EFFECTS OF HEAT TREATMENT ON THE MAGNETIC PROPERTIES OF POLYMER-BOUND IORN PARTICLE CORES

by

M. Namkung, B. Wincheski, and R. G. Bryant
NASA Langley Research Center

Presented at the
7th Joint Magnetism and Magnetic Materials Conference
January 6-9, 1998
San Francisco, California

EFFECTS OF HEAT TREATMENT ON THE MAGNETIC PROPERTIES OF POLYMER-BOUND IRON PARTICLE CORES

M. Namkung, B. Wincheski, R. G. Bryant, NASA Langley Research Center, Hampton, VA 23681 and A. Buchman, Old Dominion University, Norfolk, VA 23529

Spherical iron particles of three different size distributions, 6 - 10 μ in diameter, 100 mesh and 30 - 80 mesh, were mixed with 2.0 wt. % of soluble imide and compression molded at 300°C under 131 MPa. Post fabrication heat treatments were performed at 960°C for 6 hours resulting in a significant enhancement of the permeability in low field region for all the specimens except for the one made of 30 - 80 mesh particles. The rate of core loss of these specimens at a magnetic induction of 5 kG measured up to 1 kHz shows a noticeable increase after heat treatment which, along with the permeability enhancement, can be explained by the coalescence of particles forming a network of conductivity paths in the specimens. The scanning electron micrographs taken for the 6 - 10 μ particle specimens show no evidence of heat treatment-induced grain growth. The untreated specimens show a very weak f^2-dependence of the core loss which clearly indicates a negligible contribution from the eddy current loss. In particular, an almost perfect linearity was found in the frequency dependence of the core loss of the untreated specimen made of 100 mesh iron particles.

I. INTRODUCTION

Our previous study has shown that the permeability of polymer-bound cores of spherical iron particles of 6 - 10 μ can be enhanced by a factor of 30 - 40 through a simple post-fabrication heat treatment at 960°C for 6 hours[1]. The low field portion of the magnetization curves of these heat treated specimens showed clear evidence of contributions from reversible and irreversible domain wall motions which were absent in the untreated specimens. The most probable reason for such a heat treatment-induced permeability enhancement is the coalescence of iron particles forming a network connecting bare metal particles since the temperature of heat treatment was within the range of the typical sintering temperature of metallic particles.

Unlike the case of sintered powder metallurgy components, the presence of polyimide in these core specimens does not allow a clear visualization of inter-particle contacts in the scanning electron micrographs. An alternate way of investigating the formation of such a network structure is to measure the core loss which is sensitive to the range of eddy current flow. Hence, the purpose of the present study is to systematically investigate the effect of heat treatment on the core loss for specimens of different size distributions since it is one of the major factors controlling the structure[2] upon which the magnetic properties of interest depend.

II. EXPERIMENTS

Spherical iron particles of 6 - 10 μ in diameter, 30 - 80 mesh (0.59 - 0.17 mm in diameter) and 100 mesh (diameter less than 0.14 mm) were mixed with 2.0 wt. % of high temperature soluble thermoplastic adhesive known as LaRC™-SI and molded into cubic blocks under 131 MPa of external compressive load at 300°C for 30 minutes[3]. Two picture frame specimens of identical geometry having a cross-section of 6.35 x 6.35 mm^2, and outer edges of lengths 12.7 mm and 19.05 mm were cut from each block for core loss measurements. One of the

two picture frame specimens of each particle size distribution was heat treated at 960°C for 6 hours. The rate of heating and cooling before and after heat treatment in the furnace was approximately 0.5°C/min. The core loss was measured at the magnetic induction amplitude of 5 kG up to 1 kHz except the heat treated specimens of 6 - 10 μ and 30 - 80 mesh particles which were tested to 500 Hz. Two identical square rod specimens with the same cross-section of 6.35 x 6.35 mm^2 and 19.05 mm length were prepared from each block for the characterization of permeability, mass density and conductivity before and after an identical heat treatment.

