Journal of Magnetics 10(4), 145-148 (2005)

Microstructures and Magnetic Properties of the Annealed FeSiB Thin Films Prepared by DC Magnetron Sputtering

T. S. Jang*, D. H. Lee, J. W. Hong[1], and J. W. Park[1]

Department of Electronic Materials Engineering, Sunmoon University, Asan, Choongnam 336-708, Korea
[1]Division of Materials Science and Engineering, Hanyang University, Seoul 133-791, Korea

(Received 8 December 2005)

Effect of vacuum annealing on the microstructures and magnetic properties of $Fe_{84}Si_6B_{10}$ films has been investigated as a function of annealing temperature. X-ray diffraction and transmission electron microscopy were employed to analyze crystallization behavior of the films. Permeability of the films was measured at various frequencies by one-turn coil method. When the films were annealed below 673 K, the coercivity of the films did not change a lot (~1500 A/m) although the grain size of a crystalline phase in the partially crystallized films increased gradually up to about 16 nm. It then increased rapidly as the films became almost fully crystalline mostly with α-(Fe,Si) phase at and above 723 K. On the other hand, the electrical resistivity of the films decreased monotonically with the increase of annealing temperature. The permeabilities of the films annealed at 473~673 K were all over 1000, showing the optimum value of 3500 at 523 K, and almost constant up to 300 MHz. However, those of the as-deposited and fully crystallized films were lower than 1000 and unstable at the same frequency range.

Key words : FeSiB films, annealing, nanocrystalline, coercivity, permeability, sputtering

1. Introduction

Many studies have been carried out to develop magnetostrictive materials for sensor and actuator applications. Among various candidates, Metglas (Fe-based amorphous alloys) is known to the best choice for magnetostrictive sensors in electronic article surveillance (EAS) system due to its large saturation magnetization, magnetostriction, and permeability, with small coercivity and eddy current loss [1-3]. The Metglas alloys are available only in the form of ribbons so far. Accordingly, the production cost of a sensor is relatively high due to complicate multiple processes and it is difficult to make the sensor smaller. Modern electronic parts, however, have a strong tendency to be light, thin, simple, and small. In that sense, thin film magnetostrictive materials that have good enough soft magnetic properties should be developed for miniaturization of the sensors. In order to develop such films, in this study, we have fabricated amorphous FeSiB thin films by DC magnetron sputtering and then examined their microstructures and magnetic properties after annealing in vacuum at various temperatures.

2. Experimental

It is known that metalloid content in the ribbon alloys for a magnetic sensor plays an important role on the magnetic properties. Therefore, the composition of films in this study was chosen similarly to that of the commercially available ribbon alloys, i.e., as $Fe_{84}Si_6B_{10}$. Amorphous films were prepared by DC magnetron sputtering. Both the target holder and the substrate holder were water cooled, and the distance between them was 5 cm. Vacuum in the chamber prior to throttling was in low 10^{-4} Pa range, and process pressure and power were 0.2 Pa and 64 W, respectively, with Ar as sputter gas. Corning glass was used as a substrate. Deposit area (1.3 × 0.85 cm) of the films was equally controlled by a mask and the thickness of the films was maintained to 100 nm. Annealing was done at 473, 523, 673, 723, 823, and 873 K, respectively, in a vacuum furnace (~10^{-3} Pa) to prevent oxidation. Heating rate and holding time were kept at 10 K/min and 1 hr, respectively.

Thickness of the deposited films was measured with an alpha step. Chemical composition of the as-deposited and

---

*Corresponding author: Tel: +82-41-530-2341, Fax: +82-41-544-1857, e-mail: tsjang@sunmoon.ac.kr the annealed films were measured by the depth profile of inductively coupled plasma (ICP) and auger electron spectroscopy (AES). Phase development and microstructural change in the films by the annealing were examined by means of Cu *Kα* x-ray diffraction (XRD) and transmission electron microscopy (TEM). Magnetic properties of the films were measured with a vibrating sample magnetometer (VSM) with a maximum applied field of 0.1 T. The resistivity of the films was measured with a four-point probe and finally the permeability was investigated by one-turn coil method.

3. Results and Discussion

The phase change in $Fe_{84}Si_6B_{10}$ films with the increase of annealing temperature is shown in Fig. 1. As reported in the crystallization behavior of the ribbon specimens [4], the amorphous state was maintained up to 673 K. From 723 K, however, α-Fe started to grow and finally the silicon-contained α-(Fe, Si) phase seemed to appear. It was reported that the tetragonal Fe_2B was also found in the annealed ribbon specimens possessing similar composition [4, 5]. To clarify the existence of Fe_2B in the film specimens, transmission electron microscopy was performed.

