

Magnetostrictive properties of Sm-Fe-N and Sm-Fe-C thin film prepared by reactive sputtering process

by

Tempei TANAKAMARU^{*1}, Jamadil Azwad PAZER^{*1}, Koji MAKITA^{*1} and
Yoshihito MATSUMURA^{*2}

(Received on Mar.31th,2008 and accepted on Jul.9th,2008)

Abstract

Sm-Fe thin films as giant magnetostrictive materials were prepared by the d.c. magnetron sputtering process mixed with Ar-N₂ or mixed Ar-CH₄ gas. Diffraction peaks of Sm nitride and Fe nitride were observed in formed films prepared at 2.3 vol % N₂ partial pressure. Diffraction peaks of Sm carbide and SmFe₅ were observed in formed films prepared at 2.0 vol % CH₄ partial pressure. With increasing N₂ and CH₄ partial pressure, saturated magnetostriction of formed thin films decreased. The maximum value of magnetostrictive susceptibility of thin films prepared was seen at 1.0 vol % of N₂ partial pressure and 0.5 vol % of CH₄ partial gas pressure. In the films, compressive stress was observed with the small addition of nitrogen atoms and carbon atoms into Sm-Fe thin films, and this may lead to increase of magnetostrictive susceptibility.

Keywords: nitrogen gas, methane gas, giant magnetostriction, magnetron sputtering, internal stress

1. Introduction

Giant magnetostrictive materials (GMM) such as SmFe₂, TbFe₂, and Terfenol-D have been investigated for actuator and sensor devices^{1,2)}. The GMM have many advantages such as high responsibility, high out put power and low voltage operation comparing with other smart materials. The GMM thin film has been applied to devices for micro-machines, opto-magnetic systems and surface acoustic wave filters^{3,4)}. Large magnetostriction and high Magnetostrictive susceptibility are required for those applications of GMM thin film. We have systematically investigated Magnetostrictive properties of the GMM films by using various thin film formation processes, such as flash evaporation, ion beam sputtering, ion plating, electron beam evaporation, and magnetron sputtering⁵⁻¹⁰⁾. As a result, magnetostrictive characteristics are strongly effected by internal stress change due to ion bombardment¹¹⁾. Lim et al., have reported that Sm-Fe-B thin film shows high magnetostrictive susceptibility comparing with Sm-Fe thin films because of lattice expansion by interstitial boron atoms¹²⁾. Nitrogen and carbon atoms are similar to atomic radius of boron atom and Fe-N and Sm-Fe-N which have good magnetic properties¹³⁾. In this study, Sm-Fe-N and Sm-Fe-C thin films were prepared by d.c. magnetron reactive sputtering process in mixed Ar-N₂ or Ar-CH₄ gaseous atmosphere.

2. Experiment

2.1 Film preparation

Sm-Fe-N and Sm-Fe-C thin films were prepared by d.c. magnetron reactive sputtering process as shown in Fig.1. Argon (purity: 99.999%), argon + nitrogen (N₂ content: 5.0vol %) and methane (purity: 99.99%) were used as sputtering gas.

Reactive sputtering process SmFe₂ alloy was used as a sputtering target. Single crystalline Si (100) chips (25x5x0.28 mm) were used as substrate. A sputtering chamber was evacuated down to 1.0 x 10⁻⁴ Pa. Sputtering gas was kept at 1.0x10⁻¹ Pa. Substrate temperature was maintained at room temperature. Thin films were formed at 100 W for an hour after pre-sputtering of 5 minutes. The film thickness was ranged from 2.6 mm to 3.2mm.

