THE DISPERSION CHARACTERISTICS OF THE COMPLEX PERMEABILITY OF NiZnCu FERRITE AND ITS COMPOSITE MATERIALS

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Summary The paper deals with the frequency dependence of complex permeability for the $Ni_{0.27}Zn_{0.63}Cu_{0.1}Fe_2O_4$ sintered ferrite produced by conventional ceramic method and its composite materials made of this ferrite (in the form of powder) and non–magnetic polymer matrix (polyvinyl chloride – PVC). The experimental and theoretical curves of the complex permeability were compared. The contribution of both domain wall motion and spin rotational magnetisation processes on the complex permeability in high–frequency region (up to 1 GHz) is discussed. In the case of composite materials, the simple magnetic circuit approach is taken into account. In composites, the real part of the complex permeability in the frequency region above 100 MHz is larger than that of the prepared sintered ferrite. This is attributed to the shift of the spin and domain wall resonance frequencies toward the higher frequency region by introducing demagnetising fields of magnetic particles in the composite.

1. INTRODUCTION

Nickel–zinc ferrites (NiZn) are widely used in high frequency applications (inductors, transformer cores, microwave devices, etc.) because of their high permeability, high electrical resistivity, low eddy current loss, mechanical hardness, chemical stability and reasonable cost, [1]. This system has a cubic spinel crystal structure with the unit cell consisting of eight formula units of the form $Zn_x^{2+}Fe_{1-x}^{3+}\left[Ni_{1-x}^{2+}Fe_{1+x}^{3+}\right]O_4^{2-}$. 32 oxygen anions per unit cell form a face centered cubic cage, while the metallic cations occupy interstices. The metallic cations outside the bracket occupy the tetrahedral A-sites comprising the tetrahedral sublattice while those metallic cations enclosed by the bracket occupy octahedral B-sites comprising the octahedral sublattice [2]. The magnetic properties such as magnetic permeability of these ferrites depend strongly not only on the chemical composition, but also on the post–sintering density and the microstructure such as porosity, grain size and intra and/or intergranular pores. These are attributed to the fact that the permeability of polycrystalline ferrite can be described by the superposition of two different magnetizing processes: domain wall motion and spin rotation. Additionally, there exists a natural resonance due to the effective anisotropy field, resulting in magnetic losses. This provides some limitations (like a threshold frequency) of the application of polycrystalline ferrite to high–frequency device applications.

The task of this paper was to make a highly permeable NiZn ferrite with good thermal stability for preparation of ferrite polymer composite materials. For this purpose, the Ni–Zn ferrites are often doped with CuO. The choice of CuO is based on the fact that: (a) it increases the density and resistivity, and (b) it decreases the sintering temperature of the ferrite. The CuO reduces to Cu_2O at 1026 °C. The Cu_2O has a low melting temperature (1235 °C) and forms a liquid phase during sintering of ferrites that activates the sintering and increases the ferrite density (i.e. permeability). It allows producing highly permeable ferrites with less ZnO content and higher Curie temperature [3].

The ferrite polymer composite materials, on the other hand, consist of ferrite particles (filler) dispersed in a nonmagnetic polymer matrix [4,5]. They have the unique advantage of mouldability into complex shapes, which is not easily possible by ceramic (sintered) ferrites. The impregnation of magnetic fillers in the matrix imparts magnetic properties to the matrix and modifies the dielectric properties of the matrix [6-9].

The complex initial permeability spectra of NiZnCu ferrite and its composite materials have been studied in the frequency range from 10 kHz to 1 GHz and the contribution of domain wall motion and spin rotational magnetizing processes has been discussed. In the discussion, a simple magnetic circuit model is taken into account in the case of ferrite polymer composite materials.

