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Structural and magnetic properties of ball-milled Ni₁₁Co₁₁Fe₆₆Zr₇B₄Cu powders

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Thick films of the Ni₁₁Co₁₁Fe₆₆Zr₇B₄Cu composition were synthesized via screen printing of the ball-milled ribbons of the above composition for possible use as planar inductors. The ribbons were obtained by rapid solidification. The resulting ribbon samples were annealed at 300 °C for 2 h to cause embrittlement. They were found to have soft magnetic properties ($4\pi M_s \sim 13$ kG, $\Delta H \sim 100$ Oe, and $H_c < 0.5$ Oe). The brittle ribbons were ball milled using tungsten carbide vials and stainless steel balls in an inert atmosphere for various milling times. The sample milled for 10 h was found to have a $4\pi M_s$ of about 13 kG and a coercivity of about 73 Oe with an average particle size of about 5 μm . The screen printed (as prepared and aligned) samples showed a linewidth (ΔH) of about 1000 Oe. Similar values were obtained for screen printed films annealed for 1 h at 200 and 400 °C. © 2008 American Institute of Physics. [DOI: [10.1063/1.2839315](https://doi.org/10.1063/1.2839315)]

INTRODUCTION

Over the past several years, soft magnetic materials have been investigated extensively to meet the needs for smaller passive components that operate at higher frequencies and temperatures. Today's electronics industry requires improved alloys for use in thin film inductors, which should possess a high permeability, high saturation magnetization, and low coercivity.^{1,2} Using random anisotropy model, Herzer³ described the low coercivity values by means of a volume average magnetocrystalline anisotropy. Also, a study on the effect of the composition of the transition metal on the properties, structural and magnetic, has been done by Muller *et al.*⁴ As shown by McHenry *et al.*,⁵ there has been a great deal of work related to understand and improve these materials.

In the present work, we have synthesized screen printed films of a Ni₁₁Co₁₁Fe₆₆Zr₇B₄Cu composition and studied the dc and high frequency magnetic properties. In a previous work, the structural and magnetic properties of the thin films of the same alloy prepared by pulsed laser deposition with saturation magnetization of $4\pi M_s \sim 17$ kG were presented.⁶ The primary aim of this research is to study the effect of ball milling on the structural and magnetic properties of the alloy and to employ screen printing as a method for planar micro-inductor fabrication.

Nanocrystalline soft magnetic materials (FinemetTM) were first developed by Yoshizawa *et al.* at Hitachi Metals.² In order to enhance the magnetic properties of this alloy, Willard *et al.*¹ substituted Co and Ni, creating (Fe_{1-x-y}, Co_x, Ni_y)₈₈Zr₇B₄Cu₁ alloys. These modified alloys have attractive properties when prepared as bulk ribbons¹ where they were characterized to have a nanograined micro-

structure in which body-centered-cubic (bcc) metallic crystallites were embedded within an amorphous matrix.² Here, we report the preparation of Ni₁₁Co₁₁Fe₆₆Zr₇B₄Cu by melt spinning with subsequent annealing and ball milling. The ball-milled powders were then suspended in a binder and screen printed. The structural and magnetic properties of the as-prepared, annealed, and screen printed samples are presented.

EXPERIMENTAL PROCEDURE

The alloy with the composition Ni₁₁Co₁₁Fe₆₆Zr₇B₄Cu was prepared by arc melting of elementary starting materials of 99.9% purity in an argon atmosphere. Using the single wheel melt spinning technique, the ribbons were then prepared from the alloy ingots.¹ For this purpose, a quartz crucible with an orifice of about 0.75 mm and a Cu wheel with a surface velocity of 45 m/s were utilized. The obtained ribbons had a thickness of about 30 μm and width of about 2 mm. To make these ribbons brittle, they were then annealed at different temperatures ranging from 300 to 550 °C in argon atmosphere. The ribbons annealed at 300 °C for about 2 h were then milled in a planetary ball mill operating at a speed of 250 rpm using tungsten carbide vial and a ball to powder ratio of 20:1. To avoid oxidation during milling, the powders were milled in ethanol. Test samples were taken after every 10 h of milling. The milling was carried out for about 130 h. The powders milled for 10 h were mixed with an epoxy and a hardener and screen printed on MylarTM. These films were annealed at 200 and 400 °C to burn out the binder.

Structural properties of the samples were determined by the θ - 2θ x-ray diffraction (XRD) technique using a Cu $K\alpha$ source. The particle size determination was done by scanning electron microscopy. Magnetic properties, such as coercivity,

