Transactions of the Materials Research Society of Japan 29[4] 1631-1634 (2004)

High-Rate and Low-Temperature Sputter-Deposition of Ni-Zn Ferrite Thin-Films

Tomoharu Ogita, Setsuo Yamamoto, Gen Ishida, Hiroki Kurisu and, Mitsuru Matsuura, Yoshihiro Shimosato*, and Shigenobu Okada*
Faculty of Engineering, Yamaguchi University 2-16-1 Tokiwadai, Ube-shi,Yamaguchi, 755-8611 Japan
Fax: 81-836-85-9621, e-mail: yamamoto@yamaguchi-u.ac.jp
*SHIMADZU Corporation, 380-1 Horiyamasita, Hadano-shi Kanagawa, 259-1304 Japan
Fax: 81-463-88-8832, e-mail: s-okada@shimadzu.co.jp

Preparation method of Ni-Zn ferrite thin-films for high frequency magnetic devices has been developed. Introduction of reactive sputtering method utilizing a dense and active electron-cyclotron-resonance (ECR) microwave plasma using metal sputtering targets enabled us low-temperature and high rate deposition. Using three 100 mm square platelet targets equipped in the vicinity of the plasma extraction window in the ECR sputtering apparatus, Ni-Zn ferrite thin-films with saturation magnetization of 224 emu/cc and relatively low coercivity of 15 Oe were obtained at 200 degrees C and deposition rate of 14 nm/min. By replacing the targets to a conic target whose area is 2.5 times as large as total area of the three-platelet targets, maximum deposition rate of 44 nm/min was achieved with ensuring the same magnetic properties. Reactive ECR sputtering is one of the most promising deposition methods of ferrite thin-films.

Key words: ECR sputtering, Ni-Zn ferrite, ferrite thin-film, high deposition rate, low temperature sputtering

1. INTRODUCTION

A novel sputtering method using an Electron-Cyclotron-Resonance microwave plasma, in short "ECR sputtering" has some advantages over conventional diode or magnetron sputtering method [1,2]. Firstly, independent and precise process control is possible with respect to plasma generation, sputtering and film deposition. Secondly, the ECR sputtering is suitable for low temperature deposition of oxide or nitride thin-films, which need chemical reaction during the film growth because the ECR microwave plasma is dense and contains many energetically excited ions. We have already designed a ECR sputtering apparatus and proved that the Co-containing spinel ferrite thin-films with a high coercivity of 3000 Oe for perpendicular magnetic recording media can be deposited at a temperature lower than 200 degrees C using the ECR sputtering apparatus [3,4].

To apply soft magnetic ferrite thin-films to advanced magnetic devices such as MMIC (Monolithic Microwave Integrated Circuit), thin-film isolator and circulator, acceptable highest deposition temperature is at most 300 degrees C, and high deposition rate is necessary to meet the demand of relatively large film thickness over micrometer. In our previous study, three 100 mm square Ni-Zn-Fe platelet targets were equipped near the plasma extraction window in the ECR sputtering apparatus [5].

In this study, to increase deposition rate, a conic target whose area is 2.5 times larger than the total area of the platelet targets were introduced in our ECR sputtering apparatus.

2. EXPERIMENTAL

Figure 1 shows the configuration of the ECR sputtering apparatus (SHIMADZU corporation, SLC-75ES) used in this experiment. Plasma was generated by the combination of a 2.45 GHz microwave and an 875 Oe magnetic field, which satisfied ECR condition. Dense and active plasma was generated by ECR phenomenon. As process gas, argon and oxygen gases were separately introduced in the plasma generation chamber and near the substrate in the film deposition chamber, respectively. Ni-Zn ferrite thin-films were deposited without substrate heating. However, the

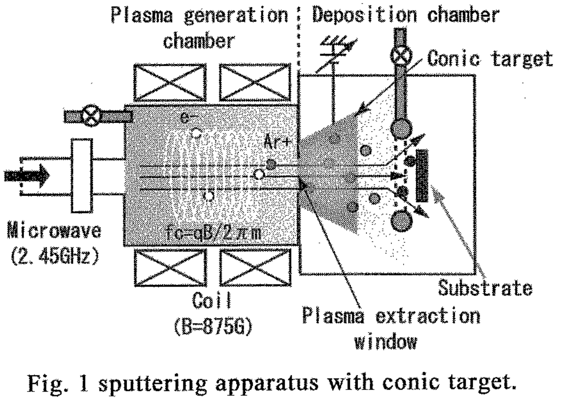


Fig. 1 sputtering apparatus with conic target.

