Journal of ELECTRICAL ENGINEERING, VOL. 54, NO. 3-4, 2003, 100–103

COMMUNICATIONS

MAGNETIC PROPERTIES OF FERRITE–POLYMER COMPOSITES

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The magnetic properties of ferrite polymer composites prepared with polyvinyl chloride (PVC) or polyvinyl alcohol (PVA) diamagnetic matrices and with an MnZn ferrite powder filler have been investigated. Emphasis was given on the influence of the filler particles morphology (shape) and granulometry (volume fraction or concentration) on the measured magnetic properties. The effective magnetic permeability of a heterogeneous ferrite-polymer composite material was measured at different ferrite concentrations and temperatures. It was observed that the ferrite-polymer composite samples prepared with the PVA matrix have more appropriate magnetic and mechanical properties in comparison with the samples prepared with a PVC diamagnetic matrix.

K e y w o r d s: composite material, polyvinyl chloride, polyvinyl alcohol, MnZn ferrite, nonmagnetic matrix

1 INTRODUCTION

Soft magnetic composite materials produced by embedding soft magnetic particles in a diamagnetic matrix have a significant advantage in comparison with metallic or ceramic materials produced using the established polymer processing techniques. The relationship describing the magnetic properties of a composite material can be modelled using an effective medium theory, magnetic circuit, etc. [1–3, 5].

Composites with magnetic fillers dispersed in a polymer matrix represent a system with physical and chemical properties that are influenced by the ferrite powder fillers and polymer matrix characteristics as well as by the interaction at the ferrite-polymer interface [2]. The study of magneto-composites is encouraged with modelling and verification of some effective parameters. The magneto-polymer composites due to their specific mechanical properties, good formability and toughness (typical for the polymers) allow to prepare the products for magnetic circuits of complicated shapes. At the same time the interesting mechanical, magnetic and chemical properties of these new materials allow to use them in many special applications, *eg* implementation of low profile coils for identification systems, sensors and/or contact-less smart cards.

2 EXPERIMENT

Ferrite cores are familiar as brittle, rigid and bulk components for high-inductance coils and transformers. The performance of such ferrites depends strongly on external influences such as temperature, pressure, and electromagnetic fields of low and high frequency. The ferrite polymer composite samples were prepared in the form of toroids with an outer diameter of 10 mm, inner diameter 6 mm and height 4 mm. As the ferrite filler, a type of MnZn ferrite was used (H40- produced by S+M Components Šumperk, Czech Republic, with Curie temperature $T_C = 130\,^\circ$C, mass density $4.8\,\mathrm{g\,cm^{-3}}$, and intrinsic permeability $4300 \pm 30\,\%$). The used manganese-zinc ferrite consists of 37 wt.% MnO, 12 wt.% ZnO and 51 wt.% $Fe_2\vee_3$ (composition $\approx Mn_{0.37}Zn_{0.57}Fe_{2.06}\vee_4$). Magnetic particles prepared by mechanical granulation of sintered MnZn ferrite with two fractions, (25-40) + (250-315) μm with the ratio of fractions $1 : 1.5$, were used for the preparation of ferrite polymer composites. As diamagnetic polymer matrices, the PVA and PVC polymers were used. The used mass concentrations of the ferrite filler in the composite samples with the PVA matrix were $\kappa = 93$, 90 and 85 wt.% (*ie* volume concentrations $\kappa_v = 73$, 66, 56 vol.%) and with the PVC matrix $\kappa = 85$ and 80 wt.% (*ie* volume concentration $\kappa_v = 64$ and 56 vol.%).

3 MEASUREMENT

As we have already mentioned, a special form was made for the prepared samples of toroidal shape. The form was filled with the prepared ferrite — PVC polymer composite mixtures and then heat-treated for about 20 minutes at a temperature of $120\,^\circ$C. Interesting results were achieved as for the photoelectric properties of PVA

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ISSN 1335-3632

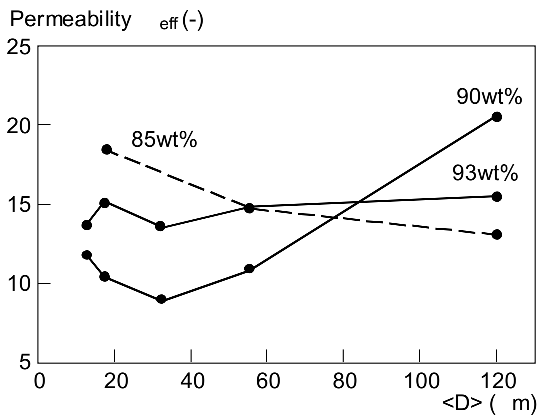

Fig. 1. The dependence of effective permeability μ_{eff} on the statistical mean value of grain size $< D >$