Fig. 2 shows bright field images and the corresponding SAD patterns of the FeSiB films. The film was virtually amorphous before annealing (Fig. 2(a)), supporting the above x-ray diffraction results. As shown in Fig. 2(b) and (c), however, the films became fully nanocrystalline when they were annealed for 1 hour above 823 K, and the Fe_2B that was not observed from the x-ray diffraction patterns seemed to be formed together with already crystallized α-(Fe,Si). It is quite possible that the formation of the Fe_2B was significantly limited because the activation energy for the formation of Fe_2B is higher than those of α-(Fe,Si) [6,

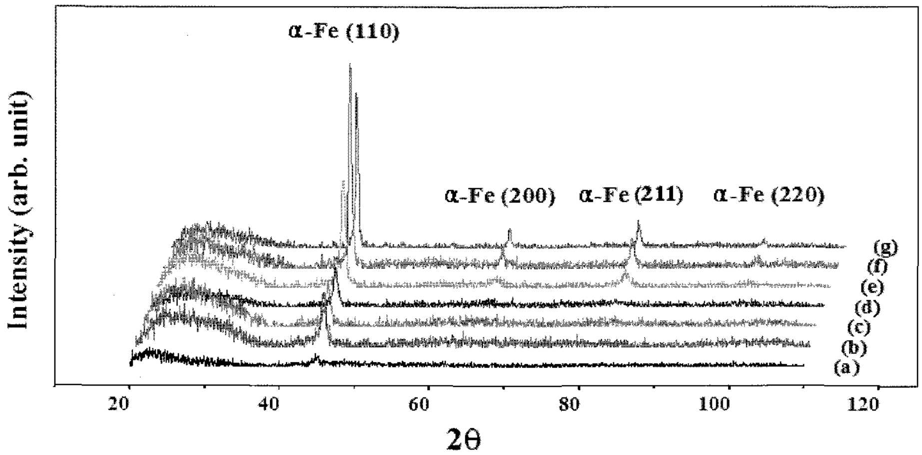

Fig. 1. X-ray diffraction patterns obtained from $Fe_{84}Si_6B_{10}$ films: (a) As deposited. From (b) to (g), annealed at 473, 523, 673, 723, 823, and 873 K, respectively.

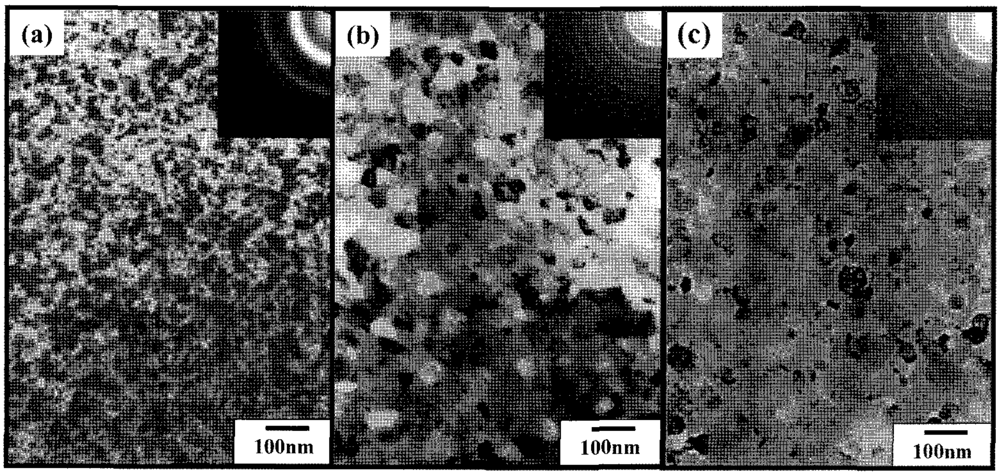

Fig. 2. TEM bright field images and the corresponding SAD patterns of $Fe_{84}Si_6B_{10}$ films: (a) As deposited. (b) Annealed at 823 K for 1 hr. (c) Annealed at 873 K for 1 hr.