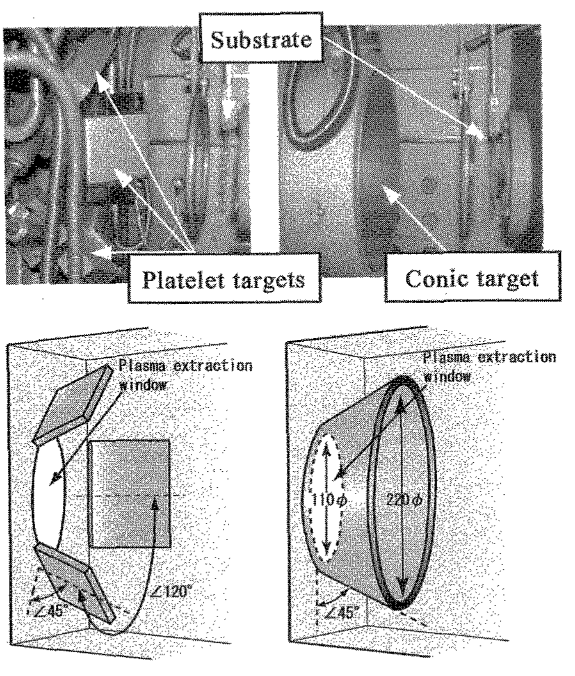
substrate temperature rose up to 250 degree C during film deposition by the plasma irradiation to the substrate. $Ni_{0.3}$-$Zn_{0.5}$-$Fe_{1.4}$ (wt%) alloy targets were used.

To achieve a high rate deposition, the followings were realized in the ECR sputtering apparatus. To obtain a high density and active plasma, a microwave power supply with a high output power of 900 W was used for the plasma volume of 5100 cm^3. Targets were placed in the vicinity of the plasma extraction window to utilize the high-density plasma, and were placed to make so-called "On-axis configuration" to the substrate. To operate the target surface in "metal mode", oxygen gas was introduced near the substrate, because it was expected that the targets are not easily oxidized and the oxygen gas excited by ECR plasma stream oxidizes the film effectively.

Figure 2 shows schematic illustrations of (a) conventional platelet targets and (b) conic target. Conventional target is composed of three 100 mm square platelet targets equipped around the plasma extraction window with an target angle separation of 120° forming an angle of 45° between the target and substrate surfaces. Total area of the platelet targets for sputtering was 300 cm^2.

To increase the deposition rate, a conic target whose area (750 cm^2) is 2.5 times larger than the total area of the conventional targets (three-platelet targets) was introduced and the effectiveness were studied in comparison with the platelet targets. The conic target has an inner and outer diameter of 110mm ϕ and 220mm ϕ, respectively. A slant angel of the sidewall of the conic target is 45°.

Replacement of the target from three platelets to conic one will bring us some merits. Firstly, the conic target surrounds the plasma extraction windows completely, sputtering target area increases. Secondly, high density and active plasma localized in the vicinity of the plasma extraction window can be used effectively because the distance between the sputtering target and plasma extraction window can be smaller in the conic target use than in platelet targets use. These two factors will lead to an increase of film deposition rate. Thirdly, the distribution in thickness or magnetic properties of the deposited thin-films will be improved as compared with the platelet targets that have 120 degrees symmetry.



(a) Platelet targets (b) Conic target

Fig 2. Sputtering targets for ECR sputtering. Targets are shown with gray color in illustrations.