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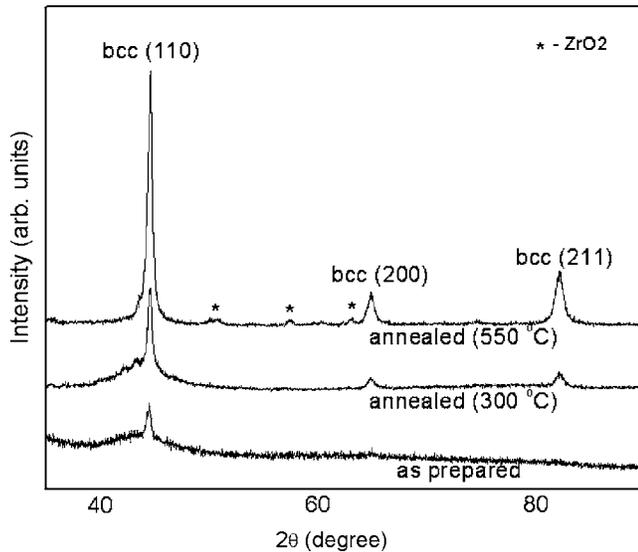


FIG. 1. X-ray diffraction of as-prepared and annealed $\text{Ni}_{11}\text{Co}_{11}\text{Fe}_{66}\text{Zr}_7\text{B}_4\text{Cu}$ ribbons.

anisotropy field, and remanent and saturation magnetizations (as $4\pi M_s$), were measured from hysteresis loops collected using a vibrating sample magnetometer. Ferromagnetic resonance (FMR) of the as-prepared annealed films and the powders was measured using a cavity technique operating in the TE_{102} mode. Room temperature FMR spectra were taken at X-band and Ka-band frequencies using a differential power absorption technique with a Varian microwave bridge as the microwave source and a Varian E-line console that included a built-in lock-in amplifier and a 100 kHz modulation signal for detection of the absorption signal.

RESULTS AND DISCUSSION

Structural properties

The XRD patterns for as-prepared and annealed samples are shown in Fig. 1. The as-prepared ribbons showed signs of crystallization. The textured crystallites were only a few nanometers in diameter and represented a small volume fraction of the ribbon since the intensities of the diffraction peaks were small. The formation of these nanocrystallites can be expected because of the presence of the high amount of ferromagnetic transition metal in this alloy that makes glass formation difficult.¹ The samples annealed at 300 and 550 °C showed broad nanocrystalline peaks from a bcc phase. From the bcc (110) peak, the lattice parameter was estimated to be about 0.287 nm, which matches closely the lattice parameter of Fe-rich FeCo alloys.⁷

The ribbons annealed at 300 °C for 2 h were ball milled for various milling times. Ball milling of these ribbons resulted in a flakelike morphology. Figure 2 shows the scanning electron micrograph of the powders milled for 10 h. The average particle size is found to be about 5 μm . Although it rapidly decreased in the first 10 h of milling, the particle size remained constant for additional milling. This can be attributed to the ductile nature of the ribbons.

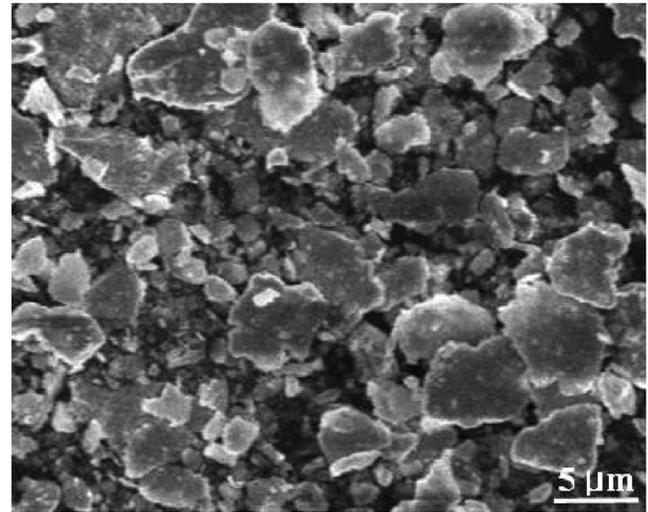


FIG. 2. Scanning electron microscope image of the powders milled for 10 h, showing the flaky particles of size less than 5 μm .

Magnetic properties

Hysteresis loops of the as-prepared, annealed, and ball-milled powders were obtained to calculate the saturation magnetization, coercivity, remanence, and uniaxial anisotropy field. The as-prepared and annealed ribbons (i.e., at 300 and 550 °C for 2 h) were found to retain the soft magnetic properties ($H_c \sim 0.5$ Oe). The coercivity for the milled powders showed a peak value after 10 h of milling ($H_c \sim 78$ Oe), decreasing as the milling time was further increased. The magnetization of the as-prepared and annealed ribbons was found to be near 13 kG. For the ribbons annealed at higher temperatures, the magnetization was found to decrease, probably due to oxidation. This is evident from the presence of zirconium oxide peaks in the sample annealed at 550 °C, as indicated in Fig. 1. For the ball-milled samples, the magnetization was found to decrease rapidly as the samples were milled beyond 10 h and remained constant at 8.5 kG as the milling time was increased. Figure 3 shows the variation of magnetization and coercivity with milling time. It can be seen that both coercivity and magnetization

