Relationship between magnetic properties and microwave-absorbing characteristics of NiZnCo ferrite composites

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The complex permeability and permittivity of the constituent materials of the microwave absorbers play a key role in determining the reflection or attenuation properties of the incident microwave. The real and imaginary (loss) components of μ_r and ε_r in the frequency range 1–12 GHz, and their relationship with microwave-absorbing properties, have been investigated in the NiZnCo ferrite composites. The composite specimens were prepared by moulding and curing a mixture of prereacted ferrite powder and silicone rubber. The quantitative estimation of microwave-absorbing properties was made by plotting the observed material constants (μ_r, ε_r) on the calculated solution map of impedance-matching. It was found that the CoO content influences the first matching frequency and the first matching thickness in the NiZnCo ferrite composites, but does not influence the second matching frequency and second matching thickness.

1. Introduction

The ferrites, which absorb energy from electromagnetic waves and are widely used in the stealth technology of aircraft, TV image interference of high-rise buildings, and microwave dark-room and protection, have attracted much attention [1]. Extensive study has been carried out to develop microwave-absorption materials with high efficiency and new absorption materials [2, 3].

One criterion for selecting the absorbing material is the location of its natural resonance region. Thus, a study of the frequency dependency of the complex permeability of the ferrite has been a field of interest.

The spinel ferrites have been utilized for many years as absorbers of various forms, such as absorbing rubber materials, sintered ceramic tiles, or paints [4–6]. The peaks in the loss spectrum in Ni–Zn ferrites are due to spin resonances and can be changed in frequency by substituting a portion of nickel or zinc ions by divalent metal ions such as cobalt. Some attempts were also made to verify the correlation between the material constants, μ_r and ε_r, and microwave absorption in the sintered ferrites containing CoO.

The purpose of the present work was to investigate the relationship between the magnetic characteristics of NiZnCo ferrite composites and their microwave-absorbing properties in the frequency range 1–12 GHz. The quantitative estimation of microwave-absorbing properties was made by plotting the observed material constants on the calculated solution map of impedance matching.

2. Experimental procedure

The ferrites were prepared by conventional solid-state reaction method. The chemical formula of the materials is $Ni_{0.5-x}Zn_{0.5-x}Co_{2x}Fe_2O_4$ ($x = 0$, 0.05, 0.1, 0.15, 0.2). Silicone rubber was used as a matrix material. The composite specimens were prepared by moulding and curing the mixture of prereacted ferrite powder and silicone rubber. The loading factor of ferrite was about 80 wt %. Toroidal samples of 3.04 mm i.d., 7.0 mm o.d., and 2 mm thick were machined for permeability spectra measurements.

The complex permeability and permittivity were determined using an HP/8720B network analyser. Measurements were made in the frequency range 1–12 GHz. The scattering function, S_{11} and S_{21}, corresponding to the reflection and transmission, of a TEM (transverse electric and magnetic wave) were measured on the surface of the sample in a coaxial line.

The complex permeability and permittivity were calculated from the measurements in the case of a short circuit and of an open circuit [7]. The matching condition was calculated by variation of the thickness of the test specimens.

3. Results and discussion

To select the basic composition from the NiZn ferrite system, measurements of the material constants were carried out. $Ni_{0.5}Zn_{0.5}Fe_2O_4$ (Ni/Zn = 1.0) was chosen as a basic composition, showing a high μ_r'' value. This results agree with Hahn *et al.*'s study [8].

While maintaining the Ni/Zn ratio = 1.0, CoO was stoichiometrically added to the basic composition to investigate the effect of occupancy of the octahedral sites in the spinel on the magnetic anisotropy. Fig. 1 shows the complex permeability ($\mu_r = \mu_r' - j\mu_r''$) spectra observed in the NiZnCo ferrite composites at

