Spin-Sprayed Ferrite Films Highly Permeable in the GHz Range, with Excellent Heat Resistance

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Previously we revealed that spin-sprayed ferrite films are usable for GHz-range noise suppressors. For actual use, the films must retain excellent characteristics after the reflow soldering process. In this study, we investigated how changing the compositions of films influences their electric resistivity and the noise suppression effects obtained after annealing at 260°C. We plated 3- μ m-thick ferrite films #1 ($\text{Ni}_{0.2}\text{Zn}_{0.1}\text{Fe}_{2.7}\text{O}_4$) and #2 ($\text{Ni}_{0.4}\text{Zn}_{0.3}\text{Fe}_{2.3}\text{O}_4$) onto polyimide sheets. The resistivity of film #1 decreased markedly, from $4 \times 10^2 \Omega \text{ cm}$ to $1 \times 10^0 \Omega \text{ cm}$ as a result of the annealing. On the other hand, film #2 exhibited a smaller decrease, from $6 \times 10^4 \Omega \text{ cm}$ to $2 \times 10^4 \Omega \text{ cm}$. The reflection parameter S_{11} and transmission loss ΔP_{loss} measured on a microstrip line for film #1, especially below 1 GHz, were increased by the annealing. This is unfavourable for use of film #1 in lowpass filters that are required to absorb noises only in the GHz range. On the other hand, S_{11} and ΔP_{loss} for film #2 were not significantly affected by the annealing. This film exhibited ΔP_{loss} of 40 % at 10 GHz. Moreover, the value of S_{11} for the film below 10 GHz was sufficiently weak, less than -12 dB. Thus, we succeeded in fabricating a GHz-range noise suppressor, with excellent heat resistance.

Key words: ferrite plating, Ni-Zn ferrite film, complex permeability, electric resistivity, noise suppressor.

1. Introduction

Current advanced downsized electronic equipments, such as Pc's and mobile phones, are currently driven at frequencies higher than several GHz. In such equipments electronic components are densely packed, which tend to cause electromagnetic interferences not only with other equipments but also with other components inside the equipments. Therefore, electromagnetic noise suppressors, which suppress high frequency noise currents generated in semiconductor devices and other components mounted on printed circuit boards, are strongly required at present. We^{1) 2)} reported that spin-sprayed NiZn ferrite films are usable for the noise suppressors working up to GHz range, because the films exhibited relatively high complex permeability, $\mu = \mu' - j\mu''$, even up to 3 GHz. The ferrite films had DC resistivity of the order of $10^4 \Omega \text{ cm}$, which is,

though, lower than that (10^6 - $10^7 \Omega \text{ cm}$) reported for bulk NiZn ferrites, but yet much higher than that ($\sim 10^{-3} \Omega \text{ cm}$) reported for metal composite films under development. Moreover, the spin-spraying is a wet process performed at low temperature ($T < 90^\circ \text{C}$) and enables us to fabricate novel two types of conducted noise suppressors, "direct deposition" type (the ferrite films are directly deposited on noise sources on printed circuit boards) and "sheet type" (the ferrite films are deposited on a flexible polymer sheet, which are cut and placed onto the noise sources). For actual use in noise suppressors, the films must keep the excellent magnetic and electric characteristics unchanged after the reflow soldering process at about 260 °C, especially when the ferrite films are embedded in multilayered printed circuit boards or electronic components. In this study we investigated how changing the compositions of films influence the electric resistivity and the noise suppression effects obtained after annealing at 260 °C.

2. Experiments

Ni-Zn ferrite films with various compositions were deposited onto polyimide sheets at 90 °C by spraying a reaction solution and an oxidising solution simultaneously. The films, as deposited and after annealed at 260 °C for 3 minutes in air, were subjected to the following measurements and analyses. The microstructures and chemical compositions were analysed by scanning electron microscopy (SEM) and inductively coupled plasma spectrometry (ICPS). In-plane DC resistivity was measured using a two-probe surface resistance meter. Complex permeability ($\mu = \mu' - j\mu''$) spectra up to 3 GHz were measured using a permeance meter equipped with a shielded loop coil³⁾. To evaluate the noise suppression characteristics the samples were cut into 35 mm x 35 mm pieces, which were placed onto a 50 Ω microstrip line of 70 mm length, as described in our previous paper⁴⁾. Measuring reflection parameter S_{11} and transmission parameter S_{21} from 10 MHz to 10 GHz with a network analyzer, we calculated transmission loss ΔP_{loss} according to the formulas given in ref.¹⁾. For comparison, measurements were also done on a commercialised, composite type noise suppressor (25 μ m thickness) in

which ferromagnetic metal flakes are embedded in a polymer matrix⁵⁾.

3. Results and discussion

The films #1 and #2 had compositions $\text{Ni}_{0.2}\text{Zn}_{0.1}\text{Fe}_{2.7}\text{O}_4$ and $\text{Ni}_{0.4}\text{Zn}_{0.3}\text{Fe}_{2.3}\text{O}_4$, respectively. As shown in Fig.1, the both films had thicknesses of about 3 μm . The film #1 had a relatively thick (ca, several hundred nanometers) columnar structure perpendicular to film plane. On the other hand, the film #2 had no columnar structure, but was composed of large (reaching several hundred nanometers) grains, having a rough surface. Thus the film #2 exhibited a weaker adhesive power against substrate than the film #1.