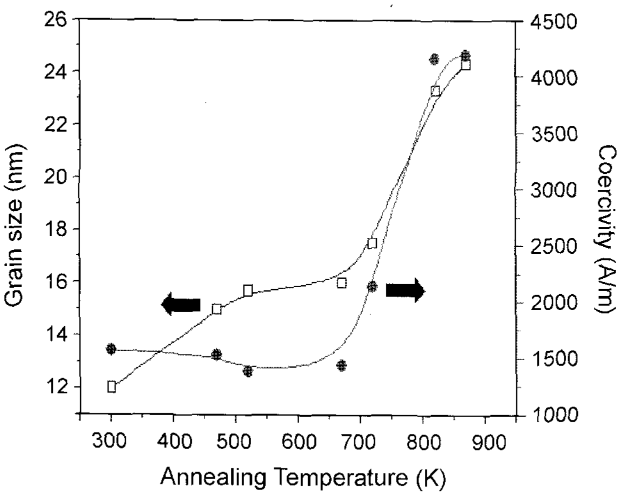

Fig. 3. Variation of the grain size and the coercivity of $Fe_{84}Si_6B_{10}$ films with the annealing temperature.

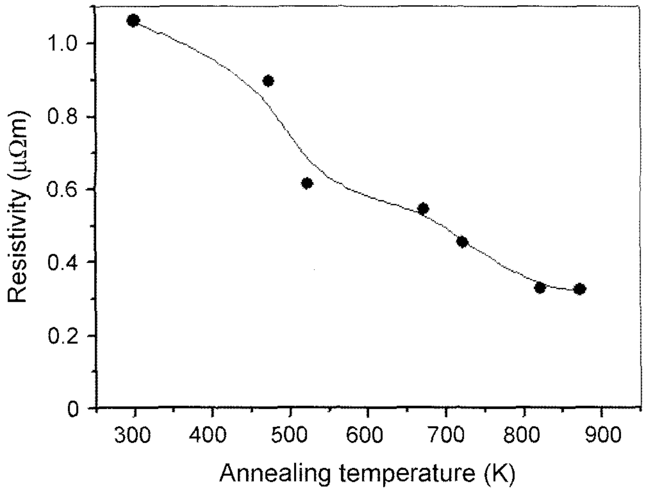

Fig. 4. Variation of the electrical resistivity of $Fe_{84}Si_6B_{10}$ films with the annealing temperature.

7].

Fig. 3 shows variation of grain size and coercivity of the FeSiB films with the annealing temperature. The gain size was estimated by using the well-known Scherrer formula [8]. The average grain size of the partially crystallized α-Fe in as-deposited film was about 12 nm. The grains soon grew to ~15 nm after annealing at 473 K, probably due to easy aggregation of unstable tiny crystallites. They then grew very slowly, maintaining the average grain size of ~16 nm up to 673 K, until the films started to become crystalline substantially. The grain size increased rapidly as the silicon-contained α-Fe fully crystallized after annealing above 723 K, resulting in the grains over 22 nm above 823 K.

It has been reported that the coercivity of a FeSiB ribbon specimen can be reduced by a low temperature annealing but it increases rapidly with the grain growth [9]. As-quenched amorphous materials generally contain many structural defects which increase internal stress throughout the material and thereby the coercivity. By a low temperature annealing that is enough to relieve the internal stress, therefore, the coercivity of the ribbon specimens can be reduced considerably. On the other hand, as shown in Fig. 3, the coercivity of the FeSiB thin films did not decrease obviously when the annealing was done below 673 K, i.e., below crystallization temperature. It indicates that the as-deposited film specimens contained fewer defects than the as-spun ribbons and, at the same time, the nano-phases partially crystallized in the amorphous films did not significantly affect the domain movement in the films. However, the coercivity increased rapidly after the films changed to crystalline by the annealing above 723 K. Main driving force for such increase of the coercivity is thought to be the formation and subsequent grain growth of α-(Fe,Si) which may suppress the domain movement. It should be noted that the coercivity of the as-deposited FeSiB film was considerably higher than that of the usual as-spun ribbon specimen although they were in similar amorphous state. It is uncertain what exactly makes such difference, but it may be partly due to higher surface anisotropy originated from larger surface to volume ratio in the film and the stress induced by the substrate of the film.

