RELATIVE MAGNETIC PERMEABILITY OF POLYMERIC COMPOSITES WITH HYBRID PARTICULATE FILLERS

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INTRODUCTION

Polymeric composites with magnetic properties can be prepared by combining a magnetic filler material with a polymeric host i.e., a binder. This can be accomplished by using polymer processing techniques which afford several advantages over the traditional techniques for shaping ceramic and metallic magnets. Polymeric magnets of complex shapes can be manufactured with high production rates and low costs by utilizing polymer processing operations including injection molding. However, such technologies have been mainly limited to the fabrication of permanent magnets [1-7].

BACKGROUND

It has been shown that at low loading levels the magnetic permeability of granular composites \( \mu_r(\phi) \), increases linearly with the volume fraction, \( \phi \), of the filler [8]:

\[
\mu_r(\phi) = 1 + A \phi
\]

(1)

Where \( A \) is a coefficient which depends on the magnetic properties of the filler, its shape, and volume fraction. For example, for spherical particles the demagnetization factor is \( N=1/3 \), so that Eqn. (1) becomes

\[
\mu_r(\phi) = 1 + 3\phi
\]

(2)

As the loading level, \( \phi \), is increased, the magnetic permeability value deviates significantly from this linear behavior and becomes a non-linear function of the volume fraction [8, 9]. Based on previous experimental studies [8, 9] the non-linear dependence of the relative permeability on the volume fraction can be approximated by using a quadratic function:

\[
\mu_r(\phi) = 1 + B \phi^2
\]

(3)

A hybrid magnetic composite may be defined as a composite containing two or more fillers with different magnetic properties, sizes, size distributions and shapes. The relative permeability of a hybrid composite consisting of similar particles (size, size distributions, and shape) differing only in magnetic properties, may be expected to obey a simple quadratic additivity rule:

\[
\mu_r(\phi_1,\phi_2) = 1 + B_1 \phi_1^2 + B_2 \phi_2^2
\]

(4)

It may be possible to achieve higher magnetic permeability values for hybrid composites by using dissimilar shapes and sizes of filler particles. For instance, Hashin and Shtrikman [11] give bounds of the magnetic permeability for a densely packed composite consisting of coated spheres. By using a variational approach they were able to determine the bounds to be:

\[
\mu_1 + \frac{3\phi_2 \mu_2 (\mu_2 - \mu_1)}{3\mu_1 + \phi_1 (\mu_2 - \mu_1)} \leq \mu_e
\]

\[
\leq \mu_2 + \frac{3\phi_1 \mu_1 (\mu_1 - \mu_2)}{3\mu_2 + \phi_2 (\mu_1 - \mu_2)}
\]

(5)

provided \( \mu_2 \geq \mu_1 \). The lower bound of the inequality corresponds to the situation where component 2 with a relative permeability of \( \mu_2 \) is coated with component 1 with a relative magnetic permeability of \( \mu_1 \). The upper bound corresponds to a composite where component 1 is coated with component 2. By tailoring a composite with flexible flakes and spherical particles it may be possible to achieve the upper bound of the effective magnetic permeability. If the flexible flakes are large in comparison to the spherical particles and have a greater magnetic permeability, then the flakes can act as if they are "coating" the spherical particles, resulting in an enhanced magnetic permeability value.

The objective of this study was to determine whether the permeability of a magnetic composite would be enhanced with the incorporation of hybrid fillers. In addition, the functional dependence of the volume fractions of each filler was studied to determine their effects on the magnetic permeability of the composites.

EXPERIMENTAL

Materials

Three magnetic materials were chosen for this study: a ferrite powder, an amorphous metal ribbon.

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cut into flakes, and a ferromagnetic metal powder. The ferrite powder, code 28, was supplied by D. M. Steward MFG Co., Chattanooga, TN. The 28 material is a fully reacted nickel-zinc spinel ferrite with an average particle diameter of 60 μm. The particle size distribution was narrow and the particles ranged in size from 50 μm to 70 μm. The amorphous ribbon, trade name Metglas "Met" 2705M, was obtained from Allied-Signal, Morristown, NJ. It has a chemical composition of 69% cobalt, 12% boron, 12% silicon, 4% nickel, 2% molybdenum, and 1% nickel. The Metglas was supplied as a continuous ribbon of width 1" and thickness 0.8 mil. The ribbon was cut into flakes with an aspect ratio of 1250 (ratio of largest dimension to thickness). The metallic ferromagnetic filler was HyMu 800 procured from Carpenter Technology Corp., Reading, PA. HyMu 800 has a composition of 80% nickel, 5% molybdenum, 0.5% manganese, 0.15% silicon, 0.10% carbon and the balance iron. It was supplied in spherical powder form at -100 mesh. The polymer matrix used in this study was low density polyethylene (LDPE), Petrothene PEV 007 available from USI Chemicals, Cincinnati, OH.

For the evaluation of hybrid composites two composites systems were used. One was a combination of PE/Met/NI2Zn ferrite and the other was a mixture of PE/Met/HyMu. Both were prepared at various loading levels. The PE/Met/NI2Zn system study was conducted as a two factor design experiment with five levels of the Metglas 2705M concentration and three levels of the Ni2Zn concentration. The PE/Met/HyMu system was run at four levels of the Metglas 2705M concentration and two levels of the HyMu concentration.

