

---

# Relative Magnetic Permeability of Injection Molded Composites as Affected from the Flow Induced Orientation of Ferromagnetic Particles

By Thomas Fiske, Halit S. Gokturk, Rahmi Yazici and Dilhan M. Kalyon

Highly Filled Materials Institute  
Hoboken , NJ 07030

---

## INTRODUCTION

Polymer composites which exhibit relative magnetic permeability values which exceed one can be prepared by mixing a magnetic filler material with a polymer binder. The magnetic permeability of composites is influenced by magnetic quality of the filler, its concentration and particle shape and size [1-5]. These microstructural variables are complex and do not allow the application of various available theoretical approaches [6, 7] to link magnetic permeability of a composite to the content and various characteristics of the magnetic powders. Thus, currently only experimental studies can provide the necessary guidance to proper selection of gating and mold design to tailor composites with targeted magnetic properties. Clearly, the orientation of ferromagnetic fibers upon the thermo-mechanical history of the injection molding process will play a key role in the development of the relative magnetic permeability value of the injection molded composite. A study similar in scope and aimed to link electrical properties of composites to the characteristics of conductive fillers was carried-out by Weber and Kamal [8].

The objective of this study was to experimentally examine the effects of processing on the orientation distributions of ferromagnetic fibers in injection molded composites and to elucidate the effects of the resultant fiber orientation distributions on the magnetic properties of the injection moldings. Furthermore, the effects of the concentration and the aspect ratio of the ferromagnetic particles were also investigated.

## EXPERIMENTAL

### *Material*

The polymer matrix used in this study was a commercial grade of high density polyethylene (HDPE), Quantum LR 734. Several different ferromagnetic materials including a NiZn ferrite powder, a nickel powder with spherical particles and two batches of nickel fibers were used as fillers.

The nickel powder with spherical particles was supplied by Johnson Matthey of Ward Hill, MA. The average particle size of the nickel powder was 110  $\mu\text{m}$ . Two grades of nickel fibers were procured from National Standard of Mishawaka, Indiana. The two batches of Ni fibers had a diameter of 20  $\mu\text{m}$  but differed in length. The shorter fibers had a length of 260  $\mu\text{m}$  (aspect ratio = 13) and the longer fibers had a length of 1000  $\mu\text{m}$  (aspect ratio = 50).

### *Injection Molding*

The mixing of the binder polymer with the reinforcing fillers was carried-out by using a Haake torque rheometer. The specimens for magnetic property characterization were made by an injection ram molding apparatus which was converted from an Instron Floor Tester and an Instron capillary rheometer. Two molds were designed and manufactured; one to produce straight cylindrical specimens with a length over diameter ratio of 10 and the second to mold disk-shaped specimens with a diameter over the thickness ratio of 8.

### ***Sample Preparation***

The cylindrical injection moldings were 0.006 m in diameter and 0.0635 m in length. The disk shaped injection molded samples were 0.006 m in thickness and 0.0508 m in diameter. Toroidal specimens with an inside diameter of 0.0417 m and an outside diameter of 0.0508 m were cut from the disk shaped injection molded samples.

### ***Characterization of Magnetic Properties***

The magnetic properties of cylindrical injection molded specimens were characterized with a HP 4284A LCR meter interfaced to a MacIntosh IIfx by an IEEE 488 card. Attached to this was an air coil in which the cylindrical samples could be slipped in and out without detachment.

The magnetic property measurements of the toroidal samples were carried-out using a HP function generator model 200CDR, a Carver Power Amplifier PT-1250, a Hitachi Oscilloscope 425, and an EG&G Lock-In Amplifier 5209 [2]. The toroidal samples were uniformly wrapped with two sets of wire windings. One functioned as the primary coil and the other served as the secondary or pick-up coil.

### ***Calibration of Two Magnetic Measurement Setups***

In order to ensure that the two magnetic measurement setups were consistent with each other, samples were prepared from HDPE/NiZn ferrite. The NiZn ferrite particles are spherical in shape and thus their orientation in different flow geometries is not a factor. Both cylindrical and toroidal samples were prepared at the 50% volume loading level. The cylindrical and toroidal samples generated relative permeability values of  $5.83 \pm 0.01$  and  $5.77 \pm 0.2$ , respectively, indicating that there is no significant difference between the magnetic behavior of the NiZn ferrite samples molded into disk and cylindrical shapes or the two experimental measurement setups.