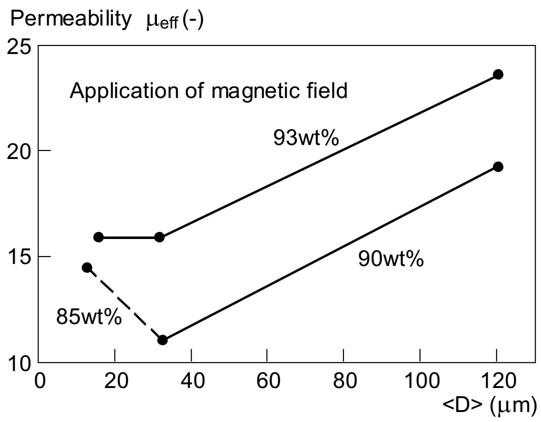

Fig. 2. The dependence of effective permeability μ_{eff} on the statistical mean value of grain size $< D >$ with application of a DC magnetic field.

polymer [4]. The measured points of the samples with drying temperatures of 80 °C were lying on a common straight line in contrary to the samples with a drying temperature of 120 °C which exhibited a higher sensitivity. This behaviour may be explained by assuming that at the drying temperature of 80 °C and 100 °C apparently the same amount of PVA molecules was converted into polyvinyl, whereas at 120 °C obviously a larger number of molecules took part in the conversion [5]. From this it follows that the composite with the PVA matrix was heat-treated at a temperature of about 80 °C or less for a longer time than previously. The effective magnetic permeability data of the prepared ferrite-polymer composites with PVA and PVC matrices were then measured at room temperature using an integration method [5]. The temperature dependence of the magnetic susceptibility was measured by means of a fully automated system equipped with a kappa bridge KLY-2, the temperature was changed between about 20 and 150 °C. The measurements were performed on ferrite powder samples. The apparatus and method used to obtain the measured data were described elsewhere [3, 5].

4 RESULTS AND DISCUSSION

It is realistic to assume that ferrite-polymer composites with inorganic magnetic fillers immersed in a PVA polymer diamagnetic matrix can innovate the properties of these new types of magnetic materials. Based on the presented experimental results and their evaluation it was shown that macroscopic magnetic parameters of the ferrite-polymer composites depend on complicated physical and chemical conditions. The analysis of the physical conditions of composites preparation shows the influence on their magnetic parameters.

Figure 1 shows the jumping dependence of the effective magnetic permeability versus statistical mean value of the grain size corresponding to different mass concentrations. From the comparison of the measured dependences in Fig. 1 it follows that the composite sample with 90 wt.% concentration of ferrite filler and with a PVA diamagnetic matrix had the highest value of the effective magnetic permeability $\mu_{\mathrm{eff}} \sim 21$ at the highest value of grain size dimension $\langle D \rangle$, in comparison with other samples. Figure 2 shows the similar behaviour of μ_{eff} versus $\langle D \rangle$. In this case, the composite samples with the PVA matrix were additionally heat-treated in the presence of a DC magnetic field (with a flux density of about 60 mT). The directions of the toroidal plane and the DC magnetic field were perpendicular to each other.

In the presented results, a measurable orientation effect is observed of the applied field on the ferrite particles during the process of preparation of the composite samples. This effect is visible in a significant increase of the effective magnetic permeability for the 85 wt.% sample in comparison with the 93 wt.% sample. From this it can be concluded that on the interface ferrite particles-polymer matrix there is probably a reinforced interaction between the ferrite particles and the thin diamagnetic polymer layer. The essence of this effect consists in a direct super exchange interaction between unpaired electron spins through —OH . . . O — hydrogen bond.

Similar measurements were carried out with the same ferrite filler but using the PVC polymer as a diamagnetic matrix. The temperature dependences of the magnetic flux Φ measured on both composite samples with PVA and PVC polymers and with different concentrations of the ferrite filler but with the same ferrite particle granulometry (particle size) can be seen in Fig. 3. The values of the magnetic flux decrease with temperature for both composite samples.

Figure 4 shows the dependence of the effective permeability μ_{eff} on the statistical mean value of the grain size for samples with higher concentrations of the ferrite filler. However, the maximum value of μ_{eff} is significantly

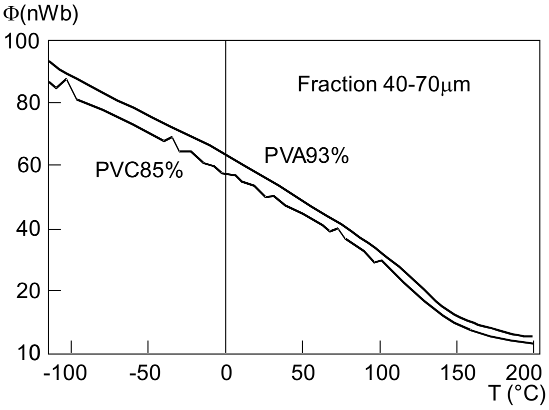

Fig. 3. Temperature dependence of the magnetic flux for ferrite-polymer composites

