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DISCLAIMER

MAGNETOSTRICTIVE PROPERTIES OF RARE EARTH-IRON LAVES PHASE MATERIALS PREPARED BY POWDER METALLURGY TECHNIQUES*

Manoochehr Malekzadeh and Milton R. Pickus**

ABSTRACT

A powder metallurgical approach has been utilized for preparation of highly magnetostrictive rare earth-iron Laves phase compounds. The results of dilatometric studies indicate that the liquid-phase sintering kinetics are in reasonable agreement with the concept of a phase boundary reaction as the rate-limiting factor. Magnetic powder orientation prior to sintering is found to improve magnetostriction of these compounds substantially.

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I. INTRODUCTION

A number of studies on the huge room temperature magnetostriction of rare earth-iron Laves phases have indicated that these materials are of particular interest in a variety of technological applications such as sonar systems and non-destructive testing in the field of reactor safety [1, 2, 3]. Recently, some powder metallurgical techniques have been developed to fabricate these hard and brittle intermetallic compounds into suitable sizes and shapes [4, 5, 6]. A notable advantage of the powder approach is its amenability to magnetic powder orientation. Considerable static magnetostriction enhancement has been observed in compacts sintered from magnetically aligned powders [7]. In this paper, we present the results of some further sintering studies as well as the dynamic characteristics of the powder metallurgically prepared samples.

II. EXPERIMENTAL PROCEDURE

The compound preparation consisted of arc-melting the elemental rare earth and iron metals, all of 99.9% purity, under a Zr gettered argon atmosphere. The arc-melted buttons were subsequently crushed and then pulverized under toluene in a steel planetary ball milling machine for 20 minutes. The resulting 15-35 μm powder was rinsed with acetone and vacuum-dried. Rubber tubing, 0.6 cm i.d. and 3.8 cm long, was manually filled with powder. The packing efficiency was improved by the application of vibratory agitation. The tubing was sealed with rubber plugs and placed inside a perforated steel tube for isostatic compaction in the range of 50-70 kg/mm^2.

For samples in which a magnetic orientation was desired, the manually filled rubber tubing was first subjected to an alternating field of 1000 Oe peak-to-peak, at frequencies up to 500 Hz, superimposed on a DC field of 20 Koe. A field of this type produced sufficient vibration to facilitate orientation of the powder. While in the magnetic field, the particles were locked in position by hand-applied end compression with a plunger prior to isostatic compaction.

The cold pressed samples were subsequently wrapped in Ta foils and sintered in a dynamic vacuum of 3×10^{-6} mm Hg at 1130° - 1150° C, for durations up to 48 hours. The true densities of the sintered samples were determined by a fluid displacement technique with appropriate temperature corrections. Permeability measurements were carried out by the fluxmetric technique [8], where corrections were made for the demagnetizing field and the cross sectional area difference between the sample and the pick-up coil. The evaluation of the dynamic magnetostriction of the samples was accomplished by a strain gauge technique. This technique is desirable because of its compactness and accuracy when used with a temperature and magnetic field compensating circuitry. In order to compensate for the temperature and magnetoresistive effects, for each of the active strain gauges on the sample, a corresponding "dummy" gauge of the same type and lot number was mounted on a piece of arc-melted and homogenized YFe_2. The choice of YFe_2 was based on its being an isostructural non-magnetostrictive ($\lambda_S < 2 \times 10^{-6}$) compound with a thermal coefficient of expansion in the same range as that of other $R\text{-}Fe_2$ compounds. The bias field was supplied by a short solenoid magnet, powered by a 180 kw source. It was established that the magnetic field of this magnet over the entire length of the sample was uniform within 4 %. The sample probe was placed accurately at the center of a solenoidal winding and then the assembly was secured in the bore of the short solenoid magnet. The circuitry shown in Figure 1 provided alternating fields up to $\pm$ 500 oe at various frequencies. The calibration of the field was done in terms of alternating currents passing through the solenoidal winding. The strain gauges were balanced by means of Q plug-in units and a dual beam oscilloscope. The dynamic strains were recorded on a light-sensitive paper by an oscillograph. To minimize any temperature rise in the sample, the alternating fields were applied only for short periods (up to 10 seconds). The frequency of the dynamic field was chosen at 160 Hz ($\omega \simeq 1000$) because it was low enough to reduce the eddy-current loss and far from the harmonics of 60 Hz.