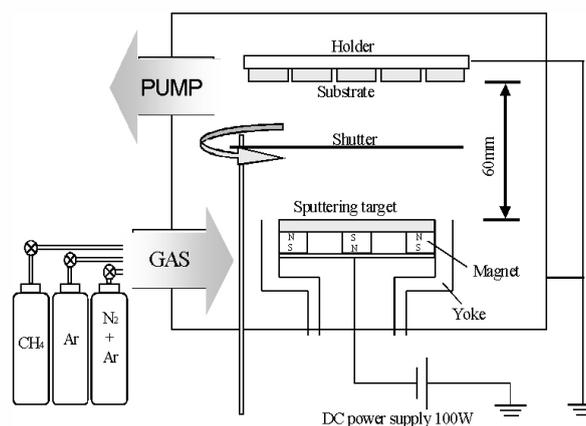


Fig.1: Schematic diagram of d.c. magnetron

2.2 Sample analysis

The structure of the formed films was examined and observed by X-ray diffraction (XRD) using Cu-K α radiation. The composition of the film samples was determined using energy dispersive X-ray spectroscopy (EDX). Magnetostriction of the films was measured using a cantilever method¹⁴⁾. Thickness of prepared thin films measured by bench-top surface profiler (DEKTAK3). Internal stress, σ was estimated by Stoney's equation (1) and was also based on results of substrate curvature measurements¹⁴⁾,

$$\sigma = E_s t_s^2 / 6(1 - \nu_s) t_f R \quad (1)$$

Subscript s and f represent substrate and film, t is thickness, E is Young's modulus, ν is Poisson's ratio and R is radius of the curvature, where $R = l^2 / 2d$.

3. Results and Discussion

3.1 Composition and Crystal structure

The film sample with Sm₂₇Fe₇₃ was obtained by sputtering process. XRD diffraction patterns of the Sm-Fe-N and Sm-Fe-C films are shown in Fig.2 and Fig.3. No distinct XRD peaks of the SmFe₂ phase were observed in as-deposited samples, suggesting amorphous like or strongly disordered structures of each film

*1 Graduate Student, Course of Applied Science,
*2 Professor, Department of Energy Engineering,

samples¹⁵). In this work, the pressure of sputtering gas was kept at 1.0×10^{-1} Pa. A mean free path of vapour flux of the target materials could be longer than target to substrate distance.

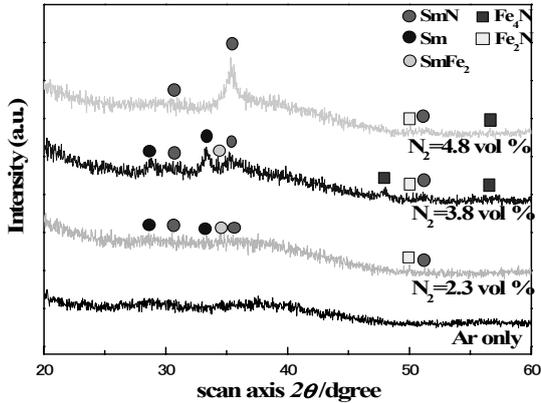


Fig.2 XRD patterns of Sm-Fe thin films prepared at various N₂ partial pressure.

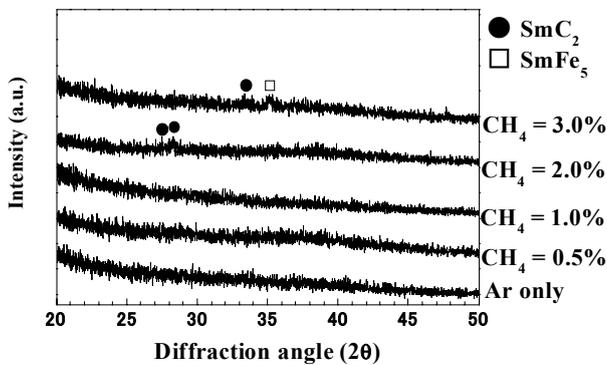


Fig.3 XRD patterns of Sm-Fe thin films prepared at various CH₄ partial pressure.