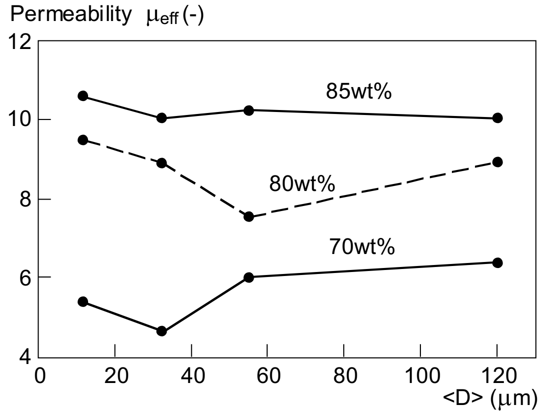

Fig. 4. The dependence of effective permeability μ_{eff} on the statistical mean value of grain size $< D >$ for samples with higher concentrations of the ferrite filler

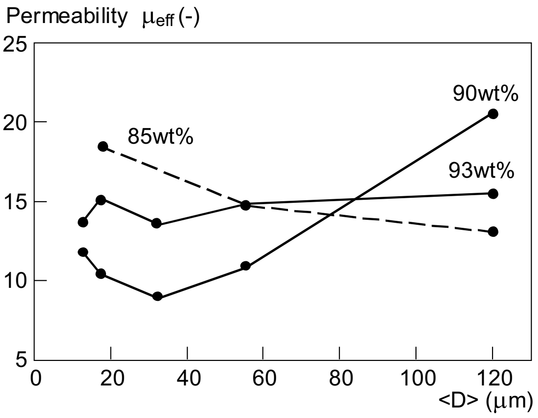

Fig. 5. The dependence of effective permeability μ_{eff} on the statistical mean value of grain size $< D >$

lower in comparison with the composite sample with PVA polymer matrix for the same particle ferrite fraction.

Figure 5 shows the smooth dependence of μ_{eff} on the statistical mean value of the grain size for different weight concentrations of ferrite in PVC. This clearly differs from the results presented in Fig. 1 for composite samples with a PVA polymer matrix. It should be noted that in the case of some ferrite concentrations there are some localized states in the polymer composites. The randomness in the interaction energy of the available spin states may arise from structural distortions around of ferrite grains covered by the polymer thin films. At higher ferrite concentrations, the states are extended and the dispersion disappears.

5 CONCLUSIONS

Calculations of the magnetic permeability or electrical conductivity of a composite material from the known parameters of the pure phases (ferrite and polymer) are connected with some specific problems [1–3, 5]. The problems connected with the preparation of ferrite-polymer composites and with the measurement of their effective magnetic parameters were presented and discussed elsewhere [2–3, 5]. We have prepared two groups of MnZn ferrite-polymer composite samples with PVA and PVC polymers. In preparation of ferrite-polymer composites with tailored properties, a suitable ferrite should be chosen with an optimal particle size (granulometry) and shape (morphology). The values of the magnetic parameters seem to be more influenced by the size effects than by the shape of filler particles at a given conditions of samples preparation, [3]. Moreover, the particle size effect, the filler concentration effect as well as the mutual orientation of the ferrite particles in the matrix can also play an important role. The polymer diamagnetic matrix should have convenient mechanical, viscoelastic and chemical properties. The compatibility of the matrix with the magnetic particles is a significant factor connected with their size and desired homogeneous distribution in the composite. It was observed that mechanical and magnetic properties of the ferrite-polymer composite samples prepared with the PVA matrix have better magnetic and mechanical properties than the samples with the PVC polymer.

Further experimental work will be carried out to estimate the effects and consequences of magnetic interactions between the magnetic particles as well as between the magnetic particles and diamagnetic polymer matrix in order to get a composite structure with much more better magnetic, mechanical and chemical properties.

Acknowledgement

This work was supported by No. 1/7610/20 grant of VEGA Agency of the Slovak Republic.

References

[1] PATERSON, J.H.—DEVINE, R.—PHELPS, A.D.R.: Complex Permeability of Soft Magnetic Ferrite/Polyester Resin Com-

posites at Frequencies above 1 MHz, J. of Magn. Magn. Mat. **196-197** (1999)), 394–396.

[2] SLÁMA, J. et al : Magnetic Permeability Study of Composite Magnetopolymers, J. of Magn. Magn. Mat. **196-197** (1999), 359–361.

[3] DOSOUDIL, R. : Investigation of Magnetic Properties of Composite Magnetic Materials, Proc. of the Conf. "Magnetic Measurements 2000", Prague, Czech Republic, Sept. 11–12 (2000), 71–74.

[4] RECHSEL, A.—GORLICH, P. : Infrared Physics **3** (1963), 229–239.

[5] DOSOUDIL, R. : Modelling and Optimisation of Magnetic Properties of Polymer Magnetic Materials, PhD Thesis, Dept. of Electromagnetic Theory, STU, Bratislava, 2000.

Received 28 June 2002

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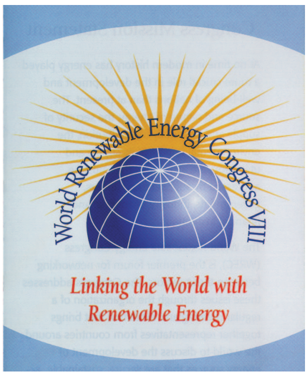

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