MICROWAVE MEASUREMENTS OF FERRITE POLYMER COMPOSITE MATERIALS

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Summary The article focuses on the microwave measurements performed on the nickel-zinc sintered ferrite with the chemical formula $Ni_{0.3}Zn_{0.7}Fe_2O_4$ produced by the ceramic technique and composite materials based on this ferrite and a non–magnetic polymer (polyvinyl chloride) matrix. The prepared composite samples had the same particle size distribution 0–250 μm but different ferrite particle concentrations between 23 vol% and 80 vol%. The apparatus for measurement of the signal proportional to the absolute value of scattering parameter S_{11} (reflexion coefficient) is described and the dependence of measured reflected signal on a bias magnetic field has been studied. By means of experiments, the resonances to be connected with the geometry of microwave experimental set–up were distinguished from ferromagnetic resonance arising in ferrite particles of composite structure. The role of local interaction fields of ferrite particles in composite material has been discussed.

1. INTRODUCTION

The nickel–zinc ferrites with high permeability and low loss are widely used in the RF frequency band (up to about 50 MHz) [1]. On the other hand, ferrite polymer composites (FPC) produced by embedding ferrite particles into a non–magnetic polymer matrix have a significant advantage over ceramic materials [2]. Because of the distribution of ferrite particles in a polymer matrix, mechanical behaviour is largely determined by the polymer and magnetic properties by the ferrite filler. The result is a rugged material with stable mechanical and electromagnetic properties. Comparison of FPC and the NiZn ceramic ferrite shows that although FPC has a lower permeability it is stable in the presence of external influences such as pressure, temperature and superimposed magnetic fields. Its magnetic dilution also yields a much broader frequency response, similar to that of NiZn sintered ferrites of low permeability, which have the disadvantage of irreversible degradation after exposure to a strong magnetic field. We have studied the complex permeability spectra of ferrite polymer composite materials based on an MnZn ferrite and PVC matrix and had shown that the permeability of composites is higher in the frequency range over 100 MHz and that their resonance frequencies are higher than that of sintered ferrite due to arising of demagnetising fields of ferrite particles in the composites [3-5].

The present work deals with the ferrite polymer composites based on a ceramic NiZn ferrite and a non–magnetic polyvinyl chloride (PVC) polymer matrix. The effect of filler volume concentration of ferrite particles in composite materials on ferromagnetic resonance field B (magnetic flux density) has been studied at a constant frequency (9.24 GHz) of microwave electromagnetic field. The relation between resonance frequency ω_r and bias field B is presented. The influence of the shape of the sample under test on the ferromagnetic resonance is also discussed from the viewpoint of the role of local interaction fields of ferrite particles dispersed in a polymer matrix.

2. EXPERIMENT

The $Ni_{0.3}Zn_{0.7}Fe_2O_4$ ferrite in the powder form was used as the filler in the composites. The powder ferrite was prepared by the conventional ceramic method and had a particle size of 0–250 μm. In its sintered form this ferrite has an initial permeability $\mu_i \approx 2500$ and is suitable for applications in the frequency band below 1 MHz. The non–magnetic matrix was polyvinyl chloride (PVC). A simple mixing and moulding process was used to produce cylindrical composite samples with ferrite volume concentrations $\kappa_v = 23$ vol%, 48 vol%, 55 vol%, 63 vol%, 73 vol%, and 80 vol%. The cylindrical samples had a height of 1.3 mm and a diameter of 9.5 mm. The sintered ferrite was also prepared in the spherical form with a diameter of 1.4 mm.