3.2 Magnetostrictive properties

Fig.4 shows in-plane saturated magnetosriction of Sm-Fe thin films prepared at N₂ or CH₄ various partial pressures. The magnetostriction of film samples decreased with increasing of N₂ and CH₄ partial pressure. It showed magnetostriction of Sm-Fe film prepared at 4.8 vol % of N₂ and 3.0 vol % of CH₄ partial pressure were 600 ppm and 740 ppm at 1200 kA/m of applied magnetic field. Fig.5 shows magnetostrictive susceptibility of Sm-Fe thin films prepared at N₂ or CH₄ various partial pressures. It showed maximum value of magnetostrictive susceptibility of Sm-Fe film prepared at 1.0 vol % of N₂ partial pressure was 34 ppm/Am⁻¹. Where maximum value of Magnetostrictive susceptibility of Sm-Fe film prepared at 0.5 vol % of CH₄ partial pressure was 26 ppm/Am⁻¹. In this study, Sm-Fe-N and Sm-Fe-C thin films show high magnetostrictive susceptibility comparing with Sm-Fe thin film because of lattice expansion by interstitial nitrogen and carbon atoms. Sm₂₇Fe₇₃ was decomposed during films deposition process because SmN and SmC₂ was precipitated when N₂ and CH₄ partial pressure increased. Magnetostriction of film was decreased with disproportionation of Sm₂₇Fe₇₃ to SmN and α-Fe at nitrogen addition as well as SmC₂ and SmFe₅ at carbon addition^{16,17}.

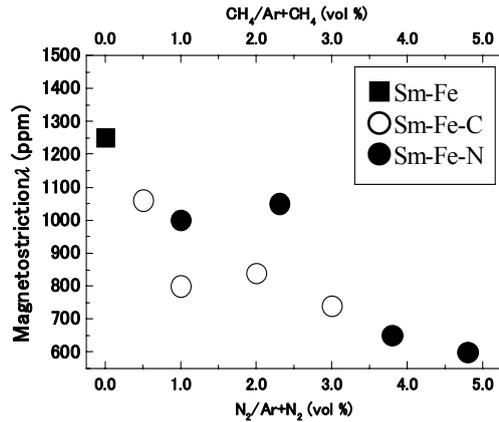


Fig.4 Saturated magnetosriction of Sm-Fe thin film prepared at N₂ or CH₄ various partial pressure.

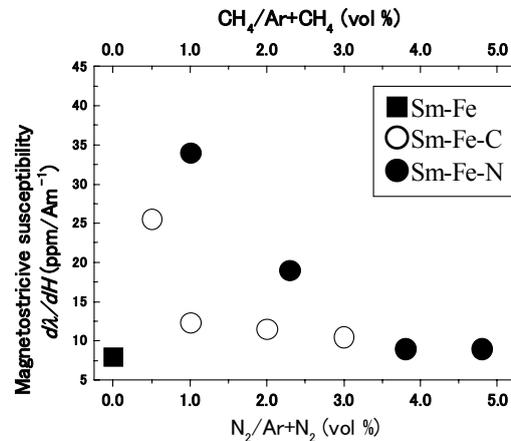


Fig.5 Magnetostrictive susceptibility of Sm-Fe thin Films prepared at N₂ or CH₄ various partial pressure.

3.3 Internal stress

Fig.6 shows internal stress of Sm-Fe thin films prepared at N₂ or CH₄ various partial pressure. In this study all, of formed film showed compressive stress. The figure shows 0.41 GPa of compressive stress has showed at Sm-Fe thin film. The compressive stress in thin films was increase to 0.70 GPa at all amount of nitrogen addition as shown in Fig.5. Where the compressive stress in thin films was increase to 0.55 GPa at all amount of carbon addition. Magnetostrictive susceptibility was increased due to magneto-elastic energy with compressive internal stress of thin film.

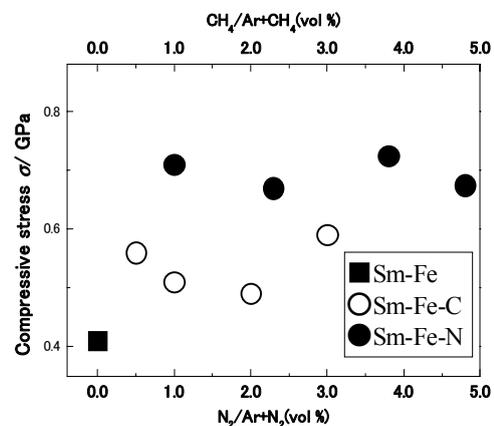


Fig.6 Internal stress of Sm-Fe thin films prepared at N₂ or CH₄ various partial pressure.

