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Variation of the Relaxation Time for NiCuZn Ferrites with Magnetic Properties

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The frequency dependence of complex permeability for various NiCuZn ferrites was investigated. The variation of complex permeability for NiCuZn ferrites can be presented as a form of a semi-circle, so called the Cole-Cole plot, and the relaxation phenomena were explained with various shapes of the plots. The relaxation time τ was calculated from f_{rx}, which is a relaxation frequency at μ''_{max}. Relations between anisotropy field H_A and relaxation time τ, initial permeability μ_i and H_A were plotted to identify the frequency dependence of complex permeability.

1. Introduction

The permeability of ferrimagnetic materials, such as ferrites that exhibit Snoek's limit, can be affected by composition and processing. The real part of permeability μ' decreases with the frequency and the imaginary part μ'' exhibits a broad peak, which is related with the relaxation phenomena. As the frequency of rf excitation increases, the material reaches a point beyond which the spins cannot fully respond to the excitation [1]. The magnetization no longer moves in phase with the excitation and losses occur. The μ' decreases with increasing the frequency and the μ'' goes through a broad resonance. The phase difference between the applied field and magnetization of the ferrite occurs due to the damping phenomena. If there is no damping in magnetization process, μ'' is zero for all frequencies, except at the resonance frequency, and then μ'' becomes infinitely large [2-4]. The broadening of the μ'' curves at higher frequencies is due to the fact that the alternating measuring field possesses a component parallel to the domain walls and also perpendicular to the saturation magnetization [4]. Magnetic relaxations appear as a decrease of μ' with increasing frequency, while μ'' has a maximum near the relaxation frequency f_{rx}.

In characterization of frequency dependence of spinel ferrite materials, the dispersion phenomena due to relaxation can be observed at some frequency range, and this is related with the ferrite composition and their initial permeability. Therefore it is important to investigate the variation of complex permeability with frequency for the applications, which gives useful information for application in the high-frequency range.

In this work, the Cole-Cole plots for NiCuZn ferrites were presented to describe its relaxation process with μ' versus μ'' spectrum. It was found that the relaxation process with a semicircle curve of complex permeability is characterized by the magnitude of initial permeability. The purpose of the present work was to investigate the frequency dependence of complex permeability for various NiCuZn ferrites and especially to discuss its relaxation phenomena with the relation of μ' versus μ''.

2. Experimental

NiCuZn ferrites were prepared by conventional ceramic processes with each metal oxides as starting materials and Co_3O_4 as additive. The mixture of raw materials were pulverized in a ball-mill for 40 hrs and calcined at 750 ℃ for 2 hrs to obtain spinel phase. The calcined powder was formed in a toroidal green body and the toroidal sample was fired at 900 ℃ for 5 hrs. The initial permeability was calculated from the measured inductances at 100 kHz and the complex permeability at 1 ~ 100 MHz using HP 4195A Network Analyzer. The magnetic flux density was measured by B-H Loop Tracer AMH5020. The anisotropy field H_A is calculated from the measured parameters.

3. Results and Discussion

The NiCuZn ferrite compositions in this study were developed on the basis of common guidelines, and then classified as the following categories : (i) the NiCuZn ferrite can be sintered at a lower temperature than NiZn ferrite, (ii) permeability depends on the Zn content, (iii) a composition deficient in Fe_2O_3 has good sintering properties, (iv) the addition of cobalt is effective to increase the resonance frequency.

For these NiCuZn ferrites, the following characterizations are reported here as the experimental results which enables us to describe the relaxation phenomena from the frequency dependence of complex permeability. The relations between relaxation time τ and initial permeability μ_i, anisotropy field H_A are also described to characterize the properties of NiCuZn ferrites.

3-1. Relaxation phenomena of the NiCuZn ferrites

As were the previous works [5, 6], the role of copper in NiCuZn ferrite is to make the green body sintered at relatively low temperature and to improve the sintering property. The magnitude of initial permeability for these ferrites is affected by the content of copper.