The resistivity for the film #1 decreased remarkably from $4 \times 10^2 \Omega\text{cm}$ to $1 \times 10^0 \Omega\text{cm}$ by the annealing. On the other hand, the film #2 exhibited a small decrease from $6 \times 10^4 \Omega\text{cm}$ to $2 \times 10^4 \Omega\text{cm}$. Electric conduction in ferrite materials is given by transfer of electrons between Fe^{2+} and Fe^{3+} ($\text{Fe}^{3+} + e^- = \text{Fe}^{2+}$) on the octahedral sites (B-sites) of spinel structure, which is enhanced as concentration of Fe^{2+} increases. Therefore, we can consider that the film #1, which was low in $\text{Ni}^{2+} + \text{Zn}^{2+}$ concentration and thus high in Fe^{2+} concentration (nominal composition: $\text{Ni}^{2+}_{0.2}\text{Zn}^{2+}_{0.1}\text{Fe}^{2+}_{0.7}\text{Fe}^{3+}_{2.0}\text{O}_4$), exhibited lower resistivity than the film #2 which was low in Fe^{2+} concentration ($\text{Ni}^{2+}_{0.4}\text{Zn}^{2+}_{0.3}\text{Fe}^{2+}_{0.3}\text{Fe}^{3+}_{2.0}\text{O}_4$).

(a) Film #1 ($\text{Ni}_{0.2}\text{Zn}_{0.1}\text{Fe}_{2.7}\text{O}_4$)

(b) Film #2 ($\text{Ni}_{0.4}\text{Zn}_{0.3}\text{Fe}_{2.3}\text{O}_4$)

Fig. 1 Cross-sectional SEM images for films #1 and #2.

As shown in Fig. 2, μ' in the lower frequency range for both films decreased by the annealing, while the natural resonance frequency f_r , where μ'' becomes maximum, shifted to the higher frequencies, from 450

Fig. 2 Permeability spectra for films #1 and #2.

Fig. 3 S-parameters and ΔP_{loss} for film #1 and a composite sheet.

MHz to 650 MHz for the film #1 and from 550 MHz to 750 MHz for the film #2. The change of the permeability spectra by the annealing may be ascribed to the increase in

uniaxial anisotropy field H_k , because μ' and f_r for the magnetic films (where spins are confined to film plane) are expressed by the following equations based on two dimensional spin dynamics ⁶⁾:

$$\mu' = \frac{4\pi M_s}{H_k} \quad (1)$$

$$f_r = \frac{\gamma}{2\pi} \sqrt{H_k \cdot 4\pi M_s} \quad (2)$$

Here M_s , and γ represent saturation magnetization and gyromagnetic ratio. Since we observed no remarkable change in microstructure of both films by the annealing, we guess that the increase of H_k as well as the decrease of resistivity by the annealing are caused by the change in valence or distribution of cations. We also guess that the lower permeability of the film #2 is ascribed to the lower M_s of 470 emu/cc than that of the film #1 of 220 emu/cc, mainly due to the deterioration of microstructure shown in

Fig. 4 S-parameters and ΔP_{loss} for film #2 and a composite sheet.

Fig.1. In order to improve the permeability as well as the adhesive power against substrate, modifying the morphology of grains, by changing the plating conditions, will be necessary.

As shown in Fig.3, S_{11} for the film #1, especially at below 1 GHz, increased by the annealing, while S_{21} decreased. ΔP_{loss} for the film #1 increased by the annealing throughout the measurement frequency range, which is reflecting the changes occurred in S_{11} and S_{21} . ΔP_{loss} for the film #1 after the annealing was much stronger than that for the composite sheet of 25 μ m thickness. However, S_{11} for the film #1 after the annealing was higher than that for the composite sheet below 300 MHz, and moreover, ΔP_{loss} was relatively high even at below 1 GHz. This is unfavourable for the use as lowpass filters that are required to absorb noises only in the GHz range. As shown in Fig. 4, on the other hand, S_{11} and ΔP_{loss} for the film #2 did not change much by the annealing, though ΔP_{loss} was lower than that for the film #1 due to the lower μ'' . The reflection parameter S_{11} for the film #2 was less than -12 dB below 10 GHz, being much lower than that reported for the composite sheet. Moreover, ΔP_{loss} for the film #2 at 10 GHz was 40 %, which is as strong as that reported for the composite sheet. This means that the film #2 of 3 μ m thickness exhibits not only excellent heat resistance but also strong noise suppression. Since the film #1 is eight times thinner than the composite sheet, our film exhibits noise suppression eight times stronger than the composite sheet.

Fig. 5 Resistivity dependence of ΔP_{loss} (shown by circles) and μ'' (shown by squares) at (a) 100 MHz and (b) 3 GHz, obtained for various annealed and as-deposited films, including #1 and #2.

We can consider that the increase of ΔP_{loss} by the annealing for the film #1 is ascribed to the remarkable decrease of resistivity, because, as shown in Fig.5(a), ΔP_{loss} for various annealed films (prepared in addition to #1 and #2) decreased with increasing resistivity at 100 MHz where μ'' was negligibly small. One possible origin of the resistivity-related power loss described above is in-plane eddy current loss caused by the incident magnetic flux perpendicular to the film plane. We can also consider that ΔP_{loss} in GHz range for the as-deposited films (with resistivity higher the $10^2 \Omega\text{cm}$) is mainly ascribed to μ'' , because, as shown in Fig.5(b), the sample having higher μ'' tends to exhibit stronger ΔP_{loss} at 3 GHz (the upper frequency limit of our permeability measurements).

4. Conclusions

We successfully fabricated thin film GHz noise suppressor of 3 μm thickness from aqueous solution, having an excellent heat resistance. The films will keep the excellent magnetic and electric characteristics unchanged even after reflow soldering process at about 260 $^{\circ}\text{C}$, which enable us to realize electromagnetic noise suppressors which are embedded in printed circuit boards and electronic parts. Modifying the morphology of grains, by changing the plating conditions, is the issue to be addressed, which may enhance the noise suppression performance further and improve the adhesion power against substrates.

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