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Ni-Zn ferrite films synthesized from aqueous solution usable for sheet-type conducted noise suppressors in GHz range

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Abstract $Ni_{0.2}Zn_{0.3}Fe_{2.5}O_4$ films (1–5 μm thick) were deposited by spin spray ferrite plating from an aqueous solutions onto polyimide sheets at 90°C. Their peel test and high-frequency permeability as well as noise suppression effects were investigated. The oxygen plasma treatment on polyimide sheet surface improved the film adhesion. There was not visible crack on the bended film surface for the Ni-Zn ferrite film thinner than 2 μm and was not peeled off even after the bending test of a million times. The films exhibited excellent high-frequency permeability profile and a natural resonance frequency (where the imaginary permeability reaches a maximum) f_r was 370 ± 30 MHz. The transmission loss increased with the film thickness, reaching the maximum $\Delta P_{\mathrm{loss}} = 70\%$ at 8 GHz for the 5 μm-thick film. The reflection loss in the measured frequency range was $S_{11} < 10\%$ which is small enough for films to be used as the conducted noise suppressors. The value of ΔP_{loss} obtained for the 5-μm thick film was about 15% higher than that ($\Delta P_{\mathrm{loss}} = 55\%$) attained by the commercialized 50-μm thick noise suppressing sheet.

Keywords Ni-Zn ferrite film · Spin spray ferrite plating · Conducted noise suppressors · Adhesion property · Mechanical durability

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1 Introduction

The electromagnetic interference (EMI) is a crucial problem for facilitating stable operation of the most advanced and downsized personal computers and cellular phones working at frequency higher than 1 GHz [1]. The noises in GHz range cannot be suppressed efficiently by lumped constant circuits using coils and capacitors. They can be suppressed sufficiently by placing magnetic bodies in the vicinity of the noise sources as distributed constant circuits. Before radiated as noise waves, noise energies are dissipated by magnetic loss that is expressed by the imaginary part, μ'', of the complex permeability $\mu = \mu' - j\mu''$ of the magnetic bodies [2]. They are called "conducted noise suppressors" [3] and composite magnetic sheets, in which Fe-Si-Al flakes are dispersed in flexible polymer resins, have already commercialized and widely used in cellular phones and mobile computers. However, these sheets are facing the dilemma that increasing permeability by increasing the volume fraction of the ferromagnetic metal flakes inevitably decreased electrical resistivity due to percolation of the metal flakes. This degrades high frequency permeability, which limits the usable frequency and the minimum thickness of the suppressors to a few GHz and 50 μm, respectively. In order to meet the demands for further downsizing of devices and increasing their working frequency, we have developed a novel type of the conducted noise suppressors using ferrite films [4]. We succeeded in preparing Ni-Zn ferrite films from an aqueous solution on smooth glass substrate only at 90°C and they exhibited high permeability up to GHz range, exceeding Snoek's limit for bulk ferrite samples by one order of magnitude [5].

The flexible printed circuits (FPC), which are used in bendable section of cellular phones and mobile computers, are one of noise sources. Therefore, the flexibility is also required for noise suppressors.

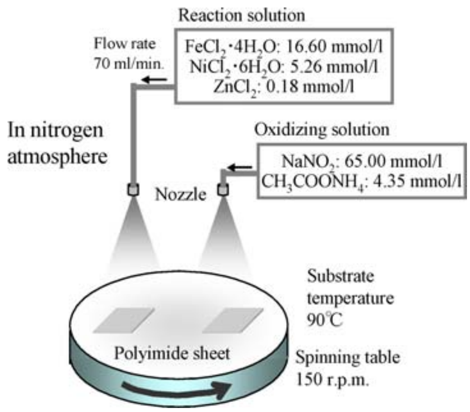

Fig. 1 Spin-spray ferrite plating

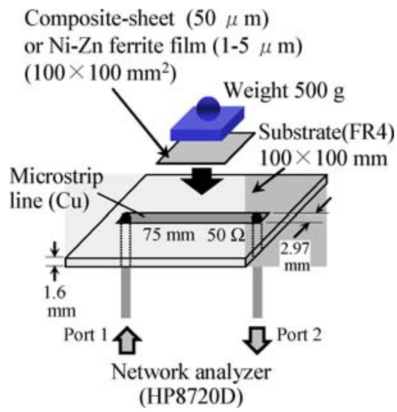

Fig. 3 Transmission and reflection measurements

In this study, therefore, we evaluated the adhesion property and the mechanical durability of ferrite film deposited on polyimide sheet as well as their noise suppression properties up to 10 GHz.

2 Experiment

Ni-Zn ferrite films 1–5 μm in thickness were deposited on to the polyimide sheets (25 μm) by spin-spray ferrite plating. In this method, a reaction solution of $FeCl_2 + NiCl_2 + ZnCl_2$ and an oxidizing solution of $NaNO_2 + CH_3COONH_4$ (pH buffer) were sprayed simultaneously onto the rotating substrates at 90°C as shown in Fig. 1. The adhesions of the films prepared with and without oxygen plasma were evaluated by the cross-cut adhesion test, in which the deposited film surfaces were scratched in a reticulate pattern with 1 mm intervals. The deposited films 17 mm in width and 50 mm in length were subjected to the bending test as shown in Fig. 2. The samples were fixed to the stator and the slider on their ends with bending radius of 1.5 mm, and the slider was reciprocated 1 million times with constant stroke of 30 mm at 2 back-and-forth motions per second.