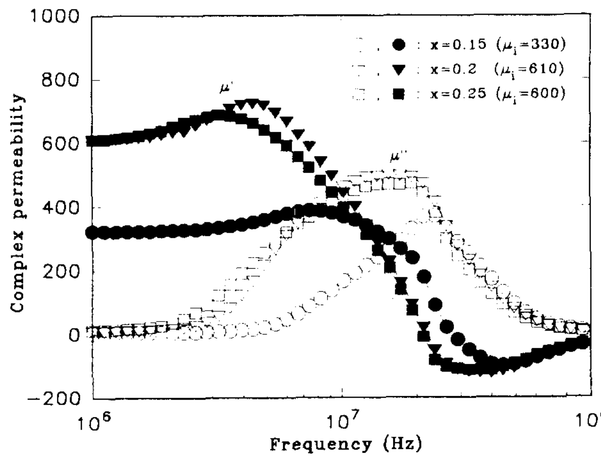

Fig. 1 Complex permeability for $(Ni_{0.4-x}Cu_xZn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$ sintered at 900 ℃ for 5 hrs.

Fig. 1 shows the variation of complex permeability of $(Ni_{0.4-x}Cu_xZn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$ ($x = 0.15 \sim 0.25$) as a function of frequency. But the curves for μ'' of higher x values such as x = 0.2, x = 0.25 forms a peak at a lower frequency range than the case of x = 0.15. This means that the high-frequency loss increases with increasing the copper content.

Fig. 2 is the Cole-Cole plot for the curves of complex permeability in Fig. 1. The Cole-Cole plots of μ' versus μ'' shows some semi-circle shaped curves, which means the relaxation dispersion. But there are some differences in the shape of curves comparing to the normalized curve of complex permeability [4]. The NiCuZn ferrites or other spinel ferrites in applications show relaxation phenomena as increasing the frequency owing to the damping in magnetization process. Fig. 2 describes the relation between the magnitude of initial permeability and relaxation of $(Ni_{0.4-x}Cu_xZn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$ ($x = 0.15 \sim 0.25$) by introducing the Cole-Cole plot of the complex permeability. The Cole-Cole plot for x = 0.15 shows an small ellipsoidal curve as shown in Fig. 2, which gives the narrower peak of complex permeability than that of other two composition, x = 0.2 and x = 0.25. This provides another method of representation to identify the relaxation phenomena for various ferrites.

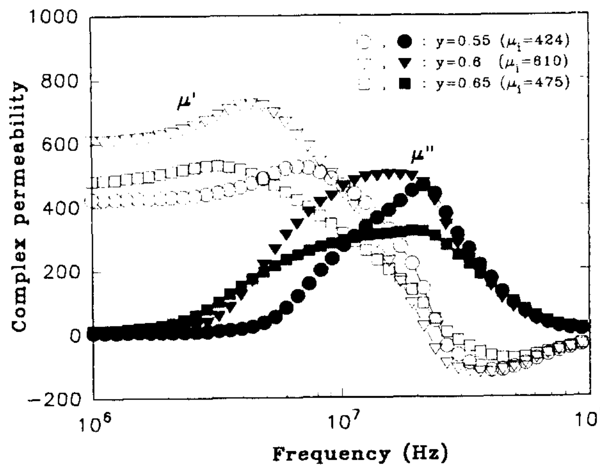

Fig. 3 Complex permeability of $(Ni_{0.8-y}Cu_{0.2}Zn_yO)_{1.015}(Fe_2O_3)_{0.985}$ sintered at 900 ℃ for 5 hrs.