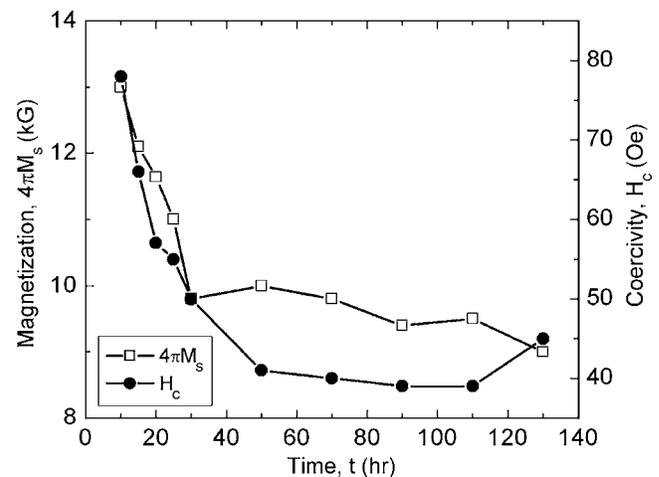


FIG. 3. Variation of magnetization and coercivity for the $\text{Ni}_{11}\text{Co}_{11}\text{Fe}_{66}\text{Zr}_7\text{B}_4\text{Cu}$ film with milling time.

decrease rapidly in the beginning and tend to remain constant as the milling time is increased. This decrease is most likely due to the partial amorphization of the alloy with increased ball milling. With continued milling, a greater volume of the powder amorphizes due to the localized heating and quenching at the point of ball-particle-ball contact. The amorphous phase has a lower coercivity and magnetization compared with the crystalline counterpart. After about 50 h of milling, there was no considerable change in $4\pi M_s$ and H_c . The samples milled for 10 h were annealed at 200 and 400 °C for about 1 h each to remove the stress generated during milling. The annealing did not have any effect on magnetization as it was found to remain constant at 13 kG.

FMR measurements were performed to determine the microwave properties of the films where the FMR linewidth ΔH was measured at a frequency of 9.63 GHz. The as prepared ribbon showed a linewidth of about 110 Oe, which remained constant in the ribbons annealed at 300 °C. The linewidth is found to increase drastically as the annealing temperature is increased, which is evident from the value of the linewidth for the ribbon annealed at 550 °C ($\Delta H = 280$ Oe). FMR measurements on the compact powders and the screen printed films show a linewidth of about 980 Oe. The screen printed films were annealed at 200 and 400 °C to burn out the binder, and FMR measurements were carried out. There was no considerable change in the linewidth, which was found to be around 1000 Oe. High field FMR measurements on the screen printed samples confirmed the magnetization values ($4\pi M_s = 13.5$ kG) of the alloy. The Lande spectroscopic splitting factor (g) for the screen printed samples was found to be 2.1. This value was deduced from the relation between resonant frequency versus resonance field, as shown in Fig. 4, was found to be similar to the values for elemental Fe, and matched the results published previously.⁶

CONCLUSIONS

The melt spun ribbons of the alloy discussed retains its soft magnetic properties on annealing at temperatures below 400 °C ($4\pi M_s \sim 13$ kG, $H_c \sim 0.5$ Oe, $\Delta H \sim 110$ Oe). Samples annealed at higher temperatures show an increase in linewidth and a decrease in $4\pi M_s$, probably due to oxidation at higher temperatures. The softest magnetic properties were obtained in the ribbons annealed at 300 °C for 2 h. These

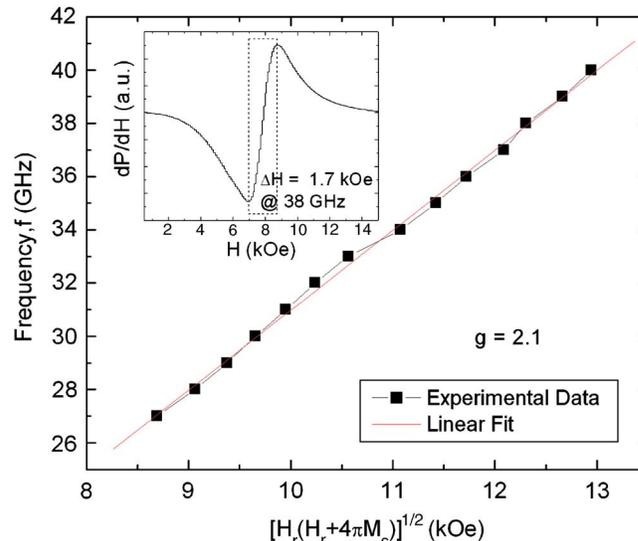


FIG. 4. (Color online) Variation of frequency vs $[H_r(H_r + 4\pi M_s)]^{1/2}$, where H_r is the resonant frequency, for the calculation of the Lande factor (g).

ribbons were ball milled for up to 130 h. The coercivity increased drastically on ball milling the sample, but it was found to decrease as the milling time was increased. Magnetization decreased rapidly with milling time and was found to remain constant as the milling time increased. The best sample obtained was after milling for 10 h ($4\pi M_s \sim 13.3$ kG, $H_c \sim 68$ Oe). This sample on screen printing showed a linewidth of about 1000 Oe, which remains constant in the samples annealed at 200 and 400 °C. This sample can be a potential candidate for application in thin film inductors.

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