Soft magnetic films need to have low loss and high permeability for high frequency application. One important source of total loss in soft magnetic materials operated at a high-frequency range is eddy current loss which can be reduced by increasing electrical resistivity of the materials. In general, the electrical resistivity of soft magnetic films is influenced by surface defect, grain size, surface oxidation, and thickness of the films [10, 11]. As shown in Fig. 4, the electrical resistivity decreased almost linearly as the annealing temperature increased mainly due to the decrease of structural defects and the increase of grain size. Meanwhile, the permeability of the FeSiB films was measured from 10 MHz up to 1 GHz as shown in Fig. 5. The permeability measured from the films annealed at 473~673 K was all above 1000, revealing the maximum value of 3500 from the one annealed at 523 K. Furthermore, it was virtually constant up to about 300 MHz while the permeability of the as-deposited and fully crystallized films was all lower than 1000 and unstable at the same frequency range. Above 300 MHz, however, the permeability of all films, regardless of annealing temperature, was severely fluctuated as the frequency increased. It indicates that usable frequency range for the films

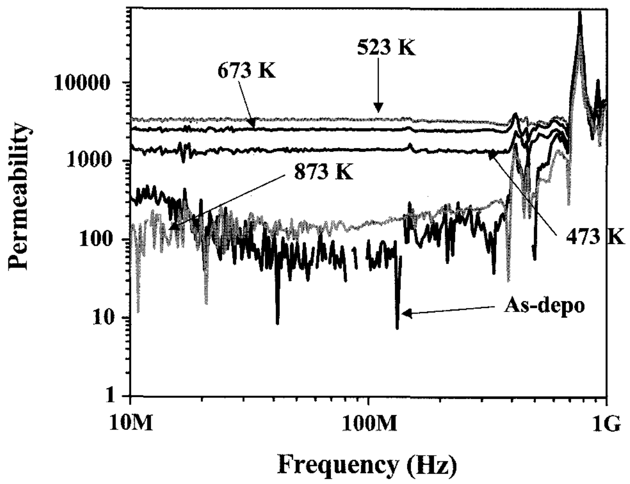

Fig. 5. Variation of the permeability of as-deposited and annealed $Fe_{84}Si_6B_{10}$ films with the applied frequency.

would be below 300 MHz. Considering the results in Fig. 4 and 5, therefore, it is desirable to anneal the FeSiB films well below 673 K to secure higher and stable permeability along with high enough electrical resistivity.

4. Conclusions

Microstructures and magnetic properties of $Fe_{84}Si_6B_{10}$ films prepared by DC magnetron sputtering were investigated after annealing at various temperatures. When the films were annealed below 673 K, the coercivity of the films did not change much although the grain size of a crystalline phase in the partially crystallized films increased gradually up to about 16 nm. It then increased rapidly as the films became fully crystalline mostly with silicon-contained α-(Fe,Si) phase above 723 K. The electrical resistivity of the films, however, decreased almost linearly with the increase of annealing temperature. The permeability of the films annealed at 473~673 K was all over 1,000 and almost constant up to 300 MHz while that of the as-deposited and fully crystallized films, annealed above 723 K, was lower and very unstable at the same frequency range. For high frequency application, therefore, it is desirable to anneal the FeSiB thin films well below 673 K to obtain good soft magnetic properties such as high permeability that is stable enough at high frequency range, high electrical resistivity as well as low coercivity.

References

[1] R. C. O'Handley, J. Mater. Eng. Performance **2**, 211 (1993).
[2] C. K. Kim and R. C. O'Handley, Metal. Mat. Trans. A **27A**, 3203 (1996).
[3] V. Franco, C. F. Conde, and A. Conde, J. Magn. Magn. Mat. **185**, 353 (1998).
[4] Alicja zaluska and Henryk Matyja, J. Mat. Sci. **18**, 1263 (1983).
[5] C. K. Kim, Mat. Sci. Eng. B **39**, 195 (1996).
[6] X. D. Liu, K. Lu, B. Z. Ding, and Z. Q. Hu, Mat. Sci. Eng. A. **179/180**, 386 (1994).
[7] Z. H. Lai, J. Conrad, G. Q. Teng, and Y. S. Chao, Mat. Sci. Eng. A. **287**, 238 (2000).
[8] B. D. Cullity, Elements of X-ray Diffraction, 2nd ed., Addison-Wesley (1978), p 284.
[9] H. Kronmuller, J. Appl. Phys. **52**, 1859 (1981).
[10] Y. K. Kim, D. Choi, K. H. Kim, J. Kim, S. H. Han, and H. J. Kim, J. Magn. Magn. Mat. **239**, 498 (2002).
[11] H. Y. Wang, Y. J. He, Z. W. Ma, E. Y. Jiang, H. S. Huang, and W. H. Mao, J. Appl. Phys. **85**, 3745 (1995).