The linear shrinkage measurements during the liquid-phase sintering were carried out by means of dilatometry. Cold pressed samples of approximately 6 mm diameter and 10 mm length were placed in the chamber of the dilatometer which had a vacuum of better than 5 x 10^{-6} mm Hg during the operation. It required less than 20 seconds for the furnace to reach a set point temperature. The temperature was controlled within $\pm$ 5°C by the feedback from the output of a sensitive thermocouple welded to the sample.

III. INITIAL SINTERING KINETICS OF RFe_2 ALLOYS

Some sintering studies of RFe_2 compounds have been previously reported [5, 6]. R. G. Johnson et al [6] have studied the shrinkage

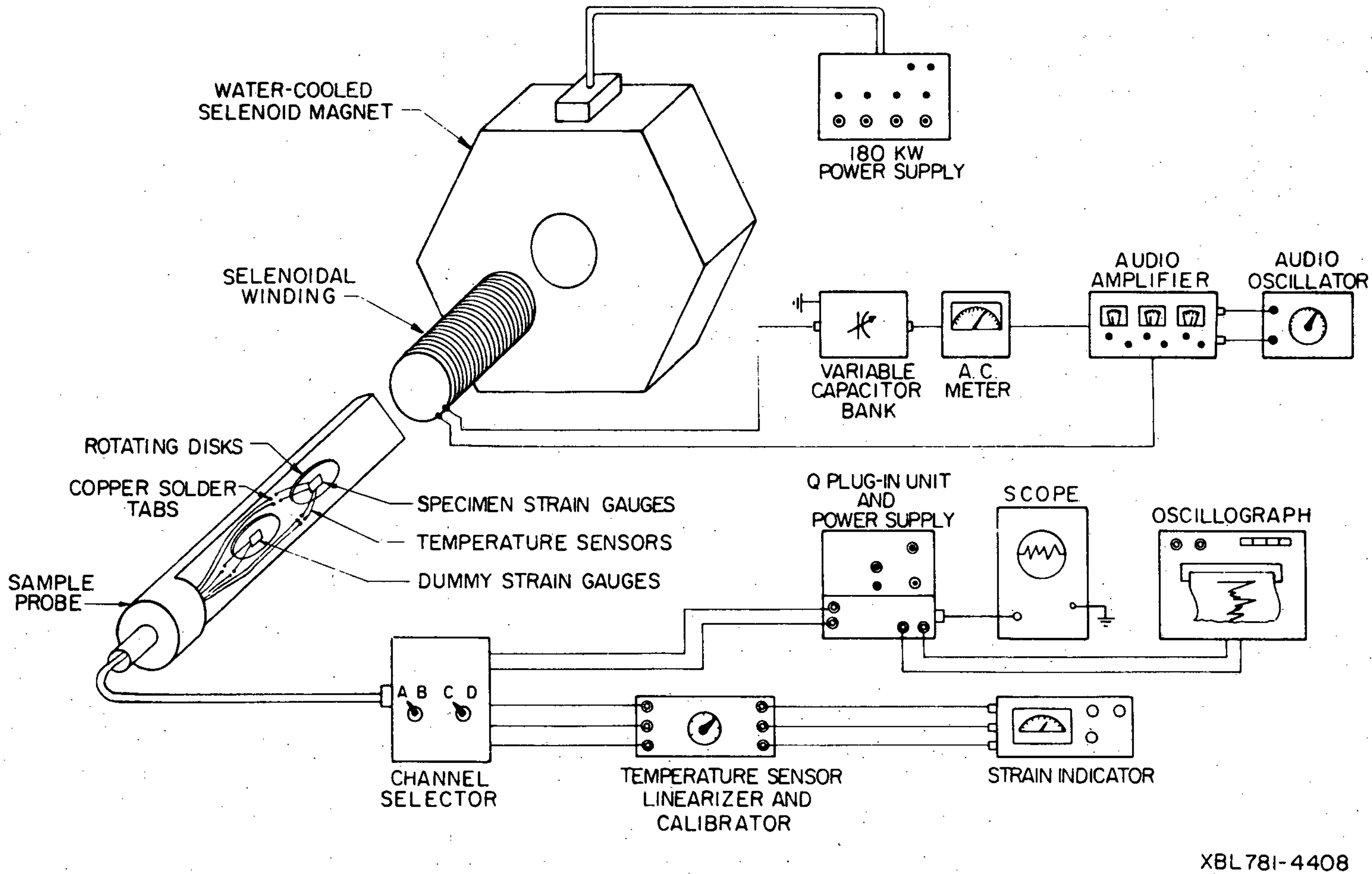

Figure 1. Experimental set-up for dyanamic magnetostriction measurements.

behavior of $DyFe_2$ alloys. Their results showed long incubation periods for stoichiometric $DyFe_2$ and for $DyFe_2$ + 10% Dy-Fe eutectic sintered at 1150°C. They were able, however, to eliminate the incubation period by sintering under 10 mm HF gas.