4. Conclusion

Sm-Fe-N and Sm-Fe-C thin films as giant magnetostrictive materials were prepared by d.c. magnetron reactive sputtering process. The saturated magnetostriction of film samples decreased with increasing of N₂ partial pressure. Where, saturated magnetostriction of film samples decreased with increasing of CH₄ partial pressure. Sm₂₇Fe₇₃ was disproportionate to SmC₂ and SmFe₅ with CH₄ with carbon content. Therefore Magnetostriction was decreased.

In spite of saturated magnetostriction, Magnetostrictive susceptibility showed maximum value when 1.0 vol % of N₂ partial pressure and 0.5 vol % of CH₄ partial pressure. These are because applied compressive stress was increased with nitrogen and carbon addition. Therefore it is effective to Magnetostrictive properties that a little amount of nitrogen or carbon addition of thin film.

Acknowledgments

This study was financially supported by the Japan Society for the Promotion of Science (JSPS) for Grant in-Aid for Scientific Research (C) as “Design of Magnetostrictive Susceptibility on Thin Film” with No.19560709. And this study was made in the frame of Development of Advanced Production processes for Energy Conversion Materials, Future Science & Technology Joint Research Center, Tokai University.

5. References

- 1) *A. E. Clark*, in E. P. Wohlfarth(ed.): Ferromagnetic Materials, Vol. 1. North Holland, Amsterdam, 1980, Chap. 7.
- 2) *H. Wakiwaka and Y. Yamada*, J. Magn. Soc. Jpn, 25, 8(2001)1425-1433
- 3) *M. Sahashi and T. Kovayashi*: J Acoustical Soc. Japan 466 (1990) 591-599.
- 4) *V. Koeninger, Y. Matsumura, H.H. Uchida and H. Uchida*: J.Alloy.Comp. 211/212 (1994) 581-584.
- 5) *M. Morimoto, T. Inagaki, T. Kobayashi, Y. Fujiwara, M. Masuda, S. Shiomi and T. Shiratori*:J. Magn. Soc. Jpn. 27 269 (2003).
- 6) *S.M. Na, S.J. Suh and S.H. Lim*: J. Appl. Phys. 93 8507 (2003).
- 7) *T.M. Danh, N.H. Duc, H.N. Thanh and J. Teillet*: J. Appl. Phys. 87, (2000) 7208-7212
- 8) *S. Esho and S. Fujiwara*: AIP Conf. Proc., 34, 331 (1976).
- 9) *N. Heiman, A. Onton, D.F. Kyser, K. Lee and C.R. Guarniere*: AIP Conf. Proc. 24, 573 (1975).
- 10) *R. Zwingman, W.L. Wilson, Jr. and H.C. Bournon*: AIP Conf. Proc. 34, 334 (1976).
- 11) *M. Takeuchi, Y. Matsumura and H. Uchida*: J.Japan Inst. Metal Vol.69 No.8 (2005) 667-670.
- 12) *S.H.Lim et al*. J.Magn. Mater. 239(2002)
- 13) *M.Takahashi et al*. J. Magn. Mater. 239 (2002)
- 14) *A.C.Tam and H.Schroeder*: IEEE Trans. Magn. 25(1989) 2629-2638
- 15) *K. Muramatsu, T.O. Yamaki, N. Matsuoka, M. Takeuchi, Y. Matsumura and H. Uchida*: Journal of Alloys and Compounds. Vol.408-412, Page335-339 (2006)
- 16) *T.Miyazaki, S.Yamaguchi, M.Takahashi, A.Yoshihara, T.Shimamori and T.Wakiyama*: J. Magn. Mater. 75 (1992) 243-251.
- 17) *T.Honda, U.Hayashi, K.I.Arai, K.Ishiyama and M.Yamaguchi*: IEEE Trans. Magn. 29 (1993) 3126-3128.