III. RESULTS AND DISCUSSION

Fig. 1 summarizes the magnetization curves of untreated and heat treated specimens of the three particle size distributions. The magnetization of the three untreated specimens increases very slowly but maintains its slope in the high field region, i. e., roughly 170 - 500 Oe, which is steeper than that of heat treated specimens of 6 - 10 μ and 100 mesh particles. This indicates that a certain level of domain wall motion is present in the untreated specimens in this field region. This is readily explainable since most iron particles are coated and well isolated in these untreated specimens providing strong barriers impeding the domain wall movement such that it takes a stronger driving force to complete the domain wall motion[4]. The effect of heat treatment on the 6 -10 μ particle specimen is seen to be dramatic. A close examination of the initial stage reveals the domain wall mobility for reversible motion is even higher in the heat treated specimen of 100 mesh particles than it is in that of 6 - 10 μ particles but the overall induction being much lower in the former is not immediately explainable. It is evident that the effect of heat treatment on the DC magnetic properties of the 30 - 80 mesh particle specimens is merely to increase the magnetization.

Speed and Elman showed that pre-fabrication annealing of the iron particles increase the maximum permeability by a factor of 2 - 5[4] which is presumably due to relieving of the surface residual stress in each particle that tends to limit the magnetization activity. The temperature of molding in the present work, i. e., 300°C, is not high enough for residual stress relief. The temperature of post-fabrication heat treatment, 960°C, is in the range of typical sintering temperature and it is safely assumed that the individual particles are stress-relieved and coalesced with the neighboring ones forming a channel which enhances the domain wall mobility. Hence, the post-fabrication heat treatment provides two major effects enhancing the initial magnetization processes observed in the 6 - 10 μ and 100 mesh specimen. An indirect evidence of coalescence of particles is the brittle-ductile transition introduced by the heat treatment as reported in our previous study[1]. The behavior of initial magnetization of the heat treated 30 - 80 mesh specimen is not readily explainable and the rupture test of this specimen, which is yet to be performed, may provide useful information.

Table I. summarizes the density and resistivity of the test specimens measured before and after heat treatment. The density of the 6 - 10 μ specimen in this work was reduced by heat treatment which is a trend that is consistent with that reported in the previous work[1]. Such a reduction in the density is thought to be due to the bubbling up of the binder material, i. e., polyimide, creating voids but why it is limited to 6 - 10 μ particles only is yet to be studied further. The resistivity of the 6 - 10 μ particle specimen is seen to decrease by a factor of 10 due to heat treatment. Such a reduction in the resistivity can be clearly explained. The individual particles were originally coated with polyimide. During the fabrication process, at 300°C under 131 MPa of compressive load, however, a certain fraction of particles rub against each other establishing bare metal to metal contacts forming an initial stage of network through the specimen. The heat treatment causes the neighboring particles in direct contact to coalesce into

each other to drastically reduce the resistivity in the conduction network. In addition, the carbonization of polyimide, which begins to occur around 500°C, transforms the insulating layers among the particles into a conducting matrix.

The density and DC magnetic properties are closely related to the particle size distribution and the fabrication condition. For certain simplified conditions, it is possible to numerically predict both properties[5]. The presence of the binder, however, causes significant complication and no particular correlation can be found. Nevertheless, all the heat treated specimens display consistently low resistivity values. It is to be noted that the resistivity of the untreated 100 mesh particle specimen is much higher than those of the other specimens. Fig. 2 shows the core loss of heat treated and untreated 6 - 10 μ particle specimens. It is immediately clear that the nonlinear frequency dependence is very weak in the loss curve of the untreated specimen, whereas a noticeable nonlinearity is present in the curve of the heat treated specimen. The simplest model of frequency dependence of core loss includes two terms; first and second order terms of frequency. The former is due to the hysteresis loss and to the eddy current loss assuming the absence of the domain structure. Fig. 3 shows a nonlinear curve of core loss per kg-cycle as a function of frequency of the heat treated 6 - 10 μ particle specimen displaying the effect of anomalous eddy current loss caused by the presence of domain wall motion[4].