The crystallographic and microstructural properties were analyzed by X-ray (Cu-K$_\alpha$) diffraction and scanning electron microscopy (SEM), respectively. Chemical composition was evaluated by inductively coupled plasma spectrometry (ICPS). Magnetic measurements were made by a vibrating sample magnetometer (VSM) for static M-H loops, and by a permeance meter equipped with a shielded loop coil [6] for complex permeability ($\mu = \mu' - j\mu''$) spectra up to 3 GHz. We evaluated the conducted noise suppression from 50 MHz to 10 GHz by using a 50-Ω microstrip line connected to a network analyzer (HP8720D) as shown in Fig. 3. Transmission parameters S_{11} and S_{21} of the Ni-Zn ferrite film (35 × 35 mm) were measured by pressing the ferrite film side to the microstrip line using a 500 g weight. The transmission loss ΔP_{loss} was calculated from S_{11} and S_{21} using the following equations:

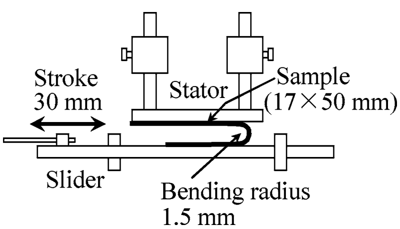

Fig. 2 Bending tester

$$P_{\text{loss}} = 1 - (|\Gamma|^2 + |T|^2)$$

$$\Delta P_{\text{loss}} = P_{\text{loss}}(\text{ferrite film} + \text{microstrip line}) - P_{\text{loss}}(\text{microstrip line}),$$

where reflection Γ and transmission T are defined as $S_{11} = 20 \log|\Gamma|$ and $S_{21} = 20 \log|T|$. For comparison, measurement was also done on a commercialized noise suppressing sheet (50 μm thick), in which ferromagnetic metal flakes are embedded in a flexible polymer matrix.

3 Results and discussion

The films deposited on polyimide sheets had the film composition of $Ni_{0.2}Zn_{0.3}Fe_{2.5}O_4$. The film thicknesses were 2 and 5 μm for 1.5 and 5 h deposition as shown in Fig. 4. All films had columnar structures aligned perpendicular to the film plane.

The films are of a single phase spinel structure having a preferential orientation of (111) parallel to the film plane as shown in Fig. 5. The (111) orientation, in which the most closely packed plane of oxygen are parallel to film plane, became more prominent as the thickness increased.

The adhesion of the 2 μm-thick Ni-Zn ferrite films prepared without oxygen plasma treatment was weak and several peeled-off area were observed after the cross-cut adhesion

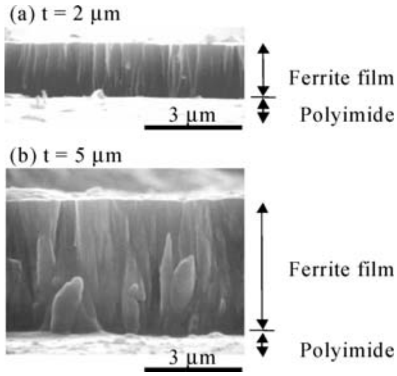

Fig. 4 Cross-sectional SEM images for the ferrite films deposited on polyimide

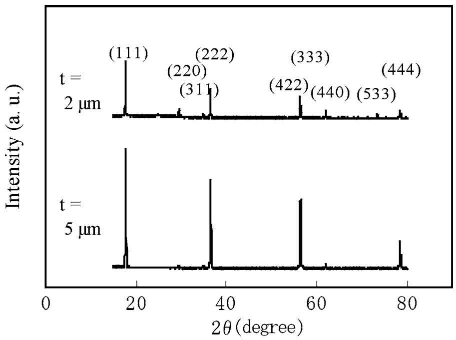

Fig. 5 X-ray diffraction diagrams for the ferrite films plated onto polyimide sheets

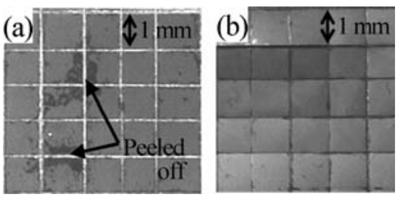

Fig. 6 Results of cross-cut adhesion test for Ni-Zn ferrite films (2 μm) prepared on polyimide sheet (a) without oxygen plasma treatment and (b) with that at 50 W for 5 min

test. However, the oxygen plasma treatment at 50 W for 5 min improved the adhesion and no peeled-off area was observed as shown in Fig. 6.