The microwave measurements were performed on a special experimental set–up utilizing the principle of measurement of ferromagnetic resonance based on the detection of microwave signal reflected from a sample under test. The block diagram of the described experimental set–up is shown in Fig.1. During the measurement, the sample is kept in a quasistatic (bias) magnetic field with added ac magnetic field of small frequency ($f_{swept} = 72$ Hz). At ferromagnetic resonance the interaction between the magnetization vector $\vec{M}$ and the microwave field $H_{\sim}$ is very strong, and the spin system absorbs the energy from the microwave field. The result is a change of voltage in a microwave detector, which detects reflected microwave signal from the sample. The microwave detector picks up a signal proportional to the voltage that depends on bias field acting on the sample. Microwave power reflected from the sample is therefore dependent on the value of external magnetic field and this dependence is called the modulation curve of ferromagnetic resonance. If during the measurement the frequency and power of microwave field is kept at a constant value and simultaneously the bias field is quasistatic (swept), then the reflected signal arising in the microwave detector is proportional to the slope of modulation curve.

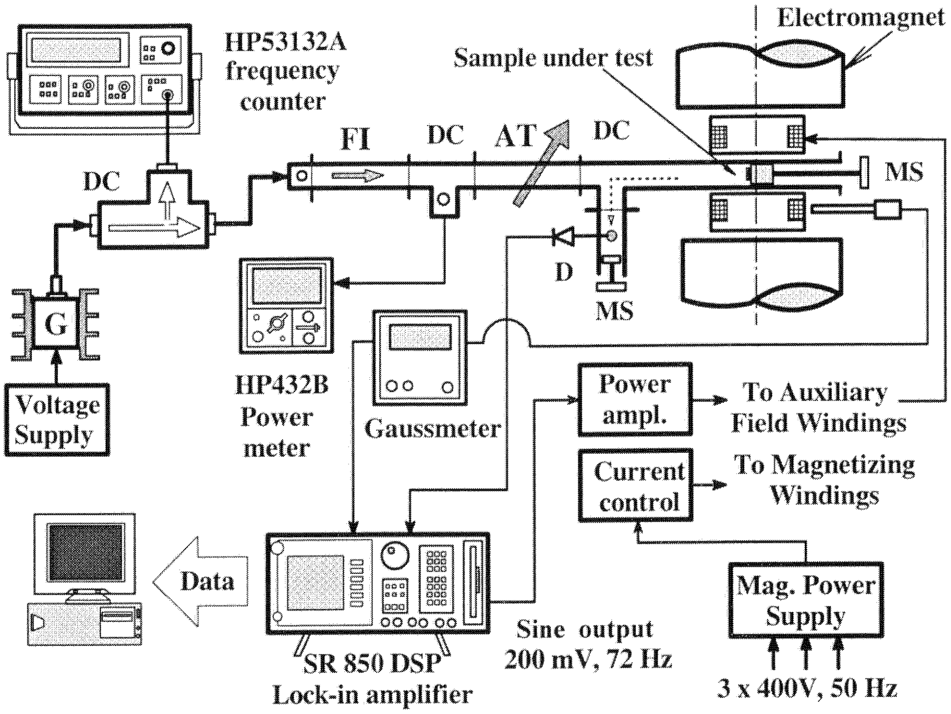

Fig.1. The block diagram of experimental set–up, MS – movable short, DC – directional bridge, FI – ferrite isolator, AT – power attenuator, D – microwave diode (detector).

If the modulation curve were linear within a small interval of the given bias field B, then the voltage in the detector would contain only the first harmonic component of the voltage with frequency f_{swept} given by an auxiliary ac field. However, the modulation curve is non–linear in nature. Therefore, the modulation of ac signal is also non–linear and the detector detects a non–harmonic signal (at a constant bias field B), with the dominant first harmonic component. The detector picks up both the measured signal and the noise, but the level of noise is higher than the amplitude of measured signal. In order to distinguish these two signals, a synchronous lock–in amplifier has been used, which stores the amplitude and phase of a corresponding harmonic component into the internal memory. At the same time, the lock–in amplifier also stores the values of bias field measured by a digital Gaussmeter equipped with an analogue output.