Fig. 4 shows the core loss of heat treated and untreated specimens of 100 mesh particles. The core loss of the untreated specimen is seen to be a remarkably linear function of frequency which is a clear evidence of the absence of eddy current loss in this material. When fitted to a second order polynomial, the coefficients of the second and first order of frequency terms are 5.8149×10^{-8} W-sec^2/gm and 1.4975×10^{-6} W-sec/gm, respectively. Compared to 1.269×10^{-6} W-sec^2/gm and 2.187×10^{-6} W-sec/gm for the curve of heat treated specimen, the linearity in the untreated specimen can be considered nearly perfect. The lack of

eddy current loss is a strong evidence of the absence of macroscopic eddy current flow in the material since the effect of microscopic eddy current contribution is linearly proportional to the frequency[6]. Fig. 5 shows the results of core loss measurements in 30 - 80 mesh particle specimens. The difference in core loss between heat treated and untreated states in these specimens is as pronounced as in the specimens of 6 - 10 μ particles.

It is evident that the core loss is noticeably lower in the untreated specimens than in the heat treated specimens as the frequency increases. Apparently, the major contributing factor for this is the resistivity. The resistivity of the untreated 100 mesh iron particle specimen, in particular, is unusually high and it contributes to the elimination of eddy current loss. At the same time, the magnetization curve of this specimen is comparable to those of the two other untreated specimens. As high resistivity and magnetization are essential requirements for a transformer core material, the untreated 100 mesh iron particle specimen provides a clear direction for the further development.

IV. CONCLUSION

The effect of heat treatment on the magnetization and core loss properties were investigated in polymer-bound iron particle cores of three different particles size distributions. It was found that the iron particle size distribution strongly affects the heat treatment-induced enhancement of magnetization as well as the frequency dependence of core loss which was investigated up to 1 kHz. The absence of eddy current loss, which was attributed to high resistivity, was observed in the untreated 100 mesh iron particle specimen.

[1]M. Namkung, R. G. Bryant, B. Wincheski and A. Buchman, J. Appl. Phys. **81**, 4112 (1997).

[2] R. M. German, *Sintering Theory and Practice* (John-Wiley & Sons, New York, NY, 1996).

[3] R. G. Bryant, M. Namkung, B. Wincheski, J. P. Fulton and R. L. Fox, NASA Patent Disclosure, LAR 15463-1-SB (995).

[4] B. D. Cullity, *Introduction to Magnetic Materials* (Addison-Wesley, Reading, MA, 1972).

[5] H. How and C. Vittoria, J. Appl. Phys. **70**, 5915 (1991).

[6] R. M. Bozorth, *Ferromagnetism* (Van Nostrand Co, New York, NY, 1952).

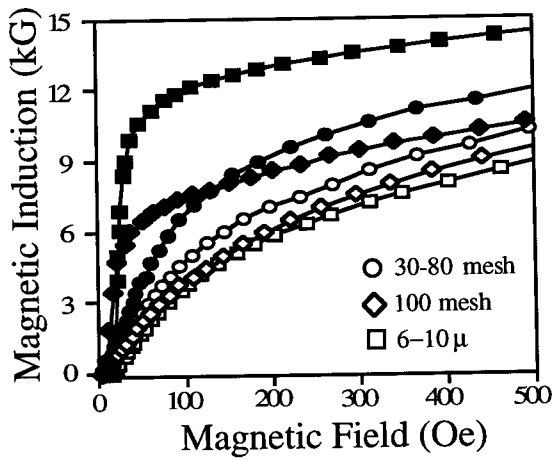

Fig. 1. Magnetization curves of heat treated (filled symbols) and untreated (unfilled symbols) specimens of three different iron particle size distributions

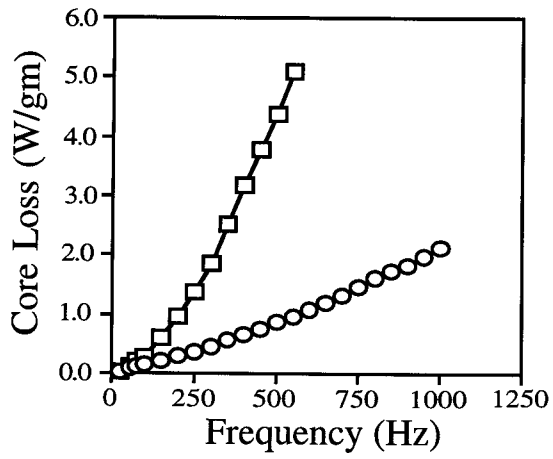

Fig. 2. Frequency dependent core loss/unit mass of heat treated (square) and untreated (circle) specimens of 6 - 10 μ iron particle cores