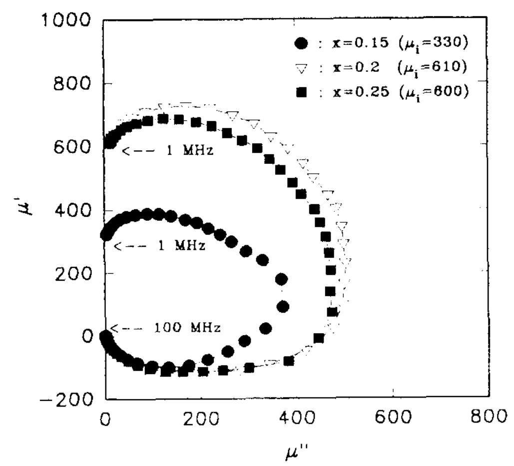

Fig. 2 Cole-Cole plots for $(Ni_{0.4-x}Cu_xZn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$ sintered at 900 ℃ for 5 hrs.

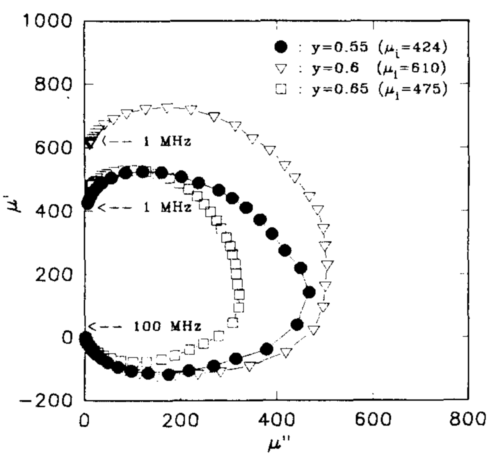

Fig. 4 Cole-Cole plots for $(Ni_{0.8-y}Cu_{0.2}Zn_yO)_{1.015}(Fe_2O_3)_{0.985}$ sintered at 900 ℃ for 5 hrs.

Fig. 3 and Fig. 4 show the relation between μ' and μ'' for $(Ni_{0.8-y}Cu_{0.2}Zn_{0.2}O)_{1.015}(Fe_2O_3)_{0.985}$ ($y = 0.15 \sim 0.25$) ferrites. In general, the saturation magnetization increases linearly with the zinc content until all divalent magnetic ions are replaced by zinc ions [4, 7]. For the larger zinc content, the saturation magnetization decreases due to the exchange interaction. The difference in magnitude of μ_i, which means a variation of ferrite composition in this study, represents the various Cole-Cole plots of μ' versus μ'' with frequency.

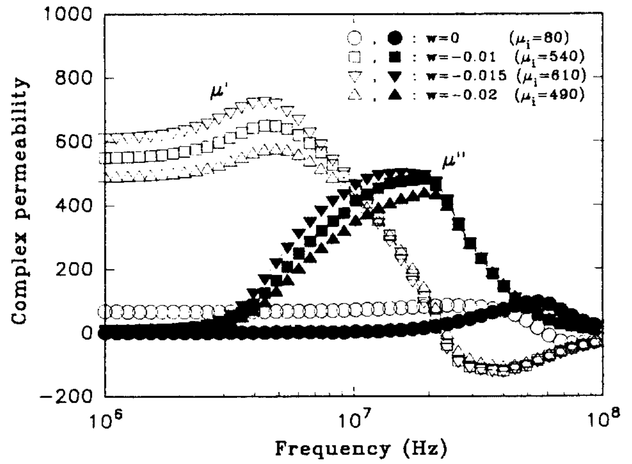

Fig. 5 Complex permeability for $(Ni_{0.2}Cu_{0.2}Zn_{0.6}O)_{1-w}(Fe_2O_3)_{1+w}$ sintered at 900 ℃ for 5 hrs.

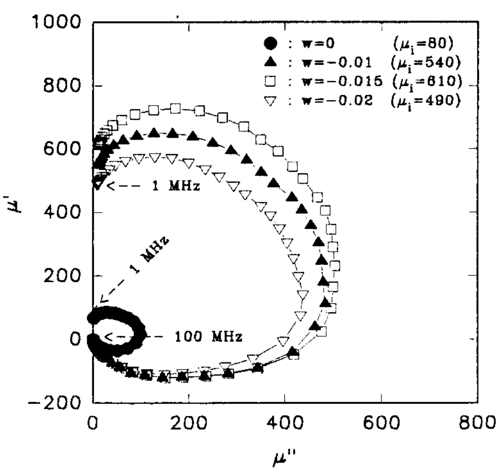

Fig. 6 Cole-Cole plots for $(Ni_{0.2}Cu_{0.2}Zn_{0.6}O)_{1-w}(Fe_2O_3)_{1+w}$ sintered at 900 ℃ for 5 hrs.