Using dilatometry, we have looked into the initial sintering kinetics of the samples prepared by the sintering approach described in Reference [5]. In this approach, the iron deficient alloy (containing 55% Fe) becomes liquid and therefore, we expect the liquid-phase sintering kinetics to prevail during the initial stages. The first stage of liquid-phase sintering involves a relatively rapid densification, due to particle rearrangement. We have used Kingery's [9] thermodynamic analysis of the densification rate during solution-precipitation (second stage) to identify the rate controlling mass-transport process. Using this analysis, the isothermal dependence of linear shrinkage on time is expressed by

$$(\Delta L/L_o)_T \quad \propto \quad t^{1/3}$$

for a process rate controlled by diffusion or

$$(\Delta L/L_o)_T \quad \propto \quad t^{1/2}$$

for a phase boundary reaction as the rate controlling step. The above equations suggest that data of linear shrinkage versus time should indicate the sintering mechanism involved.

The dilatometric results of an isothermal shrinkage of a $Tb_{0.3}Dy_{0.7}Fe_2$ sample is plotted in the form of log $(\Delta L/L_o)$ versus log t in Figure 2. The value of L_o was taken as the initial length of the cold "green"

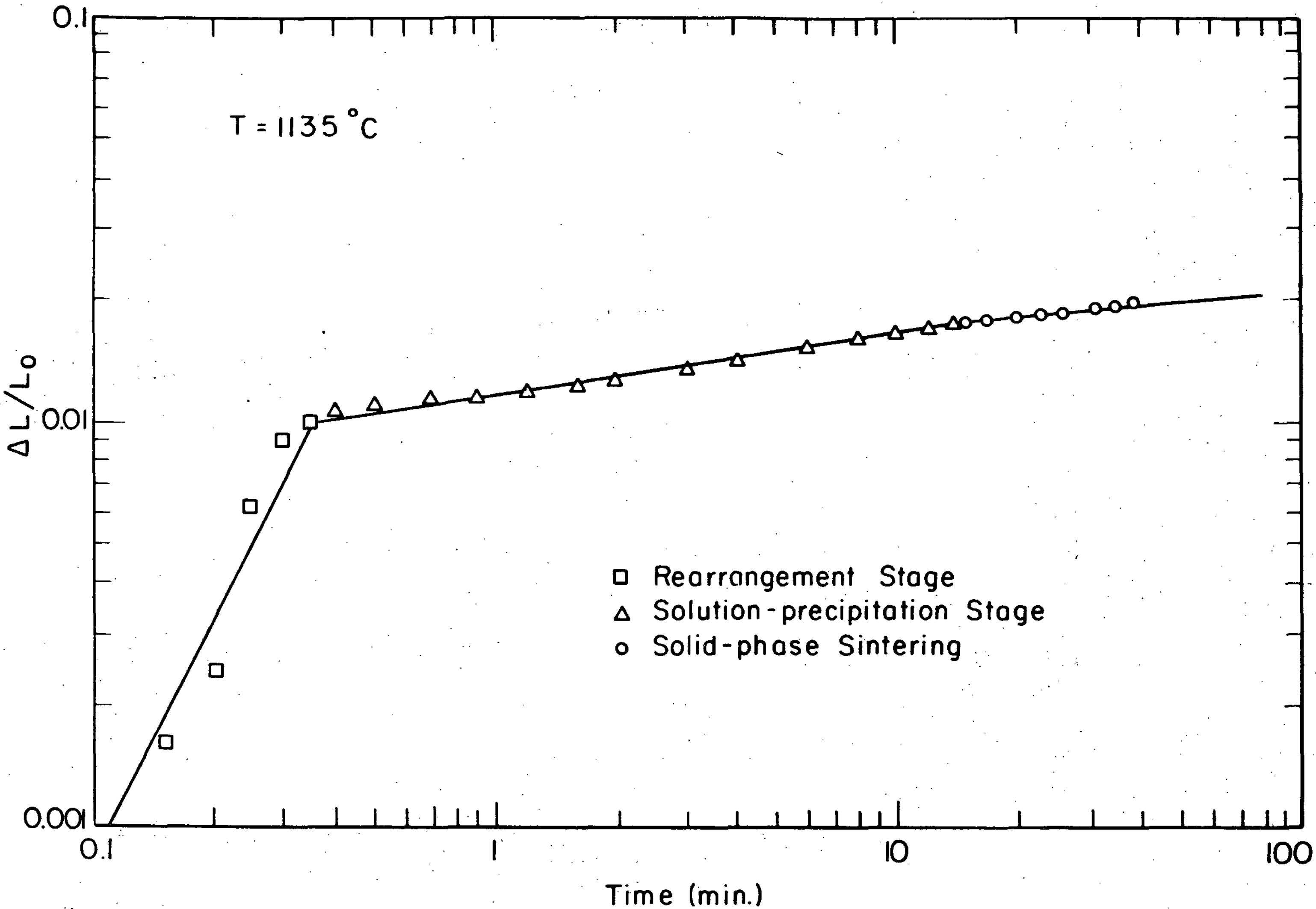

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Figure 2. Isothermal time dependence of shrinkage behavior of a $Tb_{0.3}Dy_{0.7}Fe_2$ sample.

compact corrected for thermal expansion at the sintering temperature. As the data show, a substantial amount of densification is rapidly reached by the rearrangement process, due to the presence of a large amount of liquid phase at the sintering temperature. The slope of the line representing the solution-precipitation stage is much lower than predicted by the rate-controlling mechanisms described by Kingery. A. L. Prill et al [10] have found, however, that large amounts of shrinkage during the rearrangement stage will result in an apparent low time exponent of the second stage linear shrinkage and erroneous identification of the rate-controlling process. Figure 3 shows the replot of the data which were reanalyzed, taking into account the specimen length and time at the end of the rearrangement stage. The slopes of lines through the replotted data are close to 0.5 (a least square fit through the data points corresponding to 1135°C sintering temperature gave a slope of 0.48 with a correlation coefficient above 0.99). A time exponent of 0.5 for the linear shrinkage isotherms shows the mechanism of sintering is rate limited by a phase boundary reaction process rather than diffusion in the liquid phase. The solution-precipitation stage, which starts at the first break of the curve in Figure 2, contributes to increasing the grain size. Grain boundaries have been proven to be effective barriers to domain wall motion. Globus et al [11] have given a relation between the grain size and the permeability in the form of :