3. RESULTS AND DISCUSSION

3.1 Effect of microwave input power and target voltage on target current

The target current which expresses the quantity of ion bombarding to the sputtering targets per unit time was measured with varying microwave input power from 200 to 700 W and target-voltage from –200 to –600 V in case of platelet targets and conic target. As shown in figure 3, target current is proportional to microwave input power. On the other hand, dependence of target current on target voltage is very small. This result shows that the plasma density is controlled dominantly by microwave input power in an ECR sputtering apparatus. On the other hand, it is not true in conventional sputtering where plasma density is strongly influenced by target voltage. This is the typical data showing that the plasma generation and sputtering are controlled independently in ECR sputtering.

The slope of the target current vs. microwave input power density curve for the conic target is 2.5 times as much as that for the platelet targets.

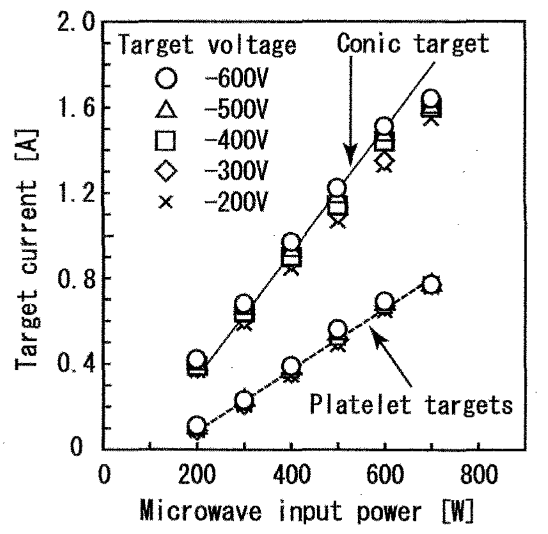


Fig. 3 Microwave input power dependence of target current in relation to target voltage.

This is due to sputtering area increased by 2.5 times. Increase in target current achieves high rate deposition.

3.2 Properties of Ni-Zn ferrite thin-films

At first, 400 nm thick Ni-Zn ferrite thin-films were deposited at a fixed total gas pressure (Ar + O_2) of 0.55 Pa. In case of platelet targets use, microwave input power was set at 500 W, target voltage, –350 V. In case of conic target use, microwave input power was set at 600 W, and target voltage, –400 V. In both cases, oxygen gas partial pressure was varied to search optimal condition to obtain high quality Ni-Zn ferrite thin-film.

Figure 4 shows the oxygen gas flow ratio dependence of saturation magnetization (M_S) and deposition rate of ferrite thin-films in use of the three-platelet targets and the conic target. Grey zone shows the expected saturation magnetization of Ni-Zn bulk ferrite with same composition as the target. In the use of platelet targets, the ferrite thin-films with M_S of 224 emu/cc was obtained at deposition rate of 14 nm/min at oxygen gas flow ratio of 5%. Because of increased area of the target, use of the conic target brought us about 3 times higher deposition ratio. To obtain ferrite thin-films with the reasonable M_S in the grey zone, use of the conic target needed about 2.6 times as high as oxygen gas flow rate than the use of the platelet targets. At the oxygen gas flow ratio of 13%, the ferrite thin-films with reasonable M_S of 290 emu/cc and low coercivity of 11 Oe could be deposited using the conic target at a high deposition rate of 44 nm/min.

Figure 5 shows the XRD diagrams for the Ni-Zn ferrite thin-films deposited at the various oxygen gas flow ratio in case of (a) platelet targets and (b) conic target use. In platelet targets use, highest diffraction peak from spinel (400) was observed at an oxygen gas flow ratio of 5%. On the other hand, in case of conic target use, the highest diffraction peak was observed at an