Fig. 5 and Fig. 6 show the results for $(Ni_{0.2}Cu_{0.2}Zn_{0.6}O)_{1-w}(Fe_2O_3)_{1+w}$ ($w = 0 \sim -0.02$) ferrites. The sintered NiCuZn ferrite of $w = 0$ have a low initial permeability because the composition of Fe_2O_3 is not so deficient as having the high density after firing at a low temperature [8, 9]. But the relation between μ' and μ'' as a function of frequency which means its relaxation process for these Fe_2O_3 deficient NiCuZn ferrite seems to be insensitive to the Fe_2O_3 deficiency (w). It is shown as in Fig. 5 that the f_{rx} value moves slightly to a lower range until the

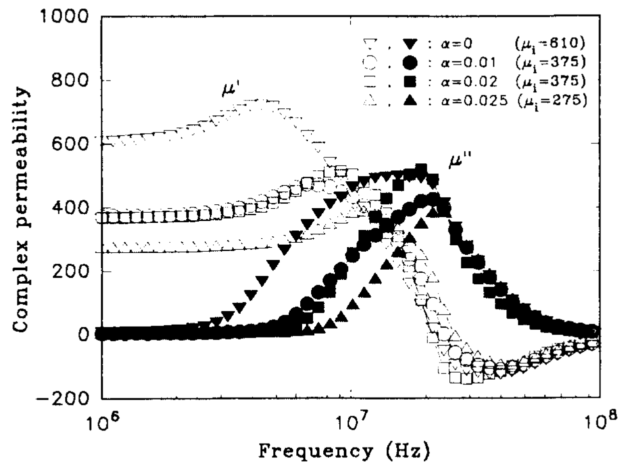

Fig. 7 Complex permeability for $(Ni_{0.2-x}Co_xCu_{0.2}Zn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$ sintered at 900 ℃ for 5 hrs.

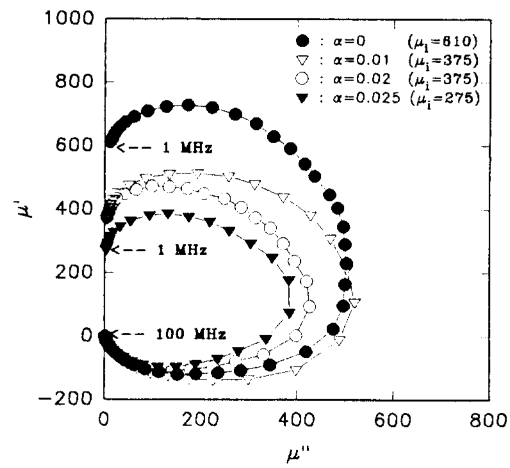

Fig. 8 Cole-Cole plots for $(Ni_{0.2-x}Co_xCu_{0.2}Zn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$ sintered at 900 ℃ for 5 hrs.

Fe_2O_3 deficiency (w) reaches at $w = -0.015$, but the higher f_{rx} is obtained at $w = -0.02$. It can be seen that the f_{rx} of Fe_2O_3 deficient NiCuZn ferrite shows a little change for the various w values because the control of each Fe_2O_3 deficiency (w) is an important factor to improve the sintering properties [8].