$$\mu - 1 = \frac{8\ \pi M_s}{H} \quad \frac{\Delta V}{V} \quad \propto r$$

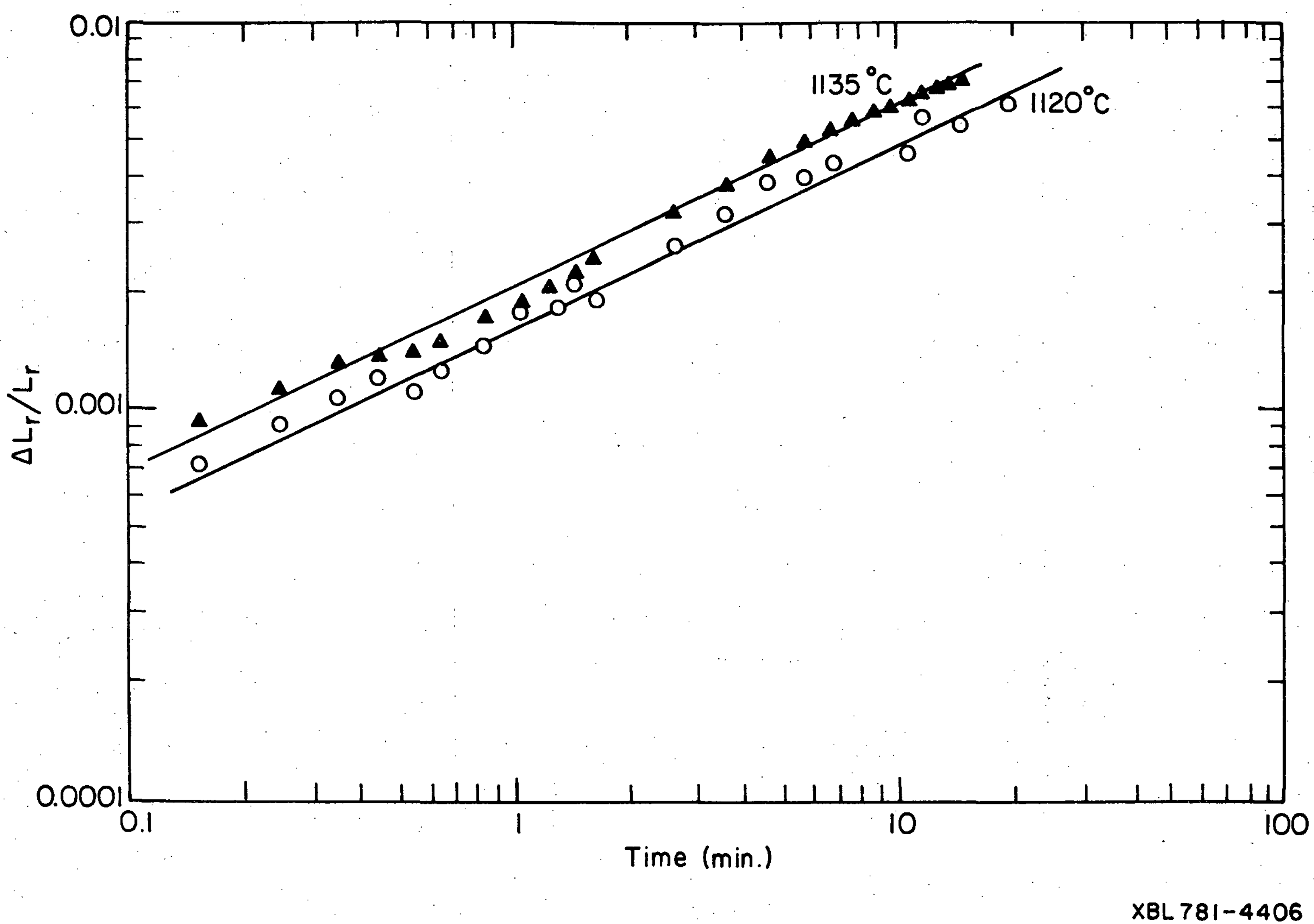

Figure 3. Time and length corrected isotherms of liquid phase sintered $Tb_{0.3}Dy_{0.3}Fe_2$.

where:

M_s	=	saturation magnetization
r	=	radius of a spherical crystallite grain
$\frac{\Delta V}{V}$	=	volume change of the domain per unit volume
μ	=	permeability.

The large grain size induced by this method of liquid-phase sintering, which gives densities as high as 97% of the theoretical density, leads also to products with a higher permeability.

IV. EFFECTS OF MAGNETIC POWDER ALIGNMENT

In the course of preparation of the magnetically aligned samples, the liquid-phase sintering approach was not applied in an attempt to reduce any possible loss of the alignment. The measured values of the static permeability of these samples are shown in Figure 4. These values are much higher than those known for the randomly oriented material. The higher permeability will be beneficial for cores working at high induction levels. The sharp increase of permeability and the occurence of permeability maxima at lower fields, as the density increases, are due to the reduction of internal demagnetizing fields of the pores and the drop in coercivity of the material.

Static and dynamic magnetostrictions of a magnetically aligned solid-state sintered compact is shown in Figure 5. At constant frequency, the magnitude of the dynamic strain attained was roughly proportional to the magnitude of the applied alternating field until the dynamic field had a value approximately equal to the bias field. As the alternating field exceeded this value, some frequency distortion of the dynamic