### ***Determination of Fiber Orientation***

Fiber orientation distributions of the injection molded samples were analyzed by applying x-ray microradiography on the transverse cross-sections of the molded samples. Microradiographs were taken with soft x-rays ( $< 104$  eV) using a GE-GA5 unit and fine-grained x-ray films [10, 11]. The images of the microradiographs were digitized and further processed using Adobe Photoshop and Scan Maker software. Orientation distributions of individual fibers with respect to reference axes were characterized employing the digitized images using NIH Image/ppc processing software.

Subsequent to the measurement of the orientation of the individual fibers, the orientation distribution and the orientation function (J) of the fibers were determined for each case. As defined by Stein [12], and utilized by Yaguchi [13], the fiber orientation function (J) for two dimensional distribution (in x-y plane) is given by:

$$J = \frac{2}{\pi} \int_{-\pi/2}^{\pi/2} \cos^2 \theta q(\theta) d(\theta) - 1 \quad (1)$$

where  $\theta$  is the fiber orientation angle and  $q(\theta)$  is the distribution of the fiber orientation angles. The orientation function J varies such that:  $J = 0$ , for random orientation;  $J = +1$ , for unidirectional orientation in x direction ( $\theta = 0$ ); and  $J = -1$  for unidirectional orientation in y direction ( $\theta = \pi/2$ ).

In this study, discrete  $\theta$  values were obtained for each fiber in the microradiographs and the orientation function was determined by numerical integration. The accuracy of the NIH-Image based image analysis tools and the subsequent calculations in determining J values were tested using computer simulated fiber distributions prepared elsewhere [13]. These control

measurements were within 2% of the simulated J values.

## RESULTS AND DISCUSSION

### *Fiber Orientation of the Toroidal and Cylindrical Profiles*

Typical micrographs of cylindrical and toroidal samples with the asymmetric Ni particles are shown at two different magnifications in Figs. 1a and 1b, respectively. It can be seen from the micrographs that the fibers in the cylindrical samples are highly aligned in the direction of flow. This is also the same direction of the applied magnetic field. The micrographs also show that the fibers in the toroidal samples are oriented in a random fashion.

The typical orientation distribution of the fibers as obtained by image analysis in the cylindrical and toroidal samples are shown in Figs. 1c and 1d, respectively. The orientation function (J) of the toroidal samples varied from -.16 to +.05 depending on the region of analysis. For the toroidal samples a value of J = -.16 indicates a relatively low fiber alignment in the injection molding flow direction (y) which is perpendicular to the applied magnetic field direction (x).

The cylindrical samples on the other hand exhibited relatively high degree of fiber orientation with the values of the orientation function ranging from 0.30 to 0.63.

Table 1 shows that for the HDPE/260 (fiber length 260  $\mu\text{m}$ , aspect ratio of 13) composites, the "overall" fiber orientation decreases as the fiber volume percent increases under injection molding conditions (temperature and injection rate) which were kept constant. This is probably due to increased interactions and friction between the fibers and the limited length of the gate used, which was not long enough to induce high alignment in the samples with high volume percent of fibers. The decrease in the orientation function due to high fiber volume percent in HDPE/260, however, was moderate.

The J values, for example, dropped from .61 to .53, when the fiber content increased (doubled) from 10% volume to 20% volume. However, in the 20% volume sample with J = .53, there are still more fibers aligned than in the 10% volume sample with J = .61. Thus, the directional values of the physical properties such as permeability along the fiber alignment axis in the 20% volume sample are still expected to be higher than the 10% volume sample.

The samples with long Ni fibers (HDPE/1000) exhibit less fiber alignment i.e., smaller J values under identical molding conditions. This effect was primarily due to the bending of these longer fibers, especially in those cases where the fiber volume percentages were low.

The effect of the gate size was very significant on the fiber orientation distribution and the fiber orientation function, J, values. The J value for 20% volume fiber samples decreased from .53 to .32 when the gate diameter decreased from 3.2 mm to 1.7 mm. This was most likely due to the different mold-filling mechanisms caused by the differences in gate design. The smaller gate might cause a jetting effect which will produce misalignment of the fibers.

The injection rate also affected the fiber orientations. For the HDPE/260 composites at 20% volume fill level, the J value increased from .53 to .63 when the injection rate was increased from the  $6 \times 10^{-8}$  m<sup>3</sup>/s to  $1.7 \times 10^{-7}$  m<sup>3</sup>/s.

### *Implications of the Fiber Orientation with Respect to the Applied Magnetic Field*

The implication of the applied magnetic field being longitudinal or transverse to the fibers should become clear by examining the simple case of a single fiber subjected to a uniform magnetic field. The magnetic fiber is magnetized by the applied external magnetic field H, inducing a magnetic field which opposes the original field. This field is called the demagnetizing field, H<sub>d</sub>. Therefore, for a non-zero demagnetization field, the effective field, H<sub>eff</sub>, will always be less than the applied field because:

$$H_{\text{eff}} = H - H_d \quad (2)$$

The apparent permeability of the fiber not only depends on the quality of the magnetic material but also on its shape. Bozorth [9] gives the relationship between the apparent permeability  $\mu'$ , the permeability  $\mu$ , and the shape or demagnetization factor N as:

$$\mu' = \frac{1}{\frac{1}{\mu} + \frac{N}{4\pi}} \quad (3)$$

For a fiber (or cylinder) N depends on the ratio of length to diameter, decreasing with increasing length to diameter ratio and also on the orientation of the fiber with respect to the applied field. The apparent permeability decreases with decreasing effective aspect ratio [9]. The effective aspect ratio is defined as the ratio of the dimension of the fiber parallel to the applied field to that of the dimension of the fiber transverse to the applied field. The effective aspect ratios of our fibers are 0.077 and 0.02 for the fiber lengths of 260 μm and 1000 μm fibers, respectively, when the fibers are oriented in the transverse direction with respect to the applied magnetic field. At both of these effective aspect ratios the demagnetization factor, N, is greater than 0.3 and the apparent permeability value approaches to a value of one. Consequently, the fibers aligned transversely to the applied field contribute very little to the magnetization of the composite. Therefore, it can be concluded that composites with fibers oriented in the direction of the applied magnetic field will exhibit a higher apparent permeability than those aligned in the transverse direction.

Figure 2 displays the magnetic permeability values of injection molded cylinders and disks for the two composite systems with different volume fractions of fibers. The confidence intervals determined according to Student's t-distribution are included. In both cases, it can be seen that the cylindrical samples indeed exhibit higher permeability values than the disk shaped samples suggesting a higher degree of fiber orientation for cylindrical samples as indeed found to be the case.

### ***Effect of Injection Rate and Gate Diameter on Permeability***

Figures 3, 4 show the effects of injection rate on the permeability of the composites. The mold and melt temperatures were held constant. Figure 3 summarizes the results of the HDPE/260 composite cylinders. It can be seen that an increase in injection volume flow rate improved the permeability values for the 20 and 30% filled composites. On the other hand, Figure 4 shows that the increase in injection rate had no effect on the permeability values of the HDPE/1000 cylindrical composites.

Gates with diameters of 0.067" (1.7 mm) and 0.125" (3.2 mm) were used to examine the effects they would have on orientation of fibers and ultimately the permeability of the composites. For these experiments the cylindrical mold was used in conjunction with the HDPE/260 composite at an injection rate of  $1.5 \times 10^{-7}$  m<sup>3</sup>/s and a mold temperature of 155oC. The difference in permeability values for the different gates is significant. The smaller gate produced samples which exhibit lower permeability values than those produced with the larger gate. The increased magnetic permeability is associated directly with the greater degree of fiber orientation.

The effects of the fiber orientation distribution function, J, on permeability were best revealed on those samples with constant fiber concentration but which were under different injection rate and gate diameter conditions. The data taken from such measurements on HDPE/260 samples with 260 μm long fibers and fill level of 20 volume percent ( $\phi = 0.20$ ) are presented in Table 2. The orientation function, J, varies between zero for spherical Ni particles to 0.63 for fiber incorporated samples injection molded at the injection volumetric flow rate of  $1.5 \times 10^{-7}$  m<sup>3</sup>/s. The experimental relative magnetic permeability versus fiber orientation function (J) behavior obeys a power law relationship:

$$\mu' = \mu_0 + 4 J^2 \quad (4)$$

where,  $\mu_0$  is the relative magnetic permeability of composite with spherical Ni particles.

In this case, if the extrapolation to higher J values is permitted the maximum magnetic permeability value, which is expected for fibers oriented uniaxially in the direction of the magnetic field, is around 6.

## **CONCLUSIONS**

Magnetic properties of composites are influenced by concentration, fiber length, and orientation of the ferromagnetic particles which are incorporated into a polymeric binder. Since fibers are anisotropic their preferential orientation will impart anisotropy in the composite material properties. Anisotropic magnetic composites were produced by injection molding of suspensions consisting of a high viscosity polymeric binder and various fillers in two custom designed molds, which produced differences in fiber orientation distributions. The magnetic properties of the composites were characterized in terms of their relative permeability values and related to their microstructure. It was determined that composites with a higher degree of orientation generate higher permeability values. Quantitative data which link orientation distribution functions to magnetic permeability values of composites