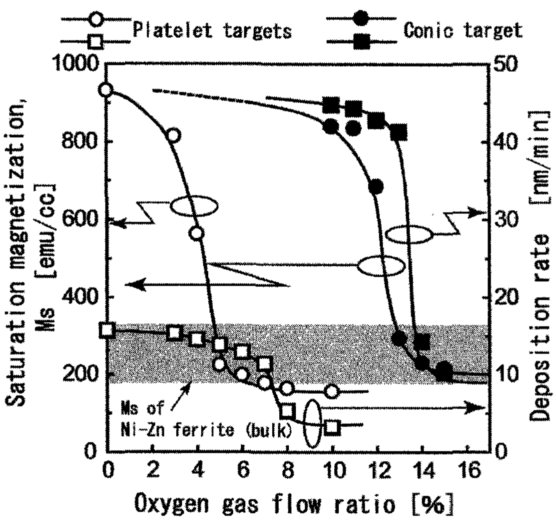
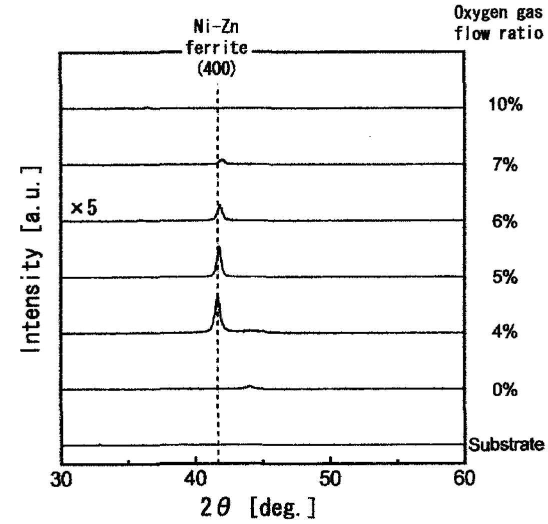
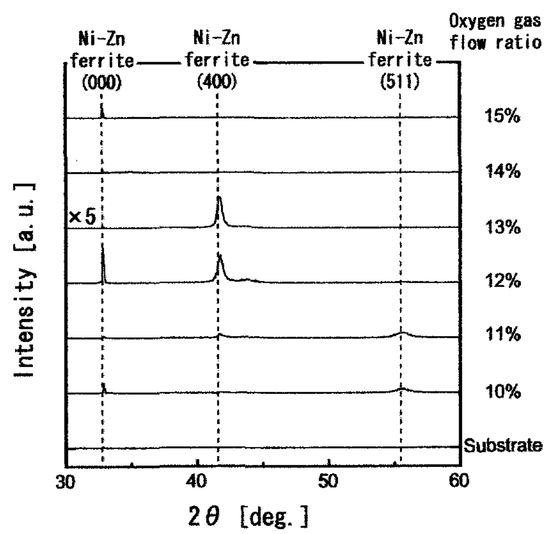


Fig. 4 Oxygen gas flow ratio dependence of saturation magnetization and deposition rate of Ni-Zn ferrite films.



(a) Platelet targets use



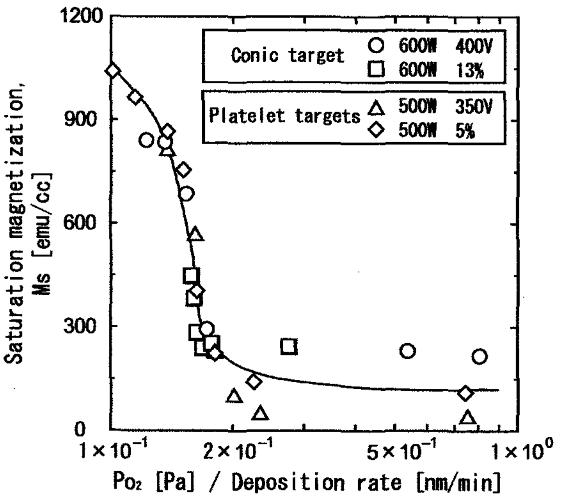
(b) Conic target use

Fig. 5 X-ray diffraction diagrams for the Ni-Zn ferrite thin-films deposited at various oxygen gas flow ratio.

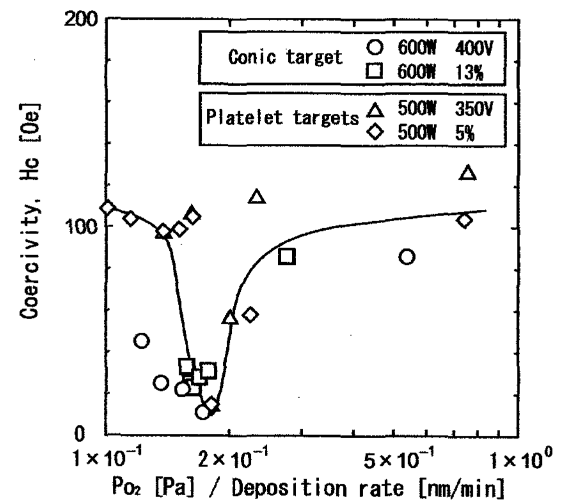
oxygen gas flow ratio of 13%. From the viewpoint of crystalinity, optimal value of oxygen gas flow ratio in case of conic target is 2.6 times as high as that of platelet target. This optimal oxygen gas flow ratio coincides with that which produces proper saturation magnetization in Ni-Zn ferrite thin-films as shown in Fig. 4.