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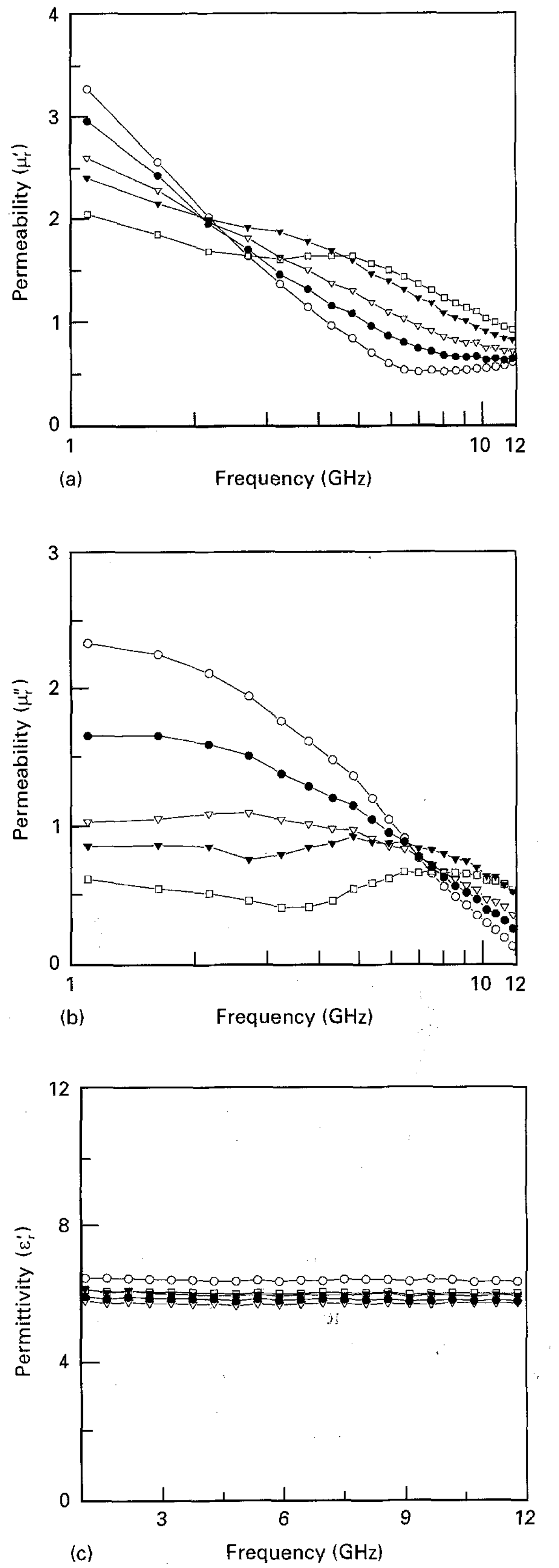


Figure 1 Material constants (μ_r, ε_r) versus frequency spectra of the $Ni_{0.5-x}Zn_{0.5-x}Co_{2x}Fe_2O_4$ composites. (a) μ_r', (b) μ_r'', (c) ε_r'. x: (○) 0, (●) 0.05, (▽) 0.1, (▼) 0.15, (□) 0.2.

1–12 GHz. In these composite specimens, μ_r' decreases sharply as a function of frequency. The value of μ_r' increases in the frequency region (>2 GHz) with increasing CoO content.

The maximum value of μ_r'' shifts to high frequency with increasing CoO content. The value of μ_r'' is aug-

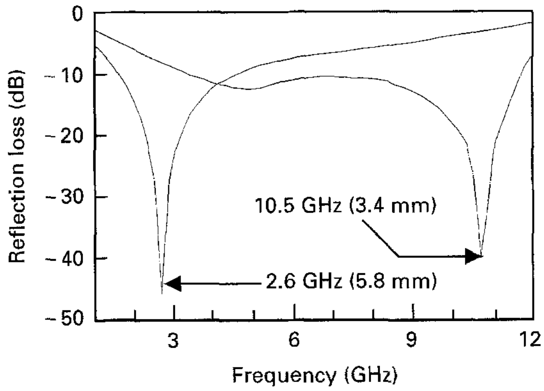


Figure 2 Absorption characteristics of the $Ni_{0.5}Zn_{0.5}Fe_2O_4$ composite with maximum attenuation.

mented with increasing CoO content, especially in the high-frequency region (> 8 GHz).

The complex permittivity ($\varepsilon_r = \varepsilon_r' - j\varepsilon_r''$) of the NiZnCo ferrite composites are almost constant ($\varepsilon_r' = 6$ and $\varepsilon_r'' = 0.12$ in the frequency range 1–12 GHz). This is likely to be due to the intrinsically small dielectric loss tangent (tan δ_ε) of the ferrite powder used in this study. The average value of tan δ_ε is approximately 0.02 and nearly constant in the frequency range of 1–12 GHz.

Fig. 2 shows the absorption behaviour for the rubber-bonded Ni–Zn ferrite ($Ni_{0.5}Zn_{0.5}Fe_2O_4$) composite. The curves were calculated from the complex permeability and the complex permittivity.

