MAGNETIC AND ELECTRIC PROPERTIES OF MAGNETOPOLYMER COMPOSITES

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Summary Permeability spectra of MnZn ferrite composite materials, prepared by mixing ferrite particles with PVC, have been studied. As the ferrite filler content decreases in composite, the quasistatic permeability of the rotation component decreases and relaxation and the resonance frequency shift is higher. The real part of the permeability in the ferrite composite materials becomes larger than that of sintered ferrite in high frequency region.

1. INTRODUCTION

The processing of good-quality magnetopolymers with ferrite powder, which are bounded with polymer binder, has received an important deal of attention during the past years. They are promising magnetocomposite materials for various industrial use, since they posses intrinsic robustness in harsh environments, the ability to work at high frequency and a price advantage over other convenient magnetic materials [1-3].

The dielectric and magnetic properties at very high frequencies up to 10 – 50 GHz can be investigated, but no macroscopic eddy currents can be observed because of low conductivity of both the ferrite powder and the polymer binder. The principal advantage of magnetocomposites lies to reduction of the losses and increasing of the operation frequency range.

It was also shown that magnetic properties of composite can be sensitive to measure temperature or mechanical effects and they can still be used successfully as sensor.

A great number of initial permeability spectra models $\tilde{\mu}(\omega)$ for polycrystalline ferrites are known. Some of them may exhibit complex plots; several dispersion regions, resonance/relaxation character symmetry or non-symmetry, etc. Some of $\tilde{\mu}(\omega)$ spectra for magnetopolymers have been modelled by a convenient mathematical representation of $\tilde{\mu}(\omega)$ components, namely dispersion part $\mu_1(\omega)$ and absorption one $\mu_2(\omega)$, and are still being worked out by others.

We try to solve the problems on the base of grain size distribution and powder concentration account for ferrite filler. The results of modelling include the relations for $\mu_1(\omega)$ and $\mu_2(\omega)$ which allow the adequate presentation of both resonance and relaxation $\tilde{\mu}(\omega)$ when appropriate parameters, having clear physical meaning are used.

In addition, the modelling makes it possible to explain several alternations in $\tilde{\mu}(\omega)$ spectra. The spectra can be influenced by several properties of composite microstructure, e.g. homogeneity of grain size distribution, processing history of sample etc.

2. THEORETICAL BASE

From the thermodynamic aspect for polycrystalline ferrites and polymer ferrite composites the permeability spectrum can be described by superposition of two components in complex domain, that can be attributed to two types of processes associated with relaxation and resonance effect

$$\tilde{\mu}(\omega)=\tilde{\mu}_r(\omega)+\tilde{\mu}_0(\omega) \qquad (1)$$

We assume that both relaxation $\tilde{\mu}_r(\omega)$ and resonance $\tilde{\mu}_0(\omega)$ components may be attributed to two types of magnetizing processes, spin rotation and domain wall motion [4]

$$\tilde{\mu}(\omega)=1+\tilde{\chi}_{sp}(\omega)+\tilde{\chi}_{dw}(\omega) \qquad (2)$$

The spin rotation complex susceptibility component $\tilde{\chi}_{sp}(\omega)$ and also the domain wall component $\tilde{\chi}_{dw}(\omega)$ are relaxation and/or resonance type.

Magnetisation processes due to rotation of magnetisation are calculated using the electromagnetic torque equation, usually with phenomenological added Landau-Lifshitz (L.L.) or Gilbert loss term for magnetic materials. Starting form L.L. equation, Kittel calculated the frequency dependence of complex initial susceptibility as

$$\tilde{\chi}_{sp}=\frac{\omega_0^2+(\Lambda/\chi_0)(j\omega+\Lambda/\chi_0)}{\omega_0^2+(j\omega+\Lambda/\chi_0)^2}\chi_i \qquad (3)$$

with the angular ω and resonance frequency

$$\omega_0=|\gamma|H_{ef} \qquad (4)$$

where γ is the gyromagnetic ratio. The effective magnetic field may be $H_{ef}=2K/J_s$ for magnetocrystalline equivalent field with constant energy K and saturated magnetic polarisation J_s. If static initial susceptibility at rotation process is $\chi_i=J_s^2/3K$, then Eq.4 is $\omega_0=2|\gamma|J_s/3\chi_i$. The mean rotation susceptibility χ_0 is roughly equal to χ_i. The parameter $\Lambda = |\gamma|\alpha J_s$ defines spin relaxation frequency in L.L. equation with damping coefficient α. For weak damping $\Lambda \ll |\gamma| J_s$ Eq.3 exhibits resonance character

$$\chi_{sp0}(\omega) = \frac{\chi_i}{1-(\omega/\omega_0)^2} \tag{5}$$

and for strong damping $(\Lambda \gg |\gamma| J_s)$ it exhibits relaxation

$$\tilde{\chi}_{spr} = \frac{\chi_i}{1+j\omega/\omega_r} \tag{6}$$

with relaxation frequency $\omega_r = \Lambda/\chi_i = |\gamma|\alpha J_s/\chi_i$. For the estimation of quasi-static permeability of the magnetocomposite sample the composite model was proposed, [5]. The derived equation can be modified as

$$\mu = \left\langle \mu_s \frac{(1+\eta)^2 \kappa_v}{1+\eta\mu_i} \right\rangle + \left\langle 1-(1+\eta)\kappa_v \right\rangle \tag{7}$$

where μ_s may be static intrinsic permeability at domain wall or rotation magnetisation of filler particles. The elementary cell of two components model considers polymer matrix surrounding the ferrite particles and it was proposed as a series-parallel magnetic circuit approximation. It assumed that, the gap length d between adjacent particles is very low in comparison with particle size D, *i.e.* $d \ll D$, in applied magnetic field direction. The other assumption was high filler concentration κ_v in composite and it is connected with demagnetising structural parameter $\eta = d/D$. The brackets $\langle\,\rangle$ denote the statistical mean value.

If one substitutes Eq.3 to $\tilde{\mu}_s = 1 + \tilde{\chi}_{sp}$ permeability in Eq.7, then this equation is rearranged to the estimation of the complex permeability spectra $\tilde{\mu}_{sp}(\omega)$ of magnetocomposite corresponding to the rotational magnetisation.

For domain wall motion magnetising process with resonance phenomena of oscillating walls and by acceptation of Eq.7 the permeability model given in [6] can be modified for the magnetocomposite as

$$\begin{aligned} \tilde{\mu}_{dw}(\omega) &= \mu_{dw1}(\omega) - j\mu_{dw2}(\omega) = \\ &= \left\langle \frac{\mu_s (1+\eta)^2 \kappa_v}{(1+\eta\mu_s)\left[1-(\omega/\omega_0)^2 + j(\omega/\omega_r)\right]} \right\rangle + \\ &+ \left\langle 1-(1+\eta)\kappa_v \right\rangle \end{aligned} \tag{8}$$

where ω_0 is angular resonance frequency connected with domain wall mass m_w. The relaxation frequency ω_r is connected with relaxation time T of domain wall motion in this magnetisation process.

3. EXPERIMENTS AND DISCUSSION

The frequency dependences of complex permeability $\tilde{\mu}(\omega)$ and its components $\mu_1(\omega)$ and $\mu_2(\omega)$ for the ferrite polymer composite were measured and can be described on samples, which successively contained 47.3, 51.0, 57.3, 66.0, 73.4 vol% MnZn filler powder in PVC matrix. The powder fraction of (40 - 80) μm was chosen, *i.e.* the average particles size was <*D*> = 60 μm. The $\tilde{\mu}(\omega)$ of samples were measured in frequency range 1 MHz to 1.66 GHz using a coaxial line cell.

For the samples containing 51 vol% MnZn filler powder the frequency spectra of $\tilde{\mu}(\omega)$ are plotted as locus in the complex plane Fig. 1, where the real component $\mu_1(\omega)$ is plotted against the imaginary component $\mu_2(\omega)$. The locus for the magnetopolymer has the shape characteristic for the rise of resonance and in this case the resonance frequency ratio is $\omega_0/\omega_r \cong 2$. Thus, in the composite the frequency spectra $\tilde{\mu}(\omega)$ are affected except, for relaxation effect by resonance phenomena too.