2. EXPERIMENTS

The prepared NiZnCu ferrite with the chemical formula $Ni_{0.27}Zn_{0.63}Cu_{0.1}Fe_2O_4$ came out of the composition $Ni_{0.3}Zn_{0.7}Fe_2O_4$ which had high initial permeability (≈ 2500) but remarkably low thermal stability (the Curie temperature was only 70 °C). The NiZnCu ferrite was prepared by the ceramic method at 1250 °C/6h in the sintered and also powder form (with the grain size distribution of 0–250 μm). The ferrite polymer composite materials were made of this ferrite and a non–magnetic polymer matrix (PVC) by the dry pressing method. Two series of composite samples were prepared to examine the effects of the ferrite particle size and the ferrite volume concentration on the complex permeability. These were: (1) constant ferrite volume concentration 73 vol% and different particle sizes 0–40, 40–80, 80–160 and 160–250 μm, and (2) constant particle size 0–250 μm and different ferrite volume concentrations 41, 48, 55, 63 and 73 vol%. All the prepared samples had the toroidal form with the dimensions: 12 x 6 x 3 mm. The complex permeability

spectra ($\tilde{\mu} = \mu' - j\mu''$) of the samples were measured by two different techniques. In the low–frequency region, $\tilde{\mu}$ was measured using an impedance analyser (HIOKI 3531 LCR HiTester) on a 5–turn wound toroidal specimen from 10 kHz to 5 MHz, keeping the ac current at 1 mA (which corresponds to the excitation ac magnetic field $H_{ac} \approx 0.4$ A/m). The complex permeability $\tilde{\mu}$ was calculated from complex impedance $Z = R + jX$ by the following equation: $\tilde{\mu} = \mu' - j\mu'' = -jKZ/\omega = -jK(R + jX)/\omega$ where ω is the angular frequency, and $K = C/N^2\mu_o$, where N is the number of turns, and C is a geometric parameter of the toroid, [4]. In the frequency range 1 MHz to 1 GHz, $\tilde{\mu}$ was obtained by measuring the input impedance of samples by the coaxial line technique with another impedance analyser (HP 4191A). The impedance analysers were controlled by a PC computer via a GPIB interface.

3. RESULTS AND DISCUSSION

The frequency dependence of real and imaginary parts (μ' and μ'') of complex initial permeability $\tilde{\mu} = \mu' - j\mu''$ for the prepared NiZnCu sintered ferrite sample is shown in Fig.1. The real part of the complex permeability μ', which is about 1100 at 10 kHz, remained almost constant until the frequency was raised to a certain value, and then began to decrease at higher frequency (≈ 1.9 MHz). The imaginary part μ'' gradually increased with the frequency, and reached a maximum of about 700 at about 6 MHz, where the real part μ' rapidly decreased. The real part μ' of $\tilde{\mu}$ in the frequency region from 2 to 20 MHz depends on the square of the frequency. This feature is well known as the natural resonance phenomenon [1].

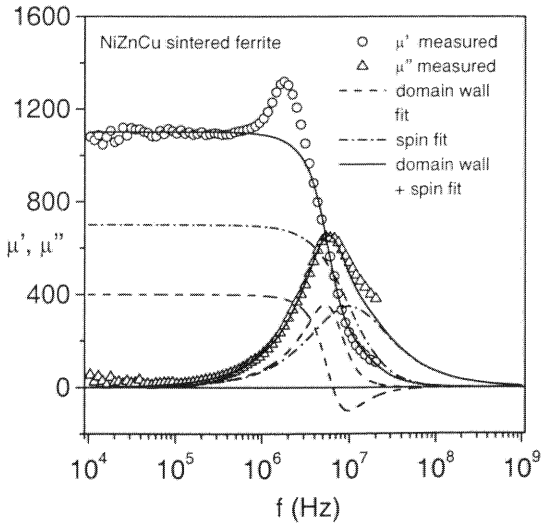

Fig.1. The complex permeability spectra for the NiZnCu sintered ferrite.

In soft polycrystalline ferrites, the magnetisation changes are due to domain wall motion and spin rotation. The former is calculated using the equation of motion of the domain wall, the damped simple harmonic oscillator equation describing motion of a planar 180° – domain wall in the ferrite. The latter is calculated using the Landau–Lipschitz electromagnetic torque equation for spin dynamics, [1, 4, 7]. Both the magnetizing processes (domain wall motion and spin rotation) affect the permeability of the ferrite. Therefore, the complex initial permeability of a polycrystalline ferrite can be calculated as a superposition of these two contributions:

$$\tilde{\mu}(f) = 1 + \tilde{\chi}_{dw}(f) + \tilde{\chi}_{sp}(f). \qquad (1)$$

Both the domain wall susceptibility $\tilde{\chi}_{dw}(f)$ as well as spin rotational one $\tilde{\chi}_{sp}(f)$ are of a resonance type and depend on the square of the frequency:

$$\tilde{\chi}_{dw}(f) = \frac{\chi_{dw} f_{dw}^2}{f_{dw}^2 - f^2 + j\frac{\beta}{m_{dw}} f}, \qquad (2)$$

$$\tilde{\chi}_{sp}(f) = \frac{(f_{sp} + jf\alpha) f_{sp} \chi_{sp}}{(f_{sp} + jf\alpha)^2 - f^2}, \qquad (3)$$

where χ_{dw} and χ_{sp} are static (or low frequency) susceptibilities for domain wall and spin rotational motions, f_{dw} and f_{sp} are resonance frequencies of domain wall and spin components, β and α are corresponding damping factors, m_{dw} is the effective mass of domain wall per unit of its area, and f is the frequency of external ac electromagnetic field. It should be noted that the domain wall component is affected by both the composition and the microstructure (which is sensitive to inner stress and/or distribution of grains), while the spin component is dominated mainly by the chemical composition of the ferrite. If α is large enough ($\alpha \to \infty$), the $\tilde{\chi}_{sp}(f)$ can be approximated by the relaxation–type frequency dispersion:

$$\tilde{\chi}_{sp}(f) = \frac{\chi_{sp}}{1 + j\frac{f}{f_{relax}}}, \qquad (4)$$

where $f_{relax} = f_{sp}/2\pi\sqrt{1+\alpha^2}$ is the relaxation frequency. We can decompose the complex permeability spectra presented in Fig.1 into the domain wall component and the spin rotational contribution by use of Eqs.(1-4). The observed permeability spectra can be well reproduced by the superposition of these two components, where we used the numerically determined parameters using a non–linear least–squares fitting method: $\chi_{dw} = 397.3$, $f_{dw} = 5.2$ MHz, $\beta/m_{dw} = 8.1 \cdot 10^6$,

$\chi_{sp} = 698.4$, $f_{sp} = 1172.1$ MHz, $\alpha = 120$ and $f_{relax} = 9.8$ MHz. It can be noted that for more precise fitting we should consider distributions of these parameters.

On the other hand, the ferrite polymer composite material is considered to be composed of ferrite particles (with intrinsic initial permeability μ_f) surrounded by a non–magnetic polymer layer (with intrinsic initial permeability $\mu_m \approx 1$). The elementary cell of idealized composite structure can be modelled using series–parallel magnetic circuit. Based on the approach described in [4, 8], it can be derived the following formula for the complex effective permeability of the composite sample:

$$\tilde{\mu}_e = \left\langle 1 + \frac{\tilde{\chi}_f (1+\eta)}{1+\tilde{\mu}_f \eta} v_i \right\rangle , \qquad (5)$$

where $\langle\rangle$ denotes the statistical mean value, $\tilde{\mu}_f$ ($\tilde{\chi}_f$) is the complex initial permeability (susceptibility) of the sintered ferrite (given by Eqs.(1-4)), $\eta = d/D$ is the demagnetising parameter (d is the average thickness of the polymer layer and D is the average ferrite particle size). The demagnetising parameter has a distribution and is connected with the volume fraction v_i [8]. Thus, the $\tilde{\mu}_e$ value of composite can be influenced by granulometry (d, D) and composite density (or concentration $\kappa_v = 100 \cdot v_i$).