For a microwave-absorbing layer terminated by a short circuit, the normalized input impedance at the absorber surface, Z_{in}, is given by

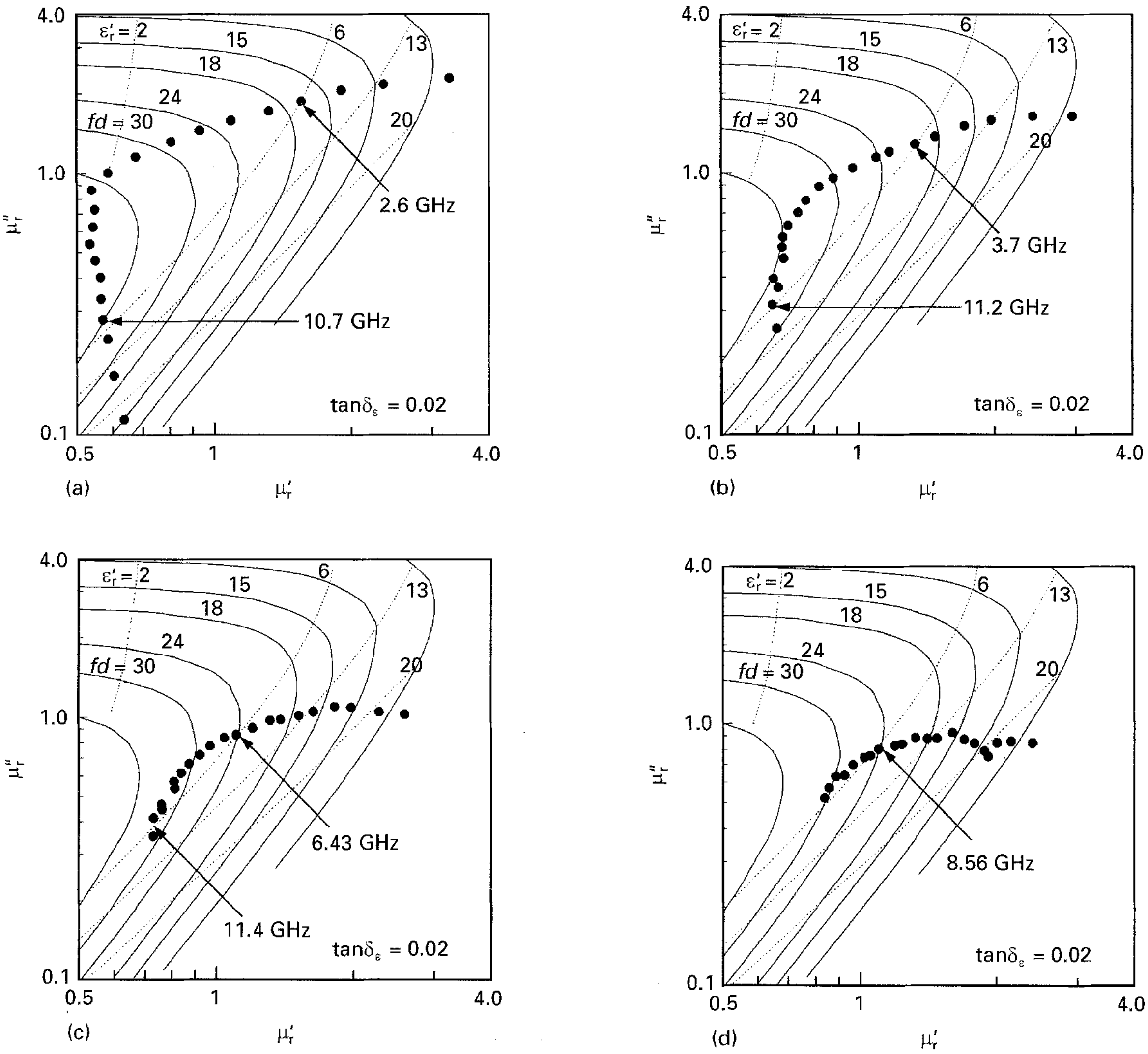
$$Z_{in} = (\mu_r/\varepsilon_r)^{1/2} \tanh[j(2\pi d/\lambda)(\mu_r\varepsilon_r)^{1/2}] \quad (1)$$

where λ is the wavelength of the microwave in free space and d is the thickness of an absorber.

The impedance matching solution map shows the relationship between the six parameters (frequency, absorber thickness, and the real and imaginary components of μ_r or ε_r), from which the zero-reflection condition can be derived from Equation 1.