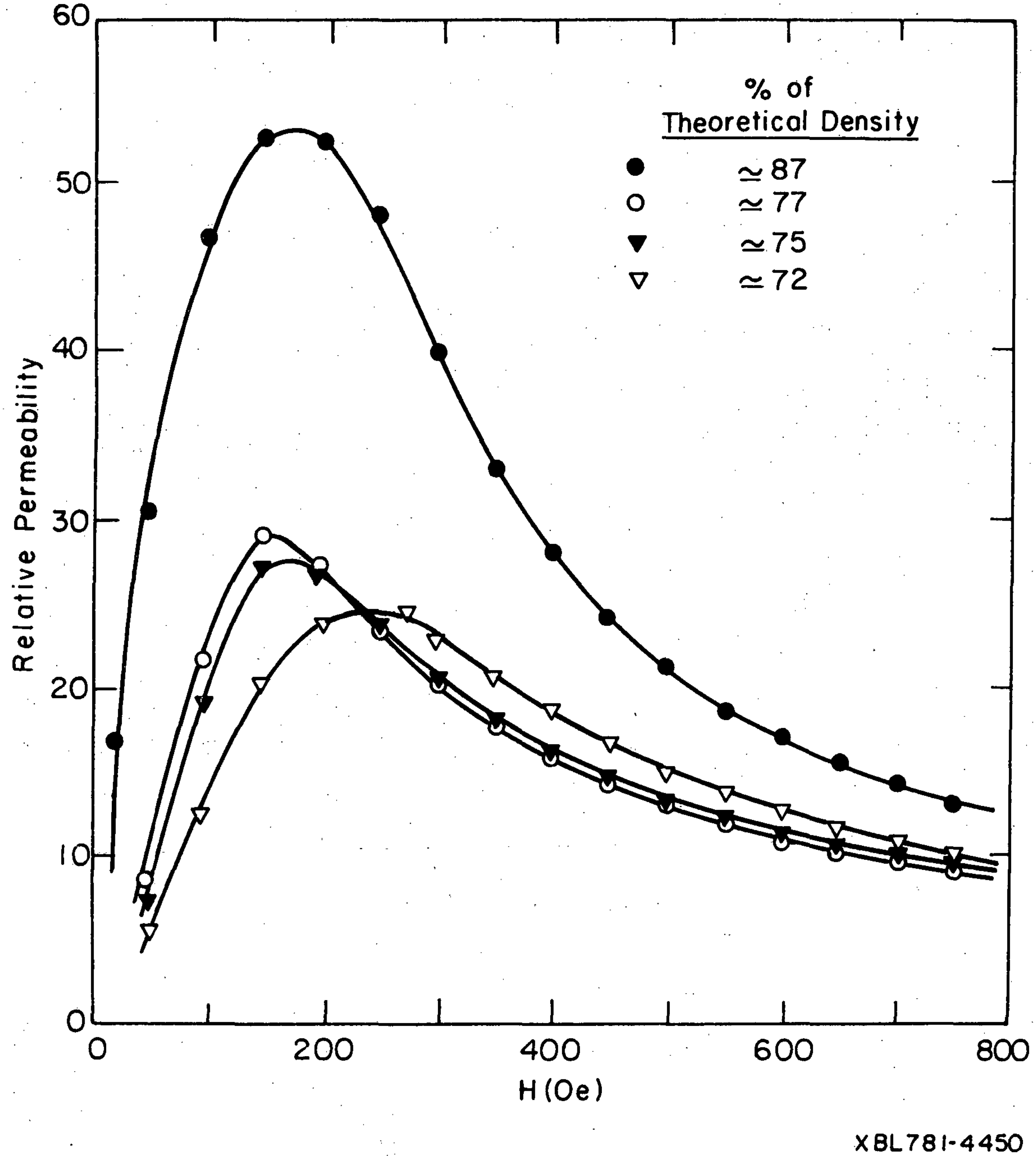

Figure 4. Effects of alignment and density on the relative permeability of $Tb_{0.3}Dy_{0.3}Fe_2$.

