is raised over 500 °C. Similar phenomenon was observed during the crystallization of Fe-B-Si system via a two-step process (Inoue, 2001; Akhter et al., 2009). The internal strain can be ascribed to grain size and the secondary phase precipitates.

The peak position, θ, can be determined from the XRD pattern using Scherrer relationship:

\[ D = \frac{0.9 \lambda}{\beta \cos \theta}, \]

\[ (\beta \cos \theta)^2 = (0.9/D)^2 + 16\epsilon^2, \]

Here, \( \epsilon \) is the strain in the sample. The strains measured from the experimental samples are shown in Fig. 2. The strain rises after 450 °C because of the Fe$_B$ precipitation. The maximum strain reaches 0.195 % at 600 °C, and it then decrease after 600 °C. The internal strain can be ascribed to grain size and the secondary phase precipitates.
The particle mean size is about 80 μm after annealing. The particle sizes are statistically measured, as shown in the SEM images. The particle mean size is about 80 μm after the first milling, and which is reduced to about 10 μm after the second milling.

In order to examine the milling effect on the FeSiB powders, XRD is utilized to check their phase structure. Besides to the original alloy ribbon, a ribbon annealed at 300 °C for 1 hour is also used for ball milling. Generally, an amorphous alloy does not crystallize below the crystallization temperature. Figure 8 shows the XRD patterns of powders from the original and annealed ribbons. Only α-Fe(Si) peaks are observed, and its FWHM decreases as the milling time increases. Comparing to the XRD patterns of annealed ribbons in Fig. 1, it is found that the ball milling process yields an effect similar to annealing at 400 °C. It is because that the abraded balls and capacity of steel provide nucleation sites for α-Fe and iron-rich areas. Milling process also introduces high energy to the powders. Even after a short period of milling time, a small fraction of the powders could have enough iron concentration and activation energy to undergo crystallization (Bansal et al., 1994; Nowosielski et al., 2007). This crystallization mode is different from thermal crystallization. The thermal crystallization involves the primary crystallization (α-Fe(Si)), and the eutectic crystallization (FeB) is similar to the crystallization induced by milling under an argon atmosphere. Only primary crystallization would occur under air atmosphere (Zhang et al., 2008). There are other reasons for the partial crystallization. For example, the composition may change due to abrasion resulted by steel milling balls or container, and the milling may induce defects as preferred sites for nanocrystal precipitation (Bednarčík et al., 2004).

Figure 9 shows the VSM analysis of origin ribbons and powders. The easy axis of the induced anisotropy is transversal magnetic anisotropy. The curves of ribbons are of a low coercivity of 0.01 Oe and a high saturation magnetization of 1.67×10^4 G. After milling, the susceptibility of the linear magnetization curve decreases, which means that internal stress increases (Hofmann et al., 1996). The Hc rises to 30 Oe after the second milling, but the Hc merely goes up to 0.41 Oe after annealing without milling. The induced anisotropy may be the major reason causing increased Hc but no difference between the first and second milling.
Figure 6. SEM images of Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ particles after 1st (a) and 2nd (b) ball milling.

Figure 7. Particle size distributions of Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ powders after the 1st and 2nd milling.

Figure 8. XRD patterns of the original and annealed ribbons, and 1st and 2nd milled powders made from the ribbon annealed at 300 °C for 1 hour.
4. Conclusions

The thermal and magnetic properties of the Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ amorphous ribbon and ball-milled powders were investigated. From the amorphous ribbon, the Curie temperature was determined as 400 °C, and two crystallization temperatures were observed at 500 and 550 °C for α-Fe(Si) and Fe$_2$B, respectively. The maximum strain was inferred to be 0.195 % in the ribbon annealed at 600 °C. The optimized soft magnetic properties of Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ ribbons were found to be $B_m = 1.84 \times 10^4$ G and $H_c = 17.95$ Oe after annealing at 450 °C. After ball milling, the Fe$_{73.5}$Si$_{13.5}$B$_9$Cu$_1$Nb$_3$ powders reached a mean size of 10 μm, with partial crystallization during the milling process. The ball milling decreased the $B_m$ while increased $H_c$ due to the anisotropy induced by stress.

Acknowledgements

This work was financially supported by the Chung-Shan Institute of Science and Technology (CSIST), Taiwan.

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