For NiZnCo ferrite composites, the locus of $\mu_r' - \mu_r''$ was replotted in the impedance matching solution map as shown in Fig. 3. In Fig. 3(a), the crossing points of the $\mu_r' - \mu_r''$ locus and the $\varepsilon_r' = 6.0$ line correspond to the impedance matching combination of the parameter groups. The frequency at this point (the matching frequency, f_m) is equal to 2.6 GHz in the low-frequency region and to 10.7 GHz in the high-frequency region. The values of the product (frequency × thickness $= f \times d$) in low- and high-frequency regions are 15 and 36.38 [GHz × mm], respectively. Therefore, the values of the matching thickness, d_m, are 5.8 and 3.7 mm, respectively. This prediction of f_m and d_m is in good agreement with the simulation results of reflection loss, as shown in Fig. 2.

When the two crossing points of the $\mu_r' - \mu_r''$ locus and the ε_r' line exist, the matching frequency and the thickness in the relatively lower frequency region are f_{m1} (the first matching frequency) and d_{m1} (the first matching thickness), respectively. The matching



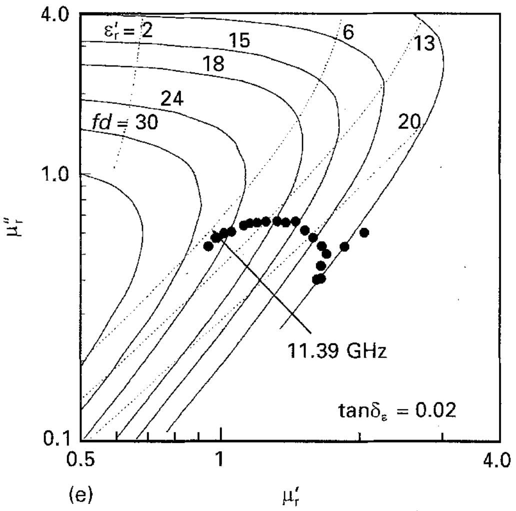


Figure 3 Impedance matching solution map for zero reflection of $Ni_{0.5-x}Zn_{0.5-x}Co_{2x}Fe_2O_4$ composites. *x*: (a) 0, (b) 0.05, (c) 0.1, (d) 0.15, (e) 0.2.

TABLE I The relationship between matching frequency and matching thickness in $Ni_{0.5-x}Zn_{0.5-x}Co_{2x}Fe_2O_4$ specimens

x	f_{m1} (GHz)	d_{m1} (mm)	f_{m2} (GHz)	d_{m2} (mm)
0	2.6	5.8	10.7	3.4
0.05	3.7	5.4	11.2	3.2
0.1	6.43	3.9	11.4	2.9
0.15	8.56	3.0	–	–
0.2	11.39	2.3	–	–

frequency and the thickness in the relatively higher frequency region are f_{m2} (the second matching frequency) and d_{m2} (the second matching thickness), respectively. The results of f_m and d_m with the variation of CoO contents are summarized in Table I.

Fig. 4 shows the matching frequency and thickness from the observed material constants of NiZnCo ferrite composites as a function of CoO content. The first matching frequency shifts towards high frequency with increasing CoO content.

The first matching frequency shows a clear dependence on the chemical composition, x, of $Ni_{0.5-x}Zn_{0.5-x}Co_{2x}Fe_2O_4$. The second matching frequency remains nearly constant with increased CoO content. It is observed from Fig. 4(b) that the second matching thickness is nearly constant, but the first matching thickness is remarkably reduced with variation in the CoO content. It is apparent that the CoO content influences f_{m1} and d_{m1} in the NiZnCo ferrite composites, but does not influence f_{m2} and d_{m2}.

In order to study these phenomena in view of the magnetic properties, the coercive force, H_c, of the ferrite powders was measured. Fig. 5 shows the

