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Frequency Variation of Complex Permeability in Dual Ferrite Filler — Single Polymeric Matrix Composites

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The frequency dependences of complex initial permeability for triple-component composite materials based on two different types of ferrite filler (MnZn and NiZn ferrite) and a nonmagnetic polymeric matrix (PVC) were studied in the frequency range of 10 kHz–1 GHz. The frequency dispersion of permeability is discussed by means of a dynamic model based on the superposition of the two types of magnetic resonance, the resonance of vibrating domain walls and the resonance of precessing magnetic moments in domains.

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1. Introduction

Composite materials based on ferrite filler and polymeric matrix have been a subject of considerable interest in the last years [1–4]. A relative new composite is the hybrid composite material, which is obtained by using two or more different kinds of fillers in a single matrix [1, 2]. Hybrids have a better all-round combination of properties than composites containing only a single filler type. In this work, the complex permeability spectra of triple-component composite materials were studied and the contribution of two basic magnetizing processes, the domain wall motion and the magnetic moments rotation in domains have been discussed. For this purpose, the dynamic model enabling us to interpret the resonance/relaxation effects in these materials is used.

2. Experimental

As magnetic fillers, a commercially available $Mn_{0.52}Zn_{0.43}Fe_{2.05}O_4$ ferrite and the $Ni_{0.33}Zn_{0.67}Fe_2O_4$ ferrite synthesized by a ceramic route at 1200°C/6 h in air have been used. Both these ferrites were prepared in sintered and also powder form (with a constant particle size of 0–250 μm). Polyvinylchloride (PVC) was used as a nonmagnetic polymer matrix to produce composite materials. Triple-component composites were prepared using the dry low-temperature hot-pressing method (at 150°C and 5 MPa). All composite samples were prepared in the form of toroids with an outer diameter of 8 mm, an inner diameter of 3.5 mm, and a height of 3 mm. The frequency dependences of real μ' and imaginary μ'' parts of complex permeability $\widetilde{\mu} = \mu' - \mathrm{j}\mu''$ were obtained by means of an impedance spectroscopy using two vector analyzers (Agilent 4192A: 10 kHz–10 MHz and Agilent 4191A: 1 MHz–1 GHz).

3. Results and discussion

Complex permeability spectra $\tilde{\mu} = \mu' - \mathrm{j}\mu''$ for both sintered ferrites (MnZn and NiZn) can be found in Fig. 1. It can be seen that at low frequencies the real part of complex permeability, μ', is about 5630 for MnZn ferrite and about 1644 for NiZn one. As the frequency increases, each measured spectrum remains level at first and then rises to a certain peak before falling rapidly to relatively low values. The loss component, μ'', rises to a pronounced peak as μ' falls. The critical frequency f_c (resonance or relaxation), at which μ'' has a maximum value, is about 850 kHz for MnZn ferrite and about 3.2 MHz for NiZn. The dispersion of permeability in the sintered ferrites is principally due to the resonance of oscillating domain walls and the resonance of rotating (precessing) magnetic moments in domains (natural ferromagnetic resonance). Accordingly, the complex permeability of a ferrite can be calculated as a superposition of the two contributions $\tilde{\mu}(f) = 1 + \tilde{\chi}_\mathrm{dw}(f) + \tilde{\chi}_\mathrm{rot}(f)$, namely the domain wall component $\tilde{\chi}_\mathrm{dw}(f)$ and magnetic moment rotation one $\tilde{\chi}_\mathrm{rot}(f)$ [3]:

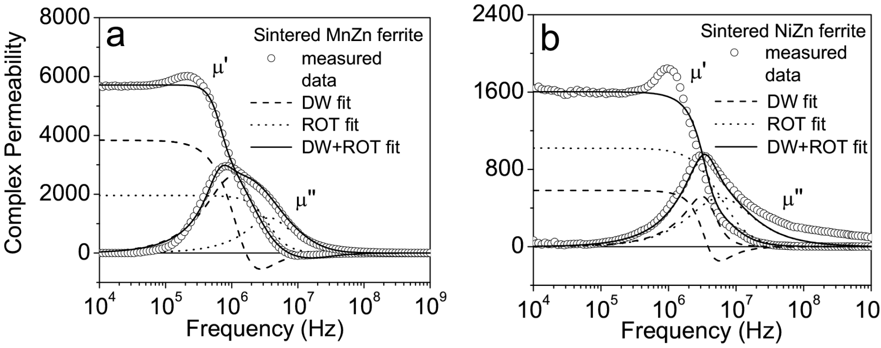

Fig. 1. Complex permeability spectra (measured and calculated data) for sintered ferrites: (a) MnZn and (b) NiZn.

$$\tilde{\chi}_\mathrm{dw}(f) = \frac{\chi_\mathrm{dw} f_\mathrm{dw}^2}{f_\mathrm{dw}^2 - f^2 + \mathrm{j}\frac{\beta}{m_\mathrm{dw}} f}, \tag{1}$$

$$\tilde{\chi}_{\mathrm{rot}}(f) = \frac{(f_{\mathrm{rot}} + \mathrm{j}\alpha f)\chi_{\mathrm{rot}} f_{\mathrm{rot}}}{(f_{\mathrm{rot}} + j\alpha f)^2 - f^2}, \tag{2}$$

where χ_{dw} and χ_{rot} are static (or low frequency) susceptibilities for domain wall (DW) and magnetic moment rotation (ROT) motions, f_{dw} and f_{rot} are resonance frequencies of DW and ROT components, β and α are corresponding damping factors, m_{dw} is the effective mass of wall, and f is the frequency of driving ac magnetic field. Measured permeability spectra for both types of ferrites can be well fitted to the relation for $\tilde{\mu}(f)$ using six fitting parameters χ_{dw}, χ_{rot}, f_{dw}, f_{rot}, β/m_{dw}, and α by means of a nonlinear least squares method. In Fig. 1 we used the numerically computed parameters: $\chi_{\mathrm{dw}} = 3835$, $f_{\mathrm{dw}} = 1.03$ MHz, $\beta/m_{\mathrm{dw}} = 8.98$ MHz, $\chi_{\mathrm{rot}} = 1858$, $f_{\mathrm{rot}} = 5.09$ MHz, $\alpha = 0.97$ for MnZn ferrite, and $\chi_{\mathrm{dw}} = 582$, $f_{\mathrm{dw}} = 3.05$ MHz, $\beta/m_{\mathrm{dw}} = 3.61$ MHz, $\chi_{\mathrm{rot}} = 1019$, $f_{\mathrm{rot}} = 854$ MHz, $\alpha = 148$ for NiZn ferrite. The magnetic moment rotation component is of a relaxation type due to relatively large damping, mainly in NiZn ferrite, and therefore the frequency at which μ'' has its maximum is lower than the resonance f_{rot} frequency. Domain wall contribution is dominant in the MnZn ferrite and rotational one is larger in the NiZn ferrite.

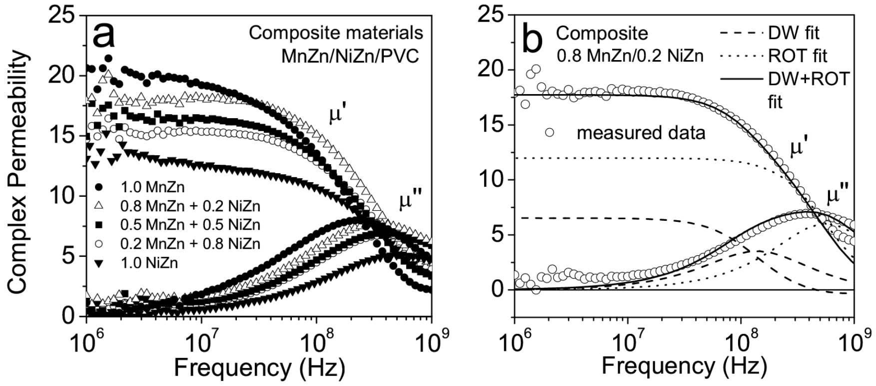

Fig. 2. Complex permeability spectra for triple-component composites: (a) measured values of all samples, and (b) measured and calculated data for selected composite sample.

Figure 2 presents the complex permeability spectra $\tilde{\mu} = \mu' - \mathrm{j}\mu''$ of triple-component composites. The volume fraction ratio of hybrid MnZn:NiZn filler in composite with PVC polymeric matrix was set to 1:0, 0.8:0.2, 0.5:0.5, 0.2:0.8, and 0:1. All the prepared composites had the total volume concentration of hybrid ferrite filler 65 vol%. Unlike sintered MnZn and NiZn ferrites, the triple-component composites show relaxation type of permeability dispersion. In composites, the values of μ' at around 60 MHz become larger than those of the sintered ferrites, and also the critical frequencies f_{c} at which μ'' has a minimum value are higher. In addition, the composites with higher content of MnZn ferrite filler (at the expense of NiZn ferrite) have larger values of μ'. One can state that in triple-component composites the complex permeability changes continuously from MnZn/PVC composite to NiZn/PVC one. Also the critical frequencies f_c change continuously between single ferrite filler composite structures and shifted towards higher values with the increase in NiZn ferrite filler fraction. These features can be explained as follows. Since the composite contains non-magnetic gaps, the demagnetizing field H_D is present along the circumference of the ring core. Hence, the μ' value at low frequencies is reduced by the H_D. On the other hand, the f_rot is related to the anisotropy field H_A antiparallel to the applied ac field and increases with H_D [3, 4]. The value of H_A is also influenced by the change of filler from MnZn to NiZn ferrite: the μ' decreases and f_rot shifts higher due to the contribution of magnetocrystalline anisotropy field to the H_A. As example, the complex permeability spectra of triple-component composite with 0.8 MnZn + 0.2 NiZn filler were decomposed into the DW and ROT components and the following values of dispersion parameters were obtained: $\chi_\mathrm{dw} = 6.1$, $f_\mathrm{dw} = 144.5$ MHz, $\beta/m_\mathrm{dw} = 1.28$ GHz, $\chi_\mathrm{rot} = 11.7$, $f_\mathrm{rot} = 5.1$ GHz, $\alpha = 15.2$. The presented results indicate that the complex permeability can be controlled continuously between two types of composite structure by triple-component composites. These materials seem to be good candidates for such applications as EMI suppressors in portable and wireless electronic equipments [4].

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