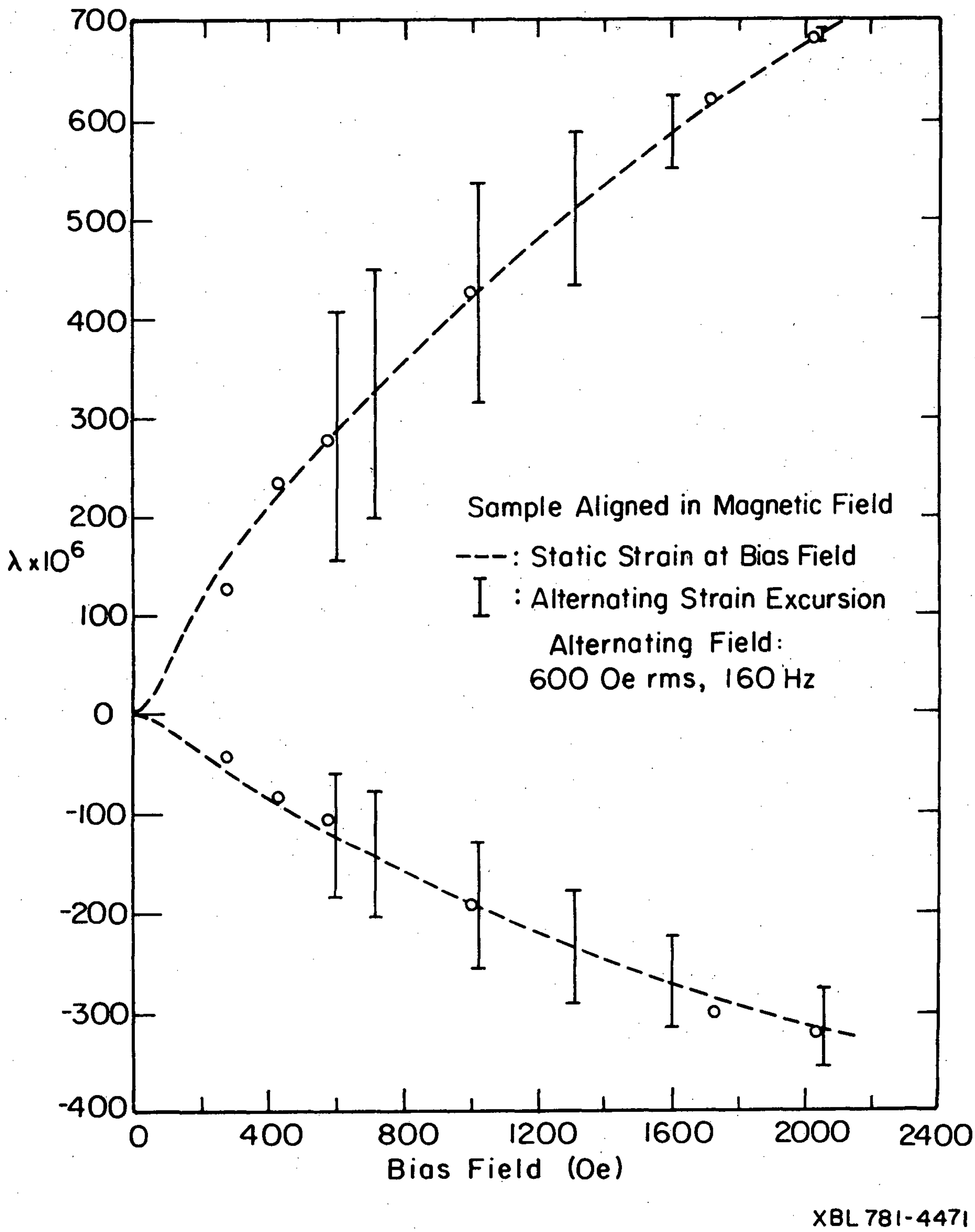

Figure 5. Dynamic magnetostrictions observed in a magnetically aligned sample of $Tb_{0.3}Dy_{0.3}Fe_2$.

strain began to occur. Comparison of the data with Figure 6 indicates that, with respect to both static and dynamic magnetostrains, the textured sample is superior to the highly dense liquid-phase sintered material. This superiority can be due to the effect of grain orientation on reducing the microstresses set up at the grain boundaries of these highly magnetostrictive materials. In the course of magnetostriction measurements, some temperature rise in the material was detected after each short period of the test. The temperature-rise rate was consistently faster with frequency increase of the applied alternating field. This is attributed to the tendency of the eddy-current loss to increase due to the formation of more widely spaced domain walls in the material as the texture improves, although the hysteresis loss should decrease.

The huge magnetostrains observed are indicative of the advantage of these materials in applications where a high power broad-band low frequency source is desirable. The magnetic powder alignment approach, however, can also be extended to the preparation of powder rolled thin plates and laminates with an improved reduction in the eddy-current loss. Preliminary data indicate that even better magnetostrictive properties can be achieved by combining the use of magnetically alligned powder with liquid-phase sintering.