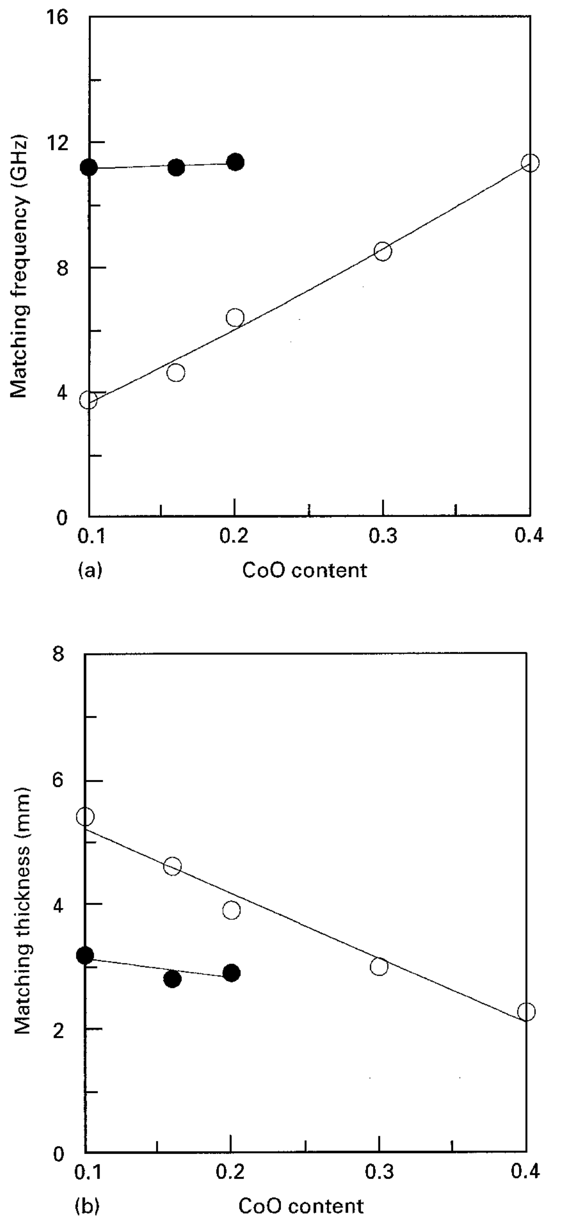


Figure 4 Relationship between CoO content and (a) matching frequency and (b) matching thickness in the $Ni_{0.5-x}Zn_{0.5-x}Co_{2x}Fe_2O_4$ composites. (a) (○) f_{m1}, (●) f_{m2}, (b) (○) d_{m1}, (●) d_{m2}.

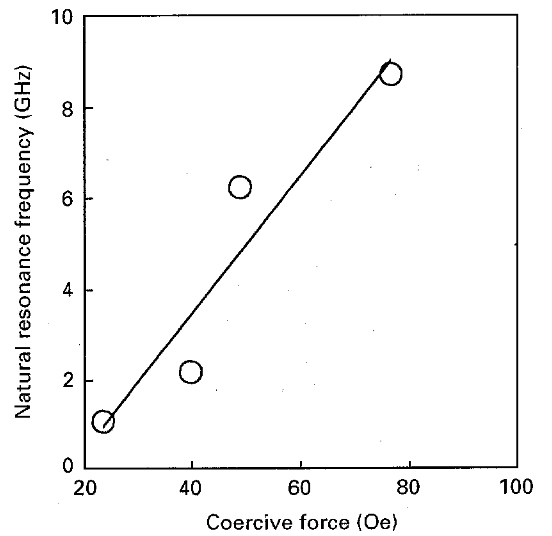


Figure 5 The natural resonance frequency as a function of coercive force, H_c.

relationship between the natural resonance frequency and the coercive force of ferrite composites. The natural resonance frequency, f_r, was determined graphically from the frequency at the maximum μ_r''. The proportional characteristics between the natural resonance frequency and the coercive force are observed in Fig. 5.

A number of authors have previously investigated the relationship between coercive force and magneto crystalline anisotropy and saturation magnetization [9, 10]. H_c is a function of many factors, such as anisotropy constant, κ, and saturation magnetization, σ_s, etc. Among them, κ and σ_s are the dominant factors determining coercive force in the spinel ferrites. With increased CoO content, σ_s is within the range of 74–84 emu g^{-1} and H_c increased remarkably from 1830 A m^{-1} to 6048 A m^{-1}. This fact suggests that the remarkable variation of coercive force caused by the cobalt substitution influences the resonance frequency. It could be deduced that the increase in H_c results from the increase of κ due to the variation of CoO content. Therefore, it is concluded that the first matching frequency is controlled by the variation of CoO content.

4. Conclusion

The natural resonance frequency shifts to a higher frequency with increasing in CoO contents. Examination of the relationship between coercive force, H_c, and saturation magnetization, σ_s, and the crystalline anisotropy constant, κ, leads us to confirm that the shift of the natural resonance frequency may be attributed to the increase in H_c with the variation of the CoO content. The quantitative estimation of microwave-absorbing properties was made by plotting the observed material constants on the calculated solution map of impedance matching. The variation of CoO contents in the NiZnCo ferrite composites gives insight into the relationship between matching frequency and magnetic parameters.

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