During the measurement of ferromagnetic resonance, the first and/or second harmonic component of swept field is stored in the amplifier's memory. The frequency of swept field is $f_{\mathrm{swept}} = 72$ Hz. The signal of this frequency, generated by an internal generator in the synchronous amplifier, is delivered to a power amplifier and then to the auxiliary windings making ac auxiliary field parallel to the direction of bias field. The auxiliary windings (with 80 turns) form a Helmholtz set–up and generate ac magnetic field with the magnitude of several mT (tens of Gauss). The resolution of the microwave detector is higher than 60 dB. It should be noted that in the case of symmetrical modulation curve, it could also be observed the maximum value of the second harmonic component of swept signal (at the ferromagnetic resonance). The value of resonance bias field is given by a change of phase in 180° and simultaneously by the drop of reflected signal down to zero. The resonances due to the geometry of microwave experimental set–up were distinguished from the ferromagnetic resonance arising in ferrite particles of composite structure.

3. RESULTS AND DISCUSSION

Figures 2 and 3 show the typical dependences of a reflected signal on bias field B measured at a constant frequency of microwave field 9.24 GHz for low (Fig.2) and high (Fig.3) ferrite volume concentration: 23 vol% and 80 vol%, respectively. The change of magnitude and phase of reflected signal indicates the presence of ferromagnetic resonance.

The spin dynamics can be evaluated by the equation of motion of the magnetization vector $\vec{M}$ in the Landau–Lipshitz form [6,7]:

$$\frac{d\vec{M}}{dt} = -\mu_0\gamma(\vec{M}\times\vec{H}_0) - \frac{\alpha\mu_0\gamma}{|\vec{M}|}[\vec{M}\times(\vec{M}\times\vec{H}_0)], \quad (1)$$

where $\vec{H}_0$ is the effective field vector responsible for the magnetic moments (spins) alignment, γ is the gyromagnetic ratio ($1.7586 \cdot 10^{11}$ $s^{-1}T^{-1}$), and α is the damping parameter. The precession of vector $\vec{M}$ around the internal field $\vec{H}_0$, called ferromagnetic resonance, appears under the action of a weak microwave field $H_\sim$ of frequency ω. Solution of Eq.(1) leads to the expression of the Polder's tensor of permeability. The ferromagnetic resonance arises at the frequency ω_r for which the imaginary part of the diagonal terms is maximum. However, as shown first by Kittel [8], the condition for resonance depends on the shape of the sample under test. The Kittel's equation applicable to an rotational ellipsoid (which is equivalent to a low cylinder) in the saturated magnetic state, placed in the empty space, and with demagnetising factors denoted by N_x, N_y, and N_z is:

$$\omega_0 = \mu_0\gamma\sqrt{[H + M_s(N_x - N_z)][H + M_s(N_y - N_z)]} \;, (2)$$

where H is the intensity of external bias field. In the presence of damping (α), the Eq.(2) must be modified:

$$\omega_r = \frac{\omega_0}{\sqrt{1+\alpha^2}} \; . \quad (3)$$

It should be noted that the z–axis is in the direction of the field H and the y–axis and x–axis are perpendicular to the z–axis. From the Eqs.(2) and (3), and from the measurement we can determine the resonant value of the applied magnetic flux density $B = \mu_0 H$ at a constant frequency of microwave field.

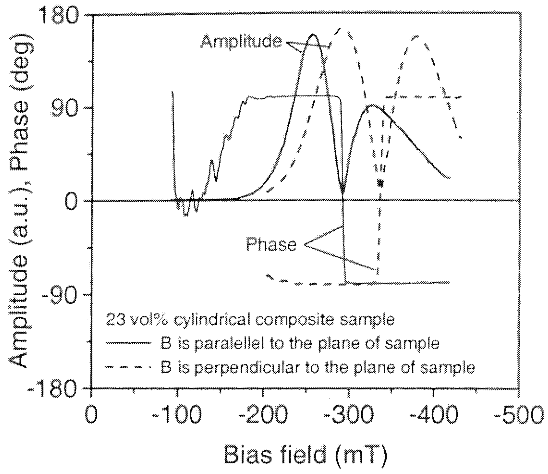

Fig.2. The dependences of amplitude and phase of reflected signal vs. bias field of 23 vol% composite sample for two different directions of bias field.

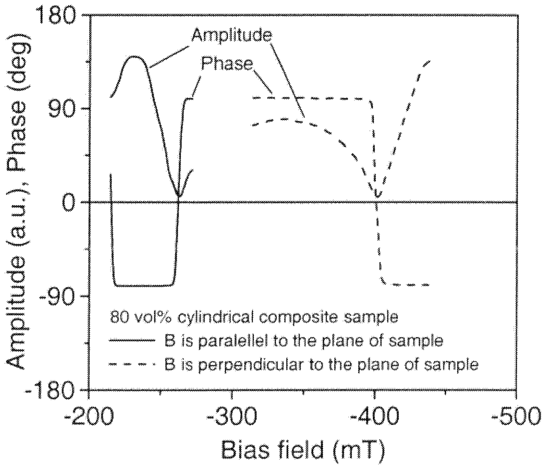

Fig.3. The dependences of amplitude and phase of reflected signal vs. bias field of 80 vol% composite sample for two different directions of bias field.

The demagnetising factors of the disc used were taken as $N_x = 0.797$ and $N_y = N_z = 0.102$. The saturation magnetization of $Ni_{0.3}Zn_{0.7}Fe_2O_4$ ceramic ferrite, measured by means of a vibrating sample magnetometer, was $M_s \approx 100$ kA/m. The calculated values of resonant fields $B \approx 280$ mT (parallel field) and $B \approx 330$ mT (perpendicular field) for the sample with the filler concentration of 23 vol%, and $B \approx 260$ mT (parallel field) and $B \approx 405$ mT (perpendicular field) for the sample with the filler concentration of 80 vol% are in good correlation with the measured values (see Figs.2 and 3).

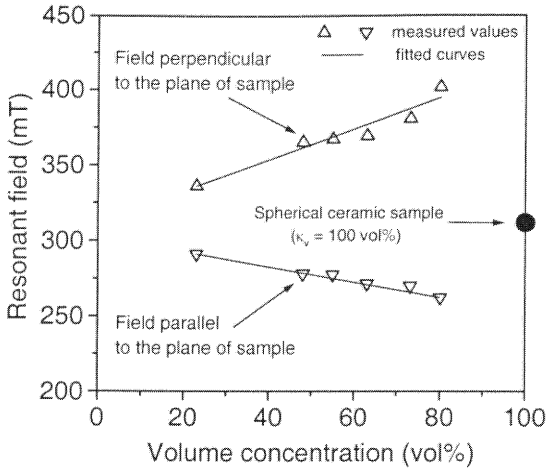

Fig.4. The dependences of resonant field vs. ferrite volume concentration in composites for two different directions of bias field.

Figure 4 shows the dependences of resonant field B vs. ferrite volume concentration κ_v of composites at two different directions of bias field B to the plane of the sample under test. At low values of concentration κ_v (around 23 vol%), the composites have almost the same values of resonant fields close to that of the spherical ceramic ferrite sample and the influence of local interaction fields of ferrite particles dispersed in polymeric matrix on ferromagnetic resonance is not significant. On the other hand, if the ferrite concentration in composite starts to increase, the resonant field B will be remarkably different for both different directions. It is assumed that the effect of local interaction fields of magnetic particles starts to act and as a result the ferromagnetic resonance is dependent on the shape of sample under test. This feature can be understood as follows. In the case of low volume concentration, particles in composite are far enough from each other. Accordingly, the reciprocal effect of particles is considered to be small. In the case of high volume concentration, particles can form clusters and also their mutual distance is much smaller, which leads to an increasing of their reciprocal effect. As a result, the local interaction fields of particles start to assert.

4. CONCLUSION

The influence of volume concentration of ferrite particles dispersed in composite materials on resonant field was shown for two special cases of directions of bias field to the plane of the sample under test. The ferromagnetic resonance depends mainly on the internal magnetic field and is highly affected by the shape of a sample under test. There is no doubt that the ferrite polymer composite materials can extend the spectrum of existing applications of soft magnetic ferrites in the future. They can be used as microwave absorbers, phase shifters, isolators, circulators, EMC shielding, filters, frequency–selective non–linear devices, etc.

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