The data for complex permeabilities and Cole-Cole plots for $(Ni_{0.2-\alpha}Co_\alpha Cu_{0.2}Zn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$ ($\alpha = 0 \sim 0.025$) are shown in Fig. 7, Fig. 8. It is shown that the f_{rx} of cobalt-contained NiCuZn ferrites becomes higher than that of some cobalt-free ferrites. As plotted in Fig. 7, the f_{rx} for the larger cobalt content (α) shift to higher frequency range. The substitution of cobalt ions in NiCuZn ferrites leads to an increase of f_{rx}. This improvement is attributed to domain wall stabilization and reduces the high-frequency losses, which is related with the increasing of magnetic induced anisotropy by substitution of cobalt ions [7, 10]. The relation between μ' and μ'' for variable frequencies in

Fig. 8 represents the variation of relaxation process as increasing the cobalt content.

3-2. Characterization of the relaxation time τ and the anisotropy field H_A

The relaxation time τ is the time elapsed after the application of a field H before the magnetization differs by less than about 37 % from its equilibrium value [4]. The relaxation time τ is given by:

$$\tau = \frac{1}{2\pi f_{rx}} \tag{1}$$

For a polycrystalline specimen consisting of randomly oriented crystallites without any magnetic interaction, the average value of the initial permeability, μ_i, is given by the following equation:

$$\frac{\mu_i - 1}{4\pi} = \frac{1}{3}\left(\frac{M_s}{H_{A1}} + \frac{M_s}{H_{A2}}\right) \tag{2}$$

where M_s is the saturation magnetization, H_{A1} and H_{A2} are two different anisotropy fields. For the ferrimagnetic spinels having the c axis as the preferred direction of magnetization, we may write $H_{A1} = H_{A2} = H_A$ and Eq. (2) can be expressed as Eq. (3):

$$\frac{\mu_i - 1}{4\pi} = \frac{2}{3}\left(\frac{M_s}{H_A}\right) \tag{3}$$

We can calculate the resonance frequency, which is determined by the magnitude of the saturation magnetization, by Snoek's relation [7] as follows:

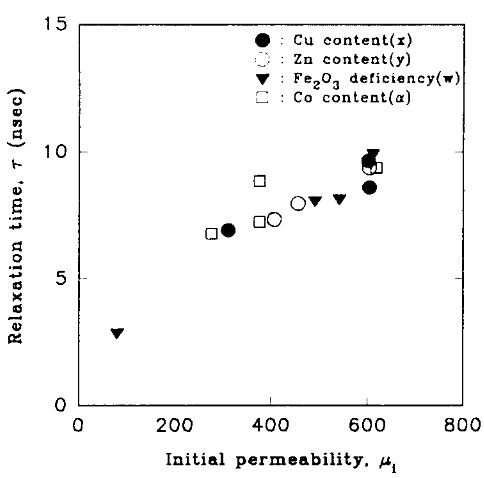

Fig. 9 Initial permeability vs. relaxation time for various NiCuZn ferrites; $(Ni_{0.4-x}Cu_xZn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$[●], $(Ni_{0.8-y}Cu_{0.2}Zn_yO)_{1.015}(Fe_2O_3)_{0.985}$[○], $(Ni_{0.2}Cu_{0.2}Zn_{0.6}O)_{1-w}(Fe_2O_3)_{1+w}$[▼], $(Ni_{0.2-z}Co_z Cu_{0.2}Zn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$[□].

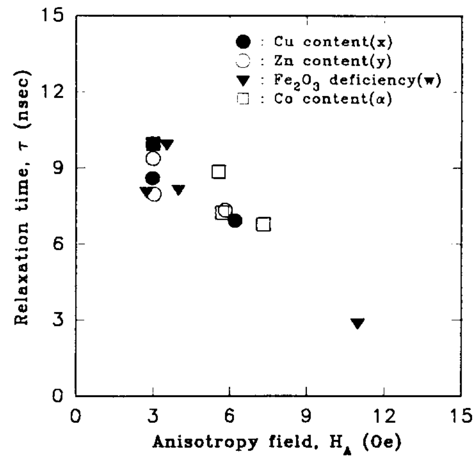

Fig. 10 Anisotropy field vs. relaxation time for various NiCuZn ferrites; $(Ni_{0.4-x}Cu_xZn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$[●], $(Ni_{0.8-y}Cu_{0.2}Zn_yO)_{1.015}(Fe_2O_3)_{0.985}$[○], $(Ni_{0.2}Cu_{0.2}Zn_{0.6}O)_{1-w}(Fe_2O_3)_{1+w}$[▼], $(Ni_{0.2-z}Co_z Cu_{0.2}Zn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$[□].