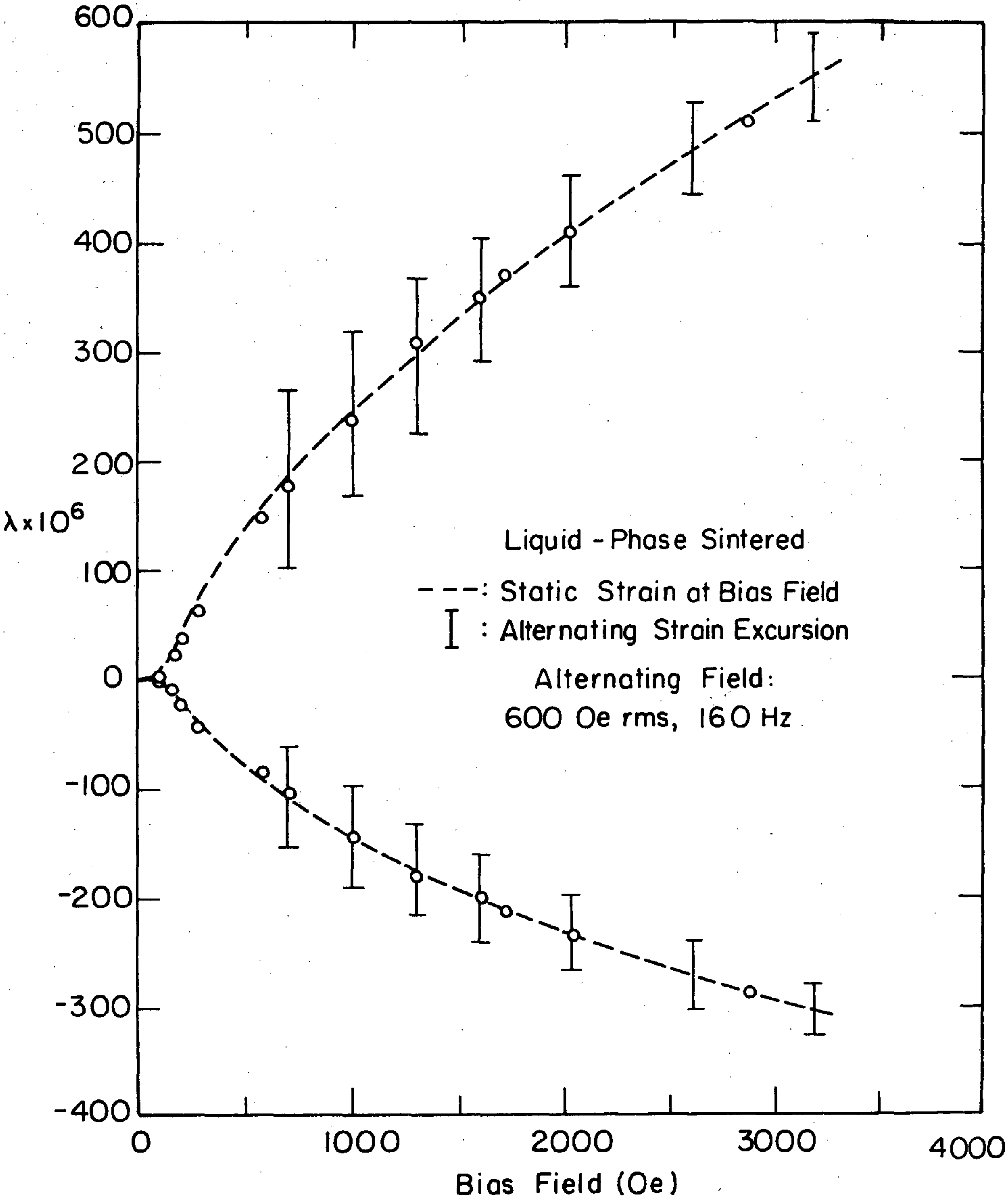

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Figure 6. Room temperature dynamic magnetostriction of liquid phase sintered $Tb_{0.3}Dy_{0.3}Fe_2$.

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FIGURE CAPTIONS

FIGURE 1. Experimental set-up for dynamic magnetostriction measurements.

FIGURE 2. Isothermal time dependence of shrinkage behavior of a $Tb_{0.3}Dy_{0.7}Fe_2$ sample.

FIGURE 3. Time and length corrected isotherms of liquid phase sintered $Tb_{0.3}Dy_{0.7}Fe_2$.

FIGURE 4. Effects of alignment and density on the relative permeability of $Tb_{0.3}Dy_{0.7}Fe_2$.

FIGURE 5. Dynamic magnetostrictions observed in a magnetically aligned sample of $Tb_{0.3}Dy_{0.7}Fe_2$.

FIGURE 6. Room temperature dynamic magnetostriction of liquid phase sintered $Tb_{0.3}Dy_{0.7}Fe_2$.

Manoochehr Malekzadeh was born in Kerman, Iran, on May 26, 1949. He received his B.S. degree from Tehran Polytechnic in 1970, and his Ph.D. degree from the University of California at Berkeley in 1978. In 1974 he joined the Materials and Molecular Research Division of the Lawrence Berkeley Laboratory to conduct research on powder metallurgical processing of magnetostrