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INFLUENCE OF APPLIED MAGNETIC FIELD UPON THE COMPLEX PERMEABILITY IN AN MnZn FERRITE AND ITS COMPOSITES

Rastislav Dosoudil * — **Vladimír Olah** * — **Vladimír Ďurman** **

The article deals with the influence of a dc magnetic field upon the complex permeability spectra in an MnZn sintered ferrite and its composites in the frequency range from 10 kHz to 1.6 GHz under magnetic fields up to 100 kA/m. In the case of the sintered ferrite, the spectrum of μ' has a resonance type frequency dispersion above 150 kHz which can be caused mainly by the domain walls vibrations and the spectrum of μ'' has a maximum at about 750 kHz (at $H_{dc} = 0$ kA/m). In the presence of a *dc* magnetic field, the dispersion character changes from resonance type to relaxation type spin resonance with disappearance of the domain walls. The values of μ' in the frequency region above 1.5 MHz are probably decreased because of eddy current effect. In the case of composites, the resonance type of frequency dispersion of permeability is observed above 150 MHz and it can be attributed to the spin resonance, which is stabilized by the demagnetising field in the ferrite particles dispersed in the non-magnetic polymer matrix.

K e y w o r d s: MnZn sintered ferrite, composite material, dc magnetic field, complex permeability spectrum, resonance

1 INTRODUCTION

Materials have two properties that determine how they interact with electromagnetic fields — permittivity (ϵ) or dielectric constant for electric fields and permeability (μ) for magnetic fields. In general, permittivity ($\epsilon^* = \epsilon' - j\epsilon''$) and permeability ($\mu^* = \mu' - j\mu''$) are complex values. The real part (ϵ' or μ') is a measure of how much energy is stored in a material. The imaginary part (ϵ'' or μ'') is a measure of how much energy is lost in a material. These properties are not constant and may change with frequency, for example. The magnetic permeability of polycrystalline (sintered) ferrite is an important factor in the application of some electronic devices. In particular, the frequency dispersion of the complex permeability μ^* determines high-frequency (RF) characteristics of these devices. Natural resonance, which originates from the effective anisotropy field, produces magnetic losses and gives limitation to some RF device applications. Thus, many experimental and theoretical investigations on the frequency dispersion in polycrystalline ferrite have been performed [1–4]. In addition, composite ferrite materials, in which ferrite particles are dispersed in a non-magnetic (polymer) matrix, have been the subject of interest in recent years because of technological application and fundamental basis [5–7]. We have studied the composite materials with MnZn ferrite filler and non-magnetic thermoplastic matrix and found that these composites have higher values of permeability in the RF frequency region (above 50 MHz) than a sintered ferrite (from which the magnetic filler was prepared), [8–9]. *Tsutaoka et al* investigated the dc magnetic field effect on the complex permeability spectra in a sintered NiZn ferrite [10] and found that the frequency dispersion is separated into two parts; two distinct peaks of μ'' corresponding to the domain wall and spin rotation resonances can be identified under several tens of kA/m. The domain wall resonance disappears under about 70 kA/m and the ferromagnetic (or ferrimagnetic) resonance like spin resonance is stabilized. *Bush* has carried out the studies of the dc magnetic field effect on the complex permeability for several ferrites and garnets in RF frequency region [11, 12]. It was found that μ' decreases with increasing magnetic field and they treated this feature as the magnetic field effect on the domain wall resonance in the ferrite materials [12].

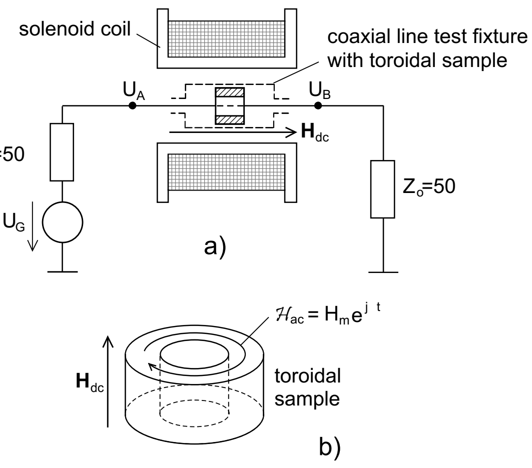

Fig. 1. a) Measuring circuit for RF frequency range (1 MHz to 1.6 GHz) and b) direction of ac (H_{ac}) and dc (H_{dc}) magnetic fields in the torodial sample.

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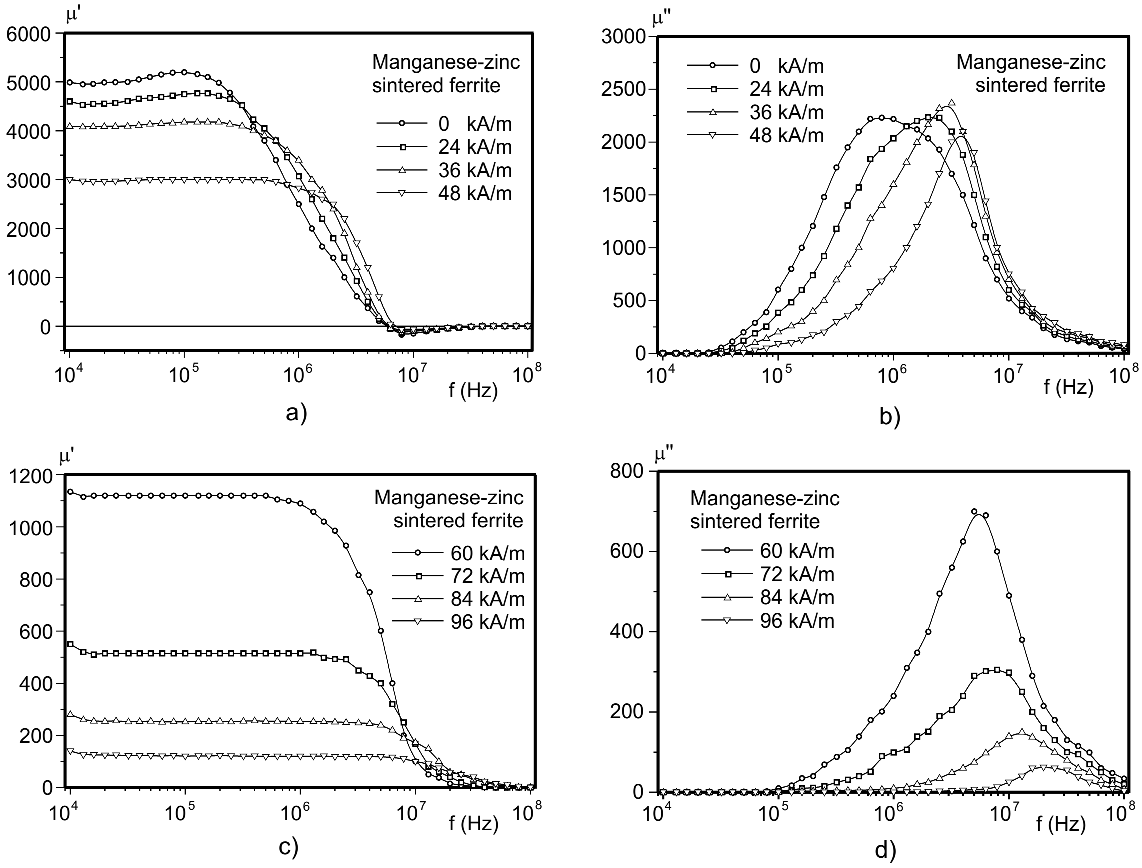

Fig. 2. The frequency dependences of real (a,c) and imaginary (b,d) parts of complex permeability for MnZn sintered ferrite under various dc magnetic fields.

In this work, we have investigated the complex permeability spectra in an MnZn sintered ferrite and its composites from the viewpoint of dc magnetic field effect. The aim of this paper was to study the improvement of permeability in the high-frequency region (10 MHz to 1.6 GHz) and to explain the measured dispersion of permeability.

2 EXPERIMENTAL

2.1 Specimen preparation

A commercially available MnZn sintered ferrite sample in a toroidal form (with an outer diameter of 10 mm, an inner diameter of 6 mm and a height of 4 mm, type H40 — produced by S+M Components, Šumperk, Czech Republic) with magnetic flux density (in saturation) $B = 0.38$ T, coercivity $H_c = 13$ A/m, Curie temperature $T_c > 130\,°\mathrm{C}$, mass density $4.8\,\mathrm{g/cm^3}$, and initial permeability $\mu_i = 4300 \pm 20\,\%$ was used. The sintered ferrite has the chemical composition $\approx \mathrm{Mn_{0.52}Zn_{0.43}Fe_{2.05}O_4}$. Ferrite particles used as the magnetic filler in the composite materials with non-magnetic matrix were prepared by mechanical grinding of the MnZn sintered ferrite (H40) with fraction 40–80 μm. In this case, polyvinylchloride (PVC) was used as a polymer non-magnetic matrix. The ferrite filler and polymer matrix were thermally treated at a temperature of $150\,°\mathrm{C}$, then pressed (in the liquid state) into a plate form, and then cooled down to room temperature. From plates were cut out the toroidal samples with dimensions $10\times6\times4$ mm. The prepared samples of composites had the following mass concentration of ferrite filler: 93 wt % (*ie* 76 vol%), 89 wt % (*ie* 66 vol%), and 81 wt % (*ie* 51 vol%).

2.2 Measurement

The complex permeability ($\mu^* = \mu' - j\mu''$) of the samples was measured by two different techniques. In the frequency range 1 kHz to 5 MHz, μ^* was obtained by measuring the inductance (ωL) and the resistance (R) differences between a coil wound (with number of turns $N = 1$) around the toroidal (sintered ferrite or composite) sample and one wound without the toroidal sample, using a low-frequency impedance analyser (HIOKI 3531 Z HiTester) with a constant effective value of the driving current ≈ 20 mA (in series mode). In the frequency range 1 MHz to 1.6 GHz, μ^* was obtained by measuring the input and output voltages U_A and U_B together with the phase angle (shift) φ_{AB} between both voltages by a vector voltmeter (HP 8405A, coaxial line technique, Fig. 1). As a signal source, the signal generator (R&S SMH 100 kHz–2000 MHz) has been used. The coaxial transmission line test fixture was inserted into a

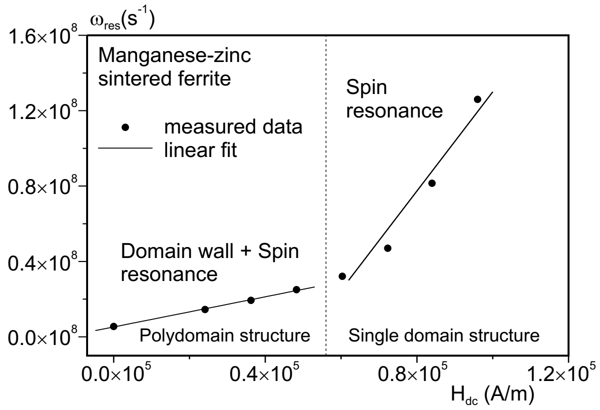

Fig. 3. The dependence of angular frequency $\omega_{\rm res}$ versus dc bias field H_{dc} for MnZn ferrite.

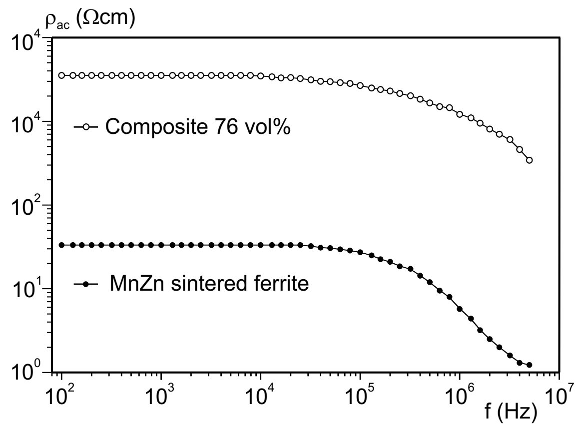

Fig. 4. The dependence of resistivity for MnZn *vs* frequency for MnZn sintered ferrite and its composite (76 vol %).

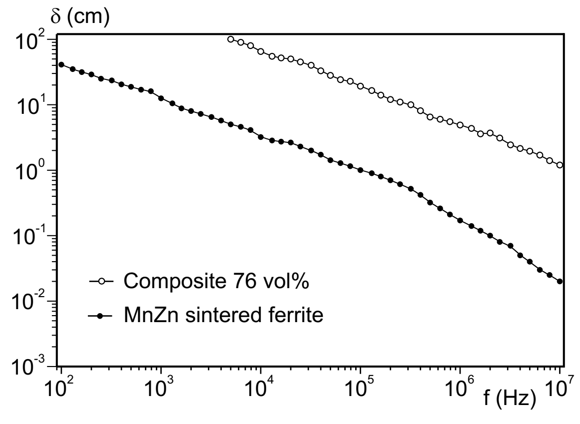

Fig. 5. The dependence of skin depth *vs* frequency for MnZn sintered ferrite and its composite (76 vol %).

solenoid coil with an inner diameter of 42 mm, an outer diameter of 160 mm, a height of 140 mm, and number of turns 3230, which can generate a dc magnetic field H_{dc} up to about 110 kA/m (1400 Oe). In this apparatus, the dc magnetic field H_{dc} is applied perpendicular to the exciting ac magnetic field H_{ac} of constant amplitude ≈ 1.1 A/m. The experimental error for the complex permeability in the frequency range 1 kHz to 100 MHz is less than 6 % and over 100 MHz less than 12 %. The relations for calculations of $\mu^* = \mu' - j\mu''$ can be found in [8]. AC electrical resistivity ρ_{ac} was measured by a two probe method in the frequency range from 100 Hz to 5 MHz at room temperature using an impedance analyser (HIOKI 3531 Z HiTester). During the measurement of ac resistivity, the samples were placed into the type 2904 Tettex plate capacitor. The diameter of electrodes was 50 mm. No additional electrodes (*eg* silver or graphite painted electrodes) were used as they can influence the results by interfacial polarisation [13].

3 RESULTS AND DISCUSSION

3.1 MnZn sintered ferrite

The frequency dependences of the real μ' and imaginary μ'' parts of complex relative permeability ($\mu^* = \mu' - j\mu''$) for an MnZn sintered ferrite are shown in Fig. 2 for several values of dc magnetic field (in Fig. 2a,b for $H_{dc} = 0$, 24, 36, 48 kA/m and in Fig. 2c, d for $H_{dc} = 60$, 72, 84, 96 kA/m). At $H_{dc} = 0$ kA/m, μ' begins to decrease at about 250 kHz and μ'' has a maximum at about 750 kHz. This resonance type of non-linear dispersion is caused mainly by the magnetic domain wall resonance. For $H_{dc} > 0$ kA/m, μ' at low frequencies decreases with increasing H_{dc} and the maximum of μ'' shifts to higher frequencies. Above 60 kA/m external field, relaxation type of dispersion can be observed, Fig. 2c,d. Since the magnetization of this MnZn ferrite almost saturates at about 80 kA/m, this MnZn ferrite is considered to have a single domain structure already at 60 kA/m external field. Therefore, this dispersion is originated by the relaxation type spin resonance due to a high spin damping factor. High permeability of MnZn ferrite at low frequencies is mainly attributed to the magnetic domain walls vibrations. We assume that the damping factor of spin rotation is larger in MnZn ferrite than that in NiZn ferrite [10] in which the resonance type permeability dispersion is observed for a single domain structure under dc magnetic field. According to this fact, it can be considered that a higher dc magnetic field over 100 kA/m is necessary to make the resonance type frequency dispersion in this MnZn ferrite. The dependence of angular frequency $\omega_{\rm res} = 2\pi f_{\rm res}$, at which μ'' has a maximum, versus dc bias field (H_{dc}) for MnZn sintered ferrite is shown in Fig. 3. It has been found that the dependence $\omega_{\rm res}$ versus H_{dc} is linear from 0 to 55 kA/m and also from 56 to 100 kA/m, and the slope of these straight lines changes at about 55.7 kA/m. Below the external field of 55 kA/m, $\omega_{\rm res}$ is determined by the superposition of two peaks originated by domain wall and spin resonance, but the contribution of spin resonance can be small due to suppression of the high frequency permeability by eddy current effect. Above 56 kA/m, maximum frequency of μ'' can be determined by $\omega_{\rm res} = \gamma\mu_o(H_a + H_{dc})$, where γ is the gyromagnetic ratio and H_a is the effective anisotropy

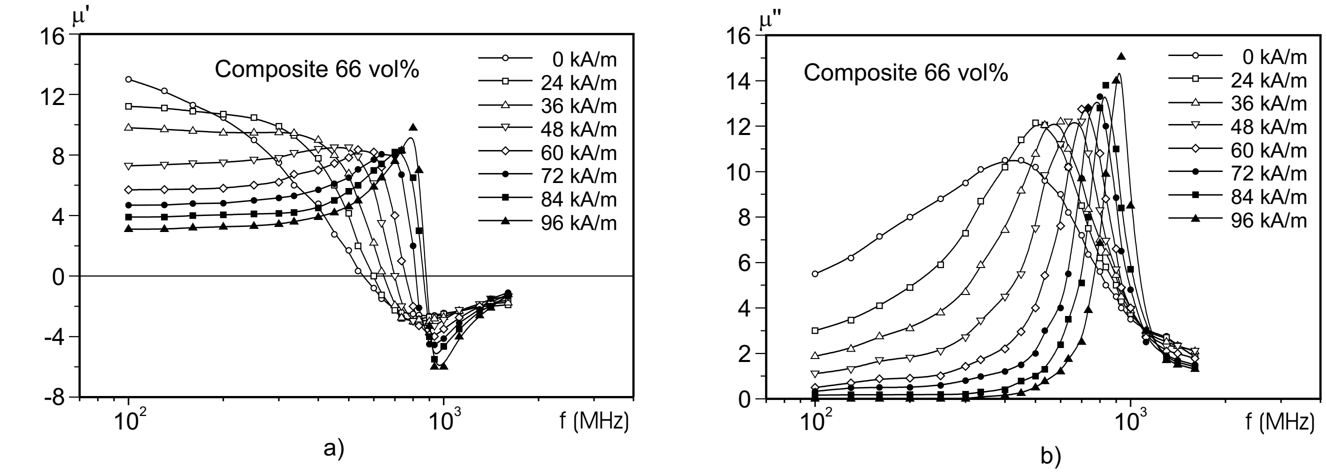

Fig. 6. The frequency dependences of real (a) and imaginary (b) parts of complex permeability for MnZn ferrite composite (66 vol %) under various dc magnetic fields

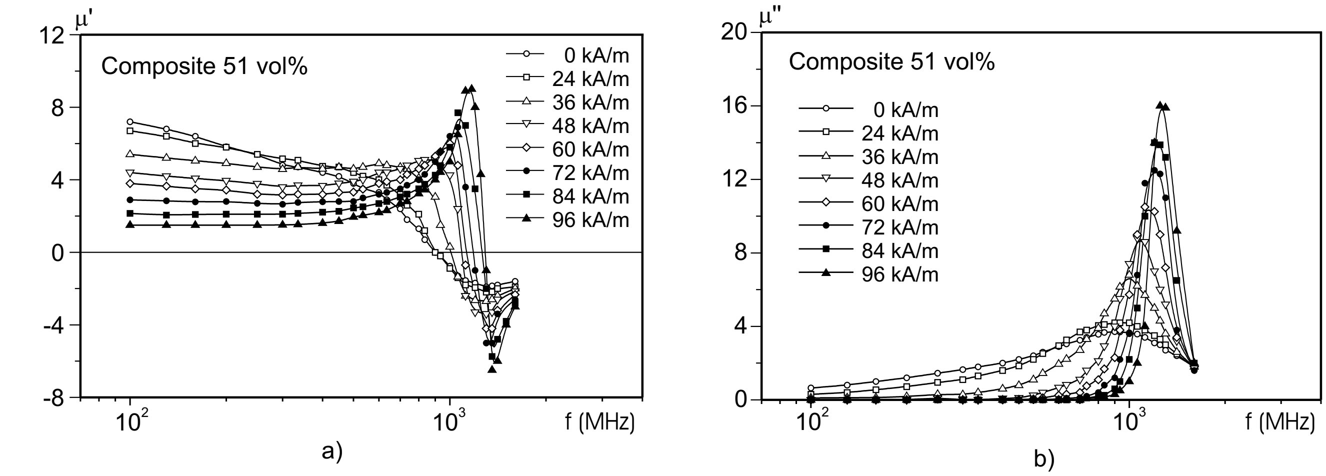

Fig. 7. The frequency dependences of real (a) and imaginary (b) parts of complex permeability for MnZn ferrite composite (51 vol %) under various dc magnetic fields

field which is written by the summation of the magnetocrystalline anisotropy field and partly also the shape anisotropy field. The magnetocrystalline anisotropy field is determined by the composition and the crystal structure of the magnetic component. For the sintered ferrite, the contribution of the magnetocrystalline field is larger than that of the shape anisotropy field, [8, 10]. We assume that the eddy current causes a decrease of the effective volume, which contributes to the permeability of samples due to skin depth effect. Therefore, ac (electrical) resistivity ρ_{ac} was also measured to approximately estimate the frequency variation of the skin depth δ. Frequency dependences of ρ_{ac} and δ for the MnZn sintered ferrite and its composite material (with volume loading of magnetic filler 76 vol %; this high volume loading sample was used as in the case of low volume loading samples ρ_{ac} values cannot be measured with appropriate measuring accuracy by the used impedance analyser) are shown in Figures 4 and 5. The skin depth δ is estimated by $\delta = \sqrt{2/\sigma\,\mu\omega}$ as in the case of high conductivity materials, where the absolute values of permeability and extrapolated ρ_{ac} values (up to 10 MHz) are used, and σ is the (electrical) conductivity. In the case of the sintered ferrite, the sample used for measurement of ρ_{ac} had a toroidal shape with dimensions $10\times6\times4$ mm. In the case of the composite with volume loading of magnetic filler 76 vol %, the sample had a form of a disc with a diameter of 10 mm and a height of 2.5 mm. In low-frequency region, δ is large enough and electromagnetic waves can penetrate into the whole sample. With increasing frequency, the eddy current starts to affect the permeability when the skin depth is smaller than the sample thickness. The skin depth, which affects the effective volume of samples, is a few mm. Accordingly, the skin depth effect can occur from about 1 MHz with increasing frequency in MnZn sintered ferrite. For composite material, ρ_{ac} increases by more than three orders of magnitude in the case of the 76 vol % sample. Since dispersion frequency is located at bout 200 MHz,

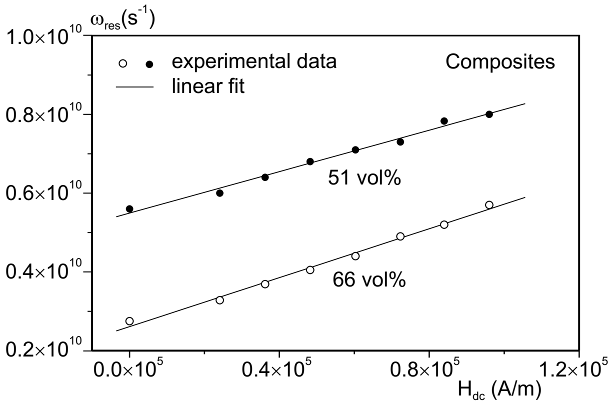

Fig. 8. The dependence of angular frequency ω_{res} versus dc bias field H_{dc} for MnZn ferrite polymer composites (51 and 66 vol %).

there can exist the skin depth effect in the permeability spectra, but it is considered small. It has also been found that with decreasing ferrite content, the eddy current effect is rapidly suppressed by the decrease of conductivity. Therefore, the skin depth effect can be negligible in low ferrite content composites.

3.2 MnZn ferrite based composites

The frequency dependences of complex permeability ($\mu^* = \mu' - j\mu''$) for MnZn ferrite — PVC polymer composites of 66 vol % and 51 vol % are shown in Figures 6 and 7 for several values of a dc magnetic field: $H_{dc} = 0$, 24, 36, 48, 60, 72, 84 and 96 kA/m. In this case, dispersion character is a resonance type and dispersion frequencies are located over 300 MHz at $H_{dc} = 0$ kA/m (the f_{res} is about 450 MHz for the 66 vol % and about 900 MHz for the 51 vol % samples, respectively). This fact can be explained in the following way. The number of magnetic domains in magnetic particles dispersed in a polymer matrix is less than in the sintered ferrite and the domain wall size is also small. Therefore, the domain wall resonance frequency can be higher than that in the sintered ferrite due to the small size of domain walls. In the RF region (several hundreds MHz), we neglect size resonance effects due to small size of ferrite particles. In the case of the used MnZn ferrite filler, the particles had only a polyhedral shape (prismatic, cubic, tetrahedral *etc*), [9]. Therefore, the shape anisotropy field is introduced, which is equivalent to the demagnetising field H_d induced by the RF magnetic field. Next, the spin resonance frequency can be increased by the demagnetising field generated by the magnetic poles on the magnetic particles dispersed in a polymer matrix [8]. In the presence of the external dc magnetic field, dispersion curves become steep in the μ' spectra and peaks become sharp in the μ'' spectra. We can state that no discontinuity is observed between polydomain and single domain structures. This fact shows that frequency dispersion at $H_{dc} = 0$ can be mainly attributable to the spin resonance in composite materials. The next Fig. 8 shows the dc magnetic field dependences of ω_{res} for measured composite samples (66 vol % and 51 vol %). It can be seen that the ω_{res} increases linearly with increasing dc magnetic field in both composite samples and the slope of the curves is almost the same. Thus, in composite materials, the relation between resonance frequency and external dc magnetic field can be expressed in the following way: $\omega_{\mathrm{res}} = \gamma\mu_o(H_a + H_d + H_{dc})$, where H_d is the demagnetising field [8, 10]. In both composite materials, distinct non-linear dispersion in the RF region up to several GHz can be achieved by applying about 80 kA/m (or more) bias field.

4 CONCLUSION

We have studied the dc magnetic field effect on the complex permeability spectra in an MnZn sintered ferrite and its composites. In the case of the sintered ferrite, dispersion character is a resonance type up to about 55 kA/m external dc bias field and this is mainly attributed to the magnetic domain wall resonance. Over about 56 kA/m external dc bias field, the MnZn ferrite is considered to have a single domain structure and the dispersion character is originated by the relaxation type spin resonance due to high spin damping factor. It has also been found that the MnZn ferrite composites show only a resonance type dispersion with and without the external dc magnetic field. At $H_{dc} = 96$ kA/m, all measured samples had a single domain structure; the sintered ferrite shows a relaxation type dispersion and the ferrite composites show resonance one. Therefore, the demagnetising field changes the dispersion character for spin resonance from relaxation type in sintered ferrite to resonance one in composite materials.

The MnZn ferrite is useful in such electronic devices which can operate at frequencies up to several MHz. On the other hand, the ferrite polymer composite materials have higher values of permeability in the RF frequency region (several hundreds MHz) and therefore can extend the spectrum of existing soft magnetic materials in the future. In the RF frequency region ferrite composite materials are suitable for various applications such as electromagnetic wave absorbers, inductor frequency converters, core transducers, ferrite antennas, EMC applications, *etc.* Further investigations for improvement of the permeability above 100 MHz are being carried out.

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