For sintered MnZn ferrite, the measured locus of $\tilde{\mu}(\omega)$ spectra have different shape and absolute values of permeability in comparison with the magnetocomposite sample. For the ferrite the real part $\mu_1 \approx 500$ in the low frequency region, begins to decrease at 0.2 MHz and reaches about $\mu_1 \approx 10$ at 200 MHz. On the other hand, the imaginary part μ_2 has a maximum of about $\mu_2 \approx 2300$ at around 1 MHz and this ferrite has relative large power loss.

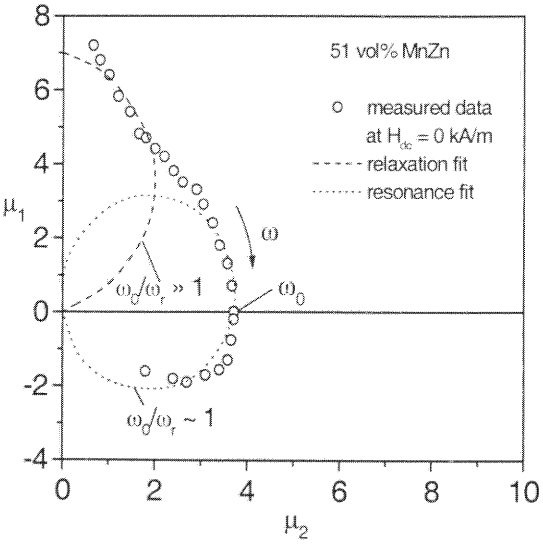

Fig. 1. The complex permeability spectra for composite with κ_v = 51.0 vol% of MnZn powder.

The complex permeability data in Fig.1 measured on composite samples have significantly lower $|\tilde{\mu}(\omega)|$ values than were that measured on sintered ferrite sample. It is due to decreasing of ferrite volume content κ_v in magnetocomposite and as well as the demagnetising effect of ferrite particles represented by η parameter, see Eq.7.

Since the domain structure of particle grains and also the energy of domain walls of powder particles in composite sample are influenced by demagnetising effect, the effective anisotropy field H_{ef} in Eq.4 is written by summation of magnetocrystalline anisotropy field $\overline{H}_a$ and the shape anisotropy $\overline{H}_d$, *i.e.* $\overline{H}_{ef} = \overline{H}_a + \overline{H}_d$. Therefore, there are probably only the rotating magnetic reversals in magnetocomposite. Both the relaxation and resonance effect of composite samples, due to shape anisotropy of small particles, rise at higher frequency region than in sintered ferrites. Thus demagnetising field increases with decreasing volume of filler and the resonance frequency becomes higher. As a result μ_1 can take larger value than that of the sintered ferrite in the high frequency region.

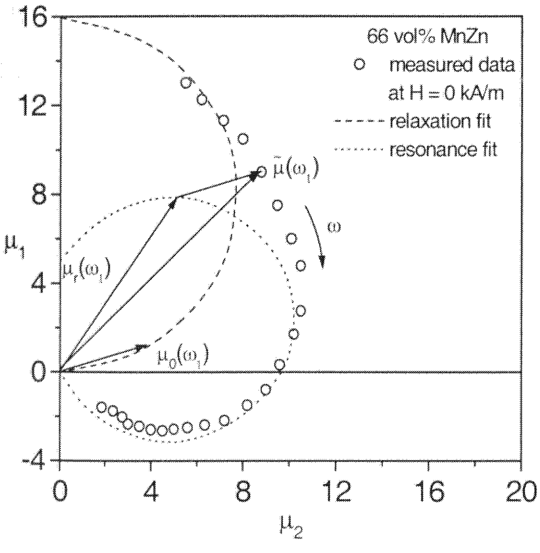

Fig. 2 The complex permeability spectra for composite with κ_v = 66.0 vol% MnZn powder.

The numerical fitting of the measured spectrum $\tilde{\mu}(\omega)$ of magnetopolymer composite follows from Eq. 8 for magnetisation due to domain wall motion or from combination of both Eq.3 with Eq.7 for magnetisation due to rotation. According to the analysis of measured spectra shown in Fig.1, they can be traced as the combination of two loci. One arc is the first part of $\tilde{\mu}(\omega)$ measured data, which corresponds to pure relaxation effect for low frequency region. The rest of relaxation $\tilde{\mu}(\omega)$ spectra, *i.e.* the part of locus plotted by dashed line can be interpreted as the contribution of relaxation effect to the total complex permeability in higher frequency region. This approximate locus can be characterised by the resonance–to–relaxation frequency ratio $\omega_0/\omega_r \gg 1$. This dashed relaxation locus can be calculated by combination of both Eq.6 and Eq.7. The second part of $\tilde{\mu}(\omega)$ data measured at higher frequency band has the resonance character and can be approximated by resonance roughly circular (dashed) locus with ratio $\omega_0/\omega_r \approx 0.1$.

The changes of measured $\tilde{\mu}(\omega)$ spectra due to the change of volume concentration κ_v = 66.0 and 73.4 vol% of MnZn fillers in composite are presented in Fig.2 and Fig.3. These experiments show the increase of absolute values $|\tilde{\mu}(\omega)|$ with increasing κ_v value. It can be estimated that demagnetising parameter η plays the important role in $|\tilde{\mu}(\omega)|$ increasing, see Eq.7. In addition the values of η is decreasing with increasing of κ_v. Thus apparent value of permeability increases with increasing of κ_v and with decreasing of η. The optimal selected type of technological procedure can influence the permeability values by both η and κ_v composite structural parameters.

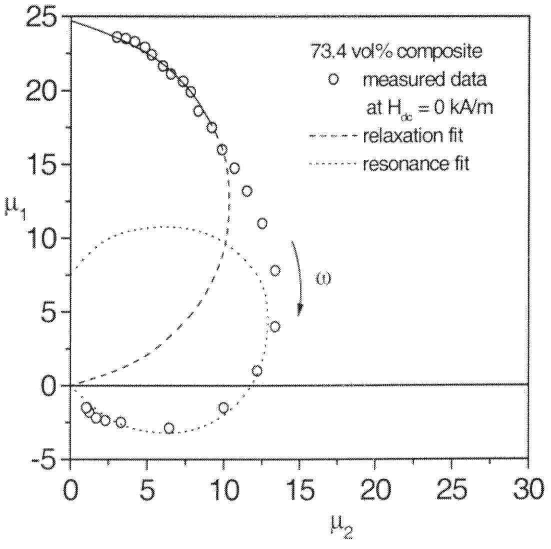

Fig. 3 The complex permeability spectra for composite with κ_v = 73.4 vol% MnZn powder.

Also each of $|\tilde{\mu}(\omega)|$ data in both Fig.2 and Fig.3 can be approximated by combination of two loci, one locus having pure relaxation behaviour with $\omega_0/\omega_r \gg 1$ and second locus is a pure resonance type with $\omega_0/\omega_r \approx 0.1$. The example of the superposition of the relaxation component of permeability $\tilde{\mu}_r(\omega_1)$ and the resonance $\tilde{\mu}_0(\omega_1)$ component to get $\tilde{\mu}(\omega_1) = \tilde{\mu}_r(\omega_1) + \tilde{\mu}_0(\omega_1)$ is shown in Fig.2 for $f_1 = 300\ MHz$.

We were examining the electromagnetic wave absorber using the magnetocomposite materials. We have designed the single layer electromagnetic wave absorber for the RF frequency using dispersion curves. The center frequency of the absorption shifts toward higher frequency and the maximum absorption is reduced as η parameter increases.

4. CONCLUSION

We have studied the frequency spectra of the complex permeability for MnZn ferrite composite materials using relaxation and resonance effect with magnetic circuit model. The model analysis respects demagnetising parameter η, which accepts average size of ferrite particles with comparison to gaps between particles and respects the volume concentration of particles in composite. We have also studied of relaxation and resonance effect contribution to frequency spectra of $\tilde{\mu}(\omega)$.

In low-frequency region, all the real μ_1 and imaginary μ_2 part and apparent permeability of composite sample are much lower and in addition these parameters decrease with decreasing ferrite content in composite samples. However in the high frequency region (for example over 100 MHz), the values of μ_1 for the ferrite composite is larger than that of the sintered ferrite. In addition the power loss is much lower in the composite sample than that in sintered ferrite sample.

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