$$f_{res}(\mu_{rot} - 1) = \frac{4}{3}\gamma M_s \tag{4}$$

where f_{res} is the ferromagnetic resonance frequency, μ_{rot} is the rotational permeability, and γ is the gyromagnetic ratio ($= 1.7588 \times 10^7\ Oe^{-1} \cdot s^{-1}$).

Fig. 9 shows the relation between initial permeability and relaxation time, which was calculated from the Eq. (1) for the NiCuZn ferrites. It is found that the relaxation time is nearly proportional to the initial permeability. This means that the magnitude of initial permeability is crucial to classify the relaxation process in application of NiCuZn ferrites at high frequencies. The relation between relaxation time and anisotropy field is shown in Fig. 10 as an description for frequency dependence of NiCuZn ferrites. Since the initial permeability becomes smaller, at the same time the coercive force also becomes larger [4], so that the anisotropy field increases as decreasing the initial permeability. As H_A increases, the peak width of μ'' curve becomes narrower, and one can find the frequency of maximum μ'' at higher frequencies.

4. Conclusion

The Cole-Cole plots of complex permeability give an example of the way to characterize the NiCuZn ferrites by introducing the relaxation process through the relation between μ' and μ'' for various frequencies. The relaxation process depends on initial permeability which is related with the anisotropy field. From the results of plots for frequency dependence of the complex permeability, the composition of NiCuZn ferrite in the actual applications can be effectively classified. These results apply well to NiCuZn ferrites in this work, and then some properties are summarized in Table I.

Table I Relation between the initial permeability μ_i and some properties for various NiCuZn ferrites with spinel structure; measured frequency $f_{\mu'' max}$, maximum magnetic flux density B_m, calculated anisotropy field H_A, relaxation time τ, calculated resonance frequency f_r.

Composition	μ_i	$f_{\mu'' max}$ [MHz] (measured)	$4\pi M_s$ [G]	H_A [Oe] (calculated)	τ [nsec]	f_r [MHz] (calculated)
$(Ni_{0.4-x}Cu_xZn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$, x = 0.15	330	23.0	3060	6.20	6.92	17.4
x = 0.20	610	17.0	3070	2.97	9.95	8.3
x = 0.25	600	18.5	3050	2.95	8.60	8.3
$(Ni_{0.8-y}Cu_{0.2}Zn_yO)_{1.015}(Fe_2O_3)_{0.985}$, y = 0.55	424	21.7	3680	5.80	7.33	16.2
y = 0.6	610	17.0	3070	2.97	9.95	8.3
y = 0.65	475	20.0	2150	3.02	7.96	8.5
$(Ni_{0.2}Cu_{0.2}Zn_{0.6}O)_{1-w}(Fe_2O_3)_{1+w}$, w = 0	80	55.0	1300	10.97	2.89	30.7
w = −0.01	540	19.7	2000	2.73	8.08	7.6
w = −0.015	610	19.5	3200	3.96	8.16	11.1
w = −0.02	490	16.0	3200	3.50	9.95	9.8
$(Ni_{0.2-\alpha}Co_\alpha Cu_{0.2}Zn_{0.6}O)_{1.015}(Fe_2O_3)_{0.985}$, $\alpha = 0$	610	17.0	3070	2.97	9.95	8.3
$\alpha = 0.01$	375	22.0	3190	5.69	7.23	15.9
$\alpha = 0.02$	375	18.0	3110	5.54	8.84	15.5
$\alpha = 0.025$	275	23.5	3000	7.30	6.77	20.4

Acknowledgement

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