Figures 2 and 3 present the dependences of real μ_e' and imaginary μ_e" parts of complex effective permeability $\tilde{\mu}_e = \mu_e' - j\mu_e''$ for composite materials based on the NiZnCu ferrite powder and PVC polymer. The parameters of both these graphs are the particle size D (μm), Fig.2, and ferrite particle concentration κ_v (vol%), Fig.3. In the case of composites, we can observe relaxation–type frequency dispersion in contrary to the sintered ferrite (see also Fig.1). The values of μ_e' rose with ferrite volume concentration, but not very much with ferrite particle size. The increase in μ_e' due to a rise of κ_v is in a good agreement with our previous studies [4-8]. The important role in $\tilde{\mu}_e(\kappa_v)$ dependence has the demagnetising parameter η, which decreases with increasing the κ_v. On the other hand, the value of μ_e' in the ferrite polymer composite materials becomes larger than that of the sintered ferrite in the high–frequency region (> 50 MHz). As the ferrite particle concentration decreases, both the shoulder frequency of μ_e' and the peak frequency of μ_e" shift higher. In the composite structure, magnetic poles, which are generated by the applied ac magnetic field produce the demagnetising field H_D, causing permeability to be lower than that of the sintered ferrite. This feature can be explained in the following way. In composite, a magnetic inert component is introduced that causes a cut–off the magnetic circuit in the ferrite. Thus, the permeability is reduced remarkably as the κ_v decreases. The magnetic poles on the surface of magnetic particles dispersed in polymer matrix create the demagnetising field H_D anti–parallel to the applied ac magnetic field. The H_D reduces the induced magnetic moment more than that calculated from the volume concentration. Therefore, the μ_e' in low–frequency region decreases with the configuration change from the sintered ferrite to the composite.

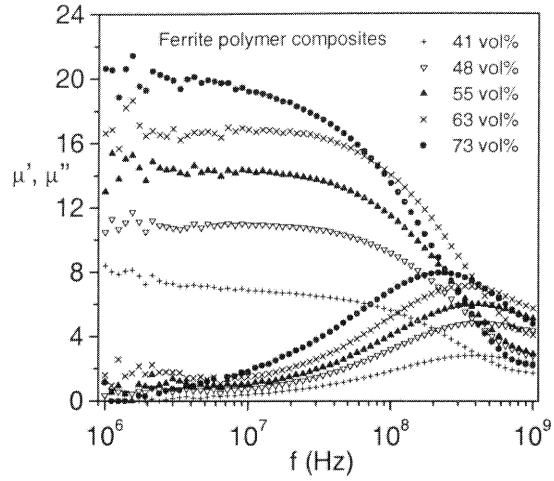

Fig.2. The complex permeability spectra of the composite samples with constant particle size 0–250 μm.

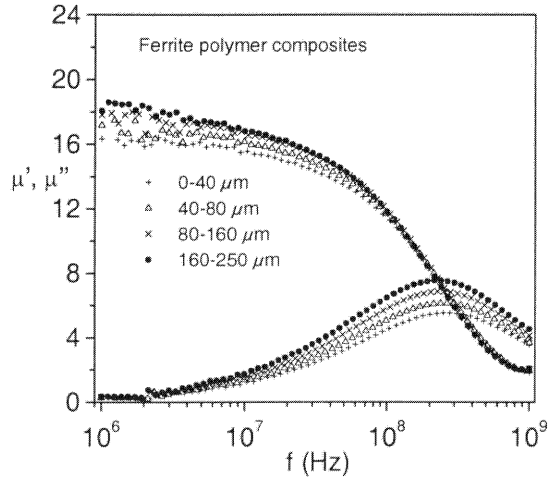

Fig.3. The complex permeability spectra of the composite samples with constant concentration 73 vol%.

On the other hand, the spin resonance frequency is represented by the equation $f_{sp} = \mu_o \gamma H_A / 2\pi$, where H_A is the effective anisotropy field given by the summation of the magnetocrystalline anisotropy field and the shape anisotropy field. In the composite sample, the shape anisotropy field is introduced, which is equivalent to the demagnetising field H_D induced by the ac magnetic field. The spin resonance frequency can then be modified by: $f_{sp} = \mu_o \gamma (H_A + H_D)/2\pi$. The value of H_D

increases with decreasing the value of κ_v. The f_{sp} shifts higher due to the contribution of the H_D.

As an example, the complex effective permeability spectra for composite sample with $\kappa_v = 73$ vol% and $\langle D \rangle = 0$–250 μm were decomposed into the domain wall and spin rotational components with the following values of fitted parameters (Fig.4): $\chi_{dw} = 6.9$, $f_{dw} = 95.4$ MHz, $\beta/m_{dw} = 11.1 \cdot 10^8$, $\chi_{sp} = 11.9$, $f_{sp} = 5.4$ GHz, $\alpha = 14$, $f_{relax} = 384.7$ MHz and $\eta = 0.01$.

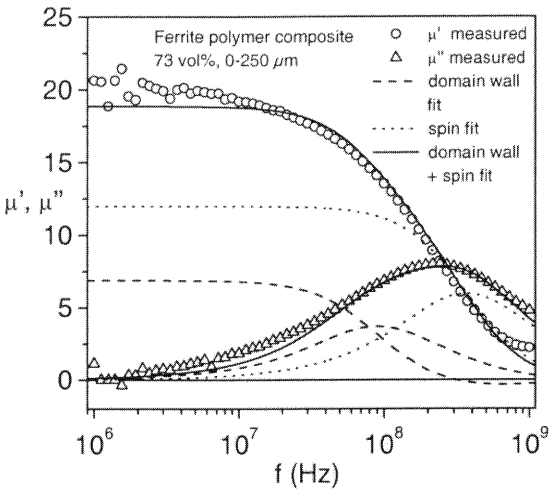

Fig.4. The complex permeability spectra (measured and calculated) for composite sample with $\kappa_v = 73$ vol% and $\langle D \rangle = 0$–250 μm.

A quantitative agreement between the calculated curves and experimental data is obtained. Accordingly, it is possible to estimate the frequency dispersion of $\tilde{\mu}_e$ for composite materials using the dispersion parameters. It should be noted that the permeability of the domain wall component is sensitive to the microstructure of the ferrite. Therefore, domain wall permeability of the composites is difficult to analyse. In addition, we have no experimental information about the damping parameter β of the domain wall permeability for composites at this stage. According to this fact, more detailed studies on the domain wall component of permeability will have to be studied in the future.

4. CONCLUSION

We have studied the variation of frequency dependence of complex initial permeability with ferrite volume concentration and particle size distribution for NiZnCu sintered ferrite of the chemical composition $Ni_{0.27}Zn_{0.63}Cu_{0.1}Fe_2O_4$ and its composite materials using the domain wall and spin resonance formulation combined with the magnetic circuit model approach. In the case of sintered ferrite, the dispersion character is of a resonance type and this is attributed to both the domain wall and spin rotational contributions. In the case of composites, the dispersion character is of a relaxation type and the permeability is larger than that of the sintered ferrite in high–frequency region. This is attributed to the shift of both the spin and domain wall resonance frequencies towards the higher frequency region by arising the demagnetising field in the composites. The magnetic circuit model can be applied to explain the effective permeability variation with ferrite concentration and particle size distribution in both the domain wall and spin components. Therefore, performance stability of some high–frequency devices can be improved using the ferrite polymer composite materials.

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