The experimental setup and material preparation techniques for mixing, determination of particle attrition, molding of ASTM specimens, and characterization of magnetic permeability values which we used here, were the same as those reported earlier [12].

RESULTS AND DISCUSSION

Figs. 1-3 show the results of magnetic relative permeability of single filler composite systems. The dashed lines in Figs. 1 and 2 represent a curve fit to the data at low loading levels, i.e. 10% and 25% loadings, to Eqn. (1). The solid lines in Figs. 1-3 represent the regression analysis, best fit to Eqn. (3), of the data for the entire range of volume loading levels. The most striking feature about Figs. 1 and 2 is the relative permeability increase at high loading levels, as indicated by the difference between the solid and dashed lines. Also note the difference between the two types of fillers. The Ni2Zn filler produces coefficients of A=4.8 and B=20.1, while the HyMu filler has coefficients of A=7.0 and B=41.1. The differences in coefficients are due to the differences in intrinsic magnetic quality, size, shape, and size distributions between the two filler particles [9].

The relative permeability of composites containing Metglas flakes exhibit a different trend than those observed with the powders as shown in Fig. 3. The relative permeability values of the composite increase with increasing volume loading level of the Metglas. However, the rate of change of the permeability with increasing concentration decreases here, as opposed to the powder samples which increase with an increasing rate (Figs. 1 and 2). This may be due to the fact that at high loading levels the mixing process produces high stresses which can degrade the intrinsic permeability as well as the aspect ratio of the flakes.

Figs. 4 and 5 are the three dimensional surface plots of the two composite systems with the hybrid fillers. Fig. 4 shows the relative permeability of the PE/Met/NI2Zn composite system as a function of the volume fractions of Metglas and Ni2Zn. The regression equation for this surface is:

$$\mu_r = 1 - 40.7\phi_1 + 233\phi_2 + 103.3\phi_1^2 + 576\phi_1\phi_2 + 330.4\phi_2^2$$  \hspace{1cm} (6)

where $\phi_1$ is the volume fraction of the Ni2Zn filler and $\phi_2$ is the volume fraction of the Metglas. It is evident from Fig. 4 that there is a significant interaction between the two fillers which manifests itself by the term $576\phi_1\phi_2$, which represents the strength of the interaction between the two fillers. The correlation coefficient for Eqn. (6) is 0.9911. The regression model accounts for over 98.2% of the variation in the data. A plot of the distribution of residuals reveals a normal behavior as shown in Fig. 6. Fig. 7 is a plot of the best fit vs. the experimental values. Figs. 6 and 7 combined with the correlation coefficient and the p-level of the parameters indicate that the proposed model of Eqn. (6) is indeed a valid one.

The relative permeability values of the PE/Met/HyMu composite samples are shown in Fig. 5. Again the enhancement is apparent with the use of hybrid fillers. The regression equation for this surface is:

$$\mu_r = 1 + 25.8\phi_1 + 277.2\phi_2 + 50.7\phi_1^2 + 788.5\phi_1\phi_2 + 286.4\phi_2^2$$  \hspace{1cm} (7)

where $\phi_1$ is the volume fraction of the HyMu filler and $\phi_2$ is the volume fraction of the Metglas. Again there is a large interaction term between the fillers, 788$\phi_1\phi_2$, which enhances the permeability.

Another way to illustrate the enhancement of relative permeability values with hybrid fillers is to compare the actual values against those predicted by Eqn. (4), i.e., the direct sum of the single filler composite systems that compose the hybrid system.
This comparison shows that the measured values are always greater than those predicted by the simple quadratic additivity rule provided by Eqn. (4). This is illustrated in Fig. 8 where the permeability values of hybrid composite samples containing 50% NiZn ferrite and varying amounts of Metglas are plotted. Also plotted are the direct sums of the single filler composite systems which make up the hybrid system. These results show that the enhancement of the relative permeability of hybrid composites is greater than that can be accomplished by using high loading levels of the individual fillers for magnetic composites prepared with conventional polymer processing methods i.e., dispersive mixing of the magnetic filler particles with a polymeric binder and molding.

CONCLUSIONS

Two hybrid composite systems were prepared at various filler concentrations and were characterized in terms of their relative magnetic permeability values. It was shown that the use of hybrid composites with symmetric and asymmetric particulates produces a synergistic enhancement of the relative magnetic permeability values when compared to the magnetic permeability values of the individual filler systems which comprise the hybrid system. This type of synergism has hitherto not been reported in the literature and generates relative permeability values which are greater than any that have been reported in the literature.

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REFERENCES


KEY WORDS:
MAGNETIC, PERMEABILITY, COMPOSITE
Fig. 1. Relative permeability of NiZn composites as a function of filler concentration.

Fig. 3. Relative permeability of Metglas 2705M composites as a function of Meiglas concentration.

Fig. 2. Relative permeability of HyMu composites as a function of HyMu concentration.

Fig. 4. Relative permeability of the hybrid NiZn/Metglas 2705M composites.