The film up to 2 μm in thickness deposited with plasma treatment did not shown visible crack when it was bended as shown in Fig. 7(a) and was not peeled off even after the bending test as shown in Fig. 7(b). In addition to the improved anchor effect, the increasing of the number of OH^- on polyimide sheet surface, which work as adsorption sites for metallic ions in ferrite plating process, caused this stronger adhesion. The columnar boundaries as seen in Fig. 4 work to relax the mechanical stress. Their numbers decreased with the increase of film thickness because of the columnar growth. The critical thickness for observing cracks by SEM after bending test was about 3 μm in this study.

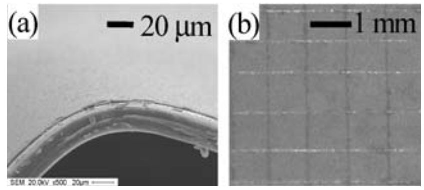

Fig. 7 (a) SEM image of bended Ni-Zn ferrite films (2 μm) on polyimide sheet and (b) result of cross-cut adhesion test after bending test of 1 million times

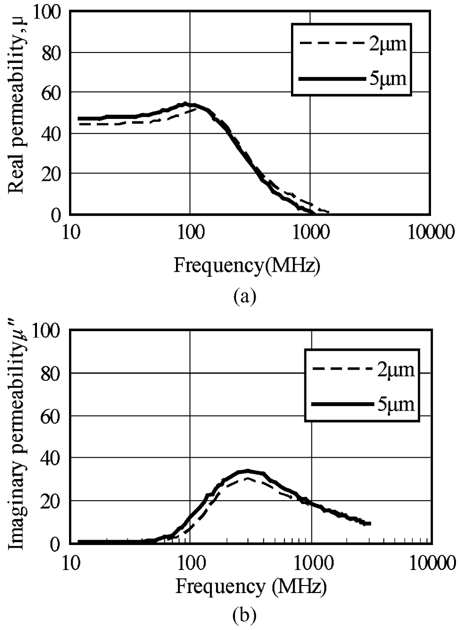

Fig. 8 (a) Real and (b) imaginary permeability spectra for the ferrite films deposited on polyimide sheets

As shown in Fig. 8, the Ni-Zn ferrite films 2 and 5 μm in thickness exhibited $\mu_r' = 45$ and the natural resonance frequency f_r, which is defined here as the frequency of the permeability of imaginary part μ_r'' peak, of about 350 MHz. The f_r was higher than that for bulk Ni-Zn ferrite samples (90–120 MHz) having the same μ_r'. This is because the magnetic moment of film samples are confined in the film plane.

The f_r for film and bulk sample is expressed by Eqs. (1) and (2), respectively [7].

$$f_r = \frac{\gamma}{2\pi}\sqrt{\frac{H_k M_s}{\mu_0}} \tag{1}$$

$$f_r = \frac{\gamma}{2\pi}\frac{H_k}{\mu_0} \tag{2}$$

Here γ is the gyromagnetic ratio, μ_0 is the permeability of a vacuum.

Figure 9(a) shows that ΔP_{loss} in the GHz range increased with the film thickness. ΔP_{loss} for the 5-μm ferrite film

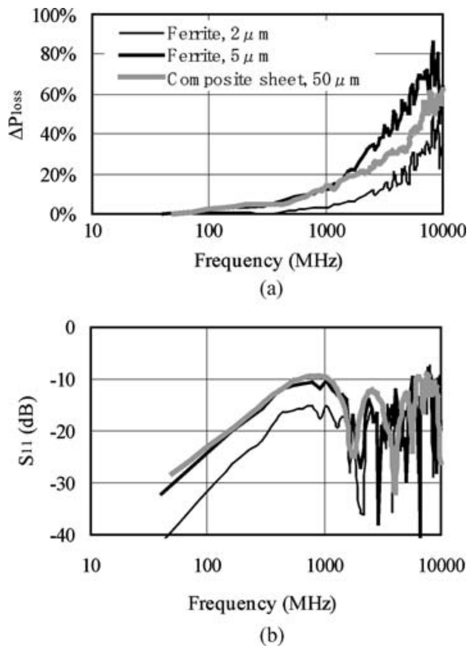

Fig. 9 (a) Transmission loss ΔP_{loss} and (b) Reflectivity S_{11} profiles for ferrite films 2 and 5 μm deposited onto polyimide sheets

sample reached 70% at 8 GHz, which is about 15% higher than that obtained for the 50-μm thick composite sheet. Though S_{11} increases with film thickness as shown in Fig. 9(b), S_{11} for the 5-μm film is 10% or less ($\leq$ 10 dB) throughout our measurement frequency range. This is sufficiently low for practical applications and about equal to that for the commercialized composite sheet.

4 Conclusions

The oxygen plasma treatment on polyimide sheets surface improved the Ni-Zn ferrite film adhesion. There was not visible crack on the bended film surface for the Ni-Zn ferrite film thinner than 2 μm and was not peeled off even after the bending test of a million times with bending radius of 1.5 mm. They exhibited sufficiently high magnetic loss and satisfactorily low reflection loss, enabling application to noise suppressors working up to 10 GHz.

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