Optimal oxygen gas flow ratio in conic target use is very critical as compared with platelet target use. Only 1% deviation of oxygen gas flow ratio from optimal condition caused the significant deterioration of crystallinity and appearance of diffraction peaks from another planes. These facts suggest that the precise oxygen gas flow control is required in high rate deposition.

From the experimental results described above, it is predicted that, in order to obtain the identical



(a) Saturation magnetization



(b) Coercivity

Fig. 6 Relationships between magnetic properties and oxygen gas partial pressure normalized by deposition rate.

ferrite thin-films in oxidation degree, magnetic and crystallographic properties, at a different deposition rate, oxygen gas flow ratio should be increased in proportional to deposition rate.

To confirm this prediction, magnetic properties were plotted against oxygen gas partial pressure normalized by deposition rate in Fig.6. The data for many Ni-Zn ferrite thin-films deposited using conic and platelets targets at different microwave input power, target voltage and oxygen gas flow ratio are included.

Oxygen gas partial pressure normalized by deposition rate dependence of (a) saturation magnetization and (b) coercivity are expressed by unique curve in both cases.

Saturation magnetization decreases with increasing oxygen gas partial pressure normalized by deposition rate. From Figure 6(a), it is found that, to obtain Ni-Zn ferite thin-films with a saturation magnetization of 200-320 emu/cc which is same as that of Ni-Zn bulk ferrite, oxygen gas partial pressure normalized by deposition rate should be in the range from 1.7×10^{-1} to 2.5×10^{-1} [Pa/(nm/min.)]. In this condition, low coercivity Ni-Zn ferrite thin-films were obtained as shown in Fig.6 (b).

4. CONCLUSIONS

A reactive sputtering method using Electron-Cyclotron-Resonance microwave plasma has been developed for deposition of Ni-Zn ferrite thin-films for high frequency magnetic devices. By using conic target, Ni-Zn spinel ferrite thin-films with a preferential orientation of (400) and relatively low coecivity of 11 Oe and reasonable saturation magnetization of 290emu/cc were obtained at a high deposition rate of 44nm/min. Also it was found that precise control of oxygen gas partial pressure normalized by deposition rate is important in this sputtering system.

Finally, we conclude that reactive ECR sputtering is one of the most promising deposition methods of ferrite thin-films.

This work was financially supported by Ministry of Economy, Trade and Industry, Ministry of Education, and Culture, Sports, Science and Technology. The authors would like to express their deepest appreciation to Professor Yoshihisa Nakamura, director of RIEC, Tohoku University and Professor Emeritus Masahiko Naoe of Tokyo Institute of Technology for their useful advice and encouragement.

5. REFERENCES

[1] T. Ono, C. Takahashi, and S. Matsuo, Jpn. J. Appl. Phys., vol.23, no.8 (1984), pp.L534-L536.

[2] O. Kinoshita, High Density Plasma Application Technologies, Realize Inc (1993), pp.160-173.

[3] S. Yamamoto, H. Wada, H. Kurisu, M. Matsuura, Special Issue of Journal of Magnetism and Materials, vol.235 (2001), pp.133-137.

[4] S. Yamamoto, K. Hirata, H. Kurisu, M. Matsuura, T. Doi, and K. Tamari, J. Magn. Magn. Mater., vol.235 (2001), pp.342-346.

[5] H. Wada, S. Yamamoto, H. Kurisu, M. Matsuura, and Y. Shimosato, Transactions of the Materials Research Society of Japan, MRS-J, Vol.128, No.4 (2003). (in press)

(Received October 8, 2003; Accepted February 18, 2004)