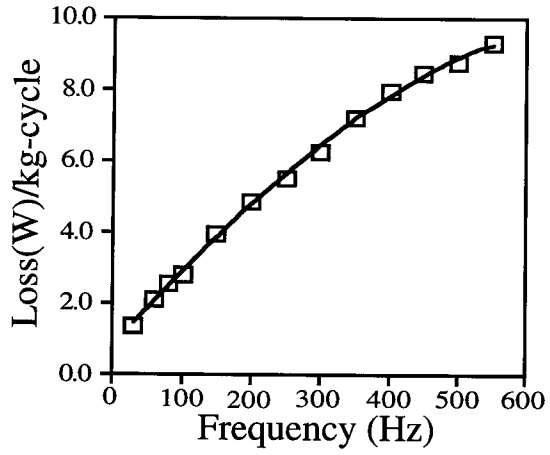

Fig. 3. Nonlinearity in core loss/unit mass-cycle in 6 - 10 μ iron particle core indicating the effect of anomalous eddy current loss due to a nonuniform distribution of magnetization across the cross-section

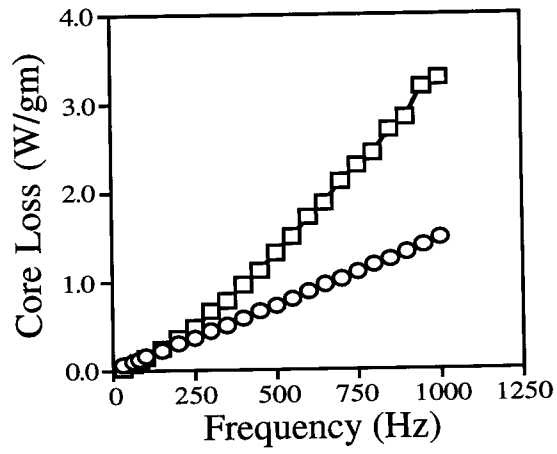

Fig. 4. Frequency dependent core loss/unit mass of heat treated (square) and untreated (circle) specimens of 100 mesh particles displaying a nearly perfect linearity in the latter due to the absence of eddy current loss

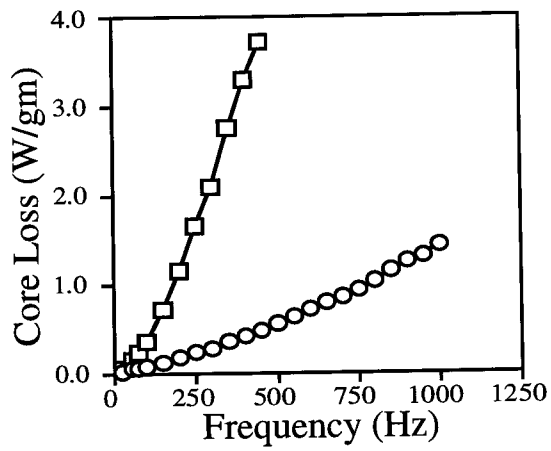

Fig. 5. Frequency dependent core loss/unit mass of heat treated (square) and untreated (circle) specimens of 30 - 80 mesh iron prticles

TABLE I. Density and resistivity of test specimens

Specimens[a]	Density[b](gm/cc)	Resistivity (Ω-cm)
6 -10 μ	6.3[c]/6.22[d]	0.467/0.043
100 mesh	5.88/7.41	1.686/0.057
30 - 80 mesh	5.75/6.92	0.161/0.064

[a]Specimen designation based on the iron particle sizes

[b]Density measured using the weight of an object measured immediately after it was submerged in water (It changes as water fills up the voids. See Ref. 1 for details)

[c]Values for untreated specimens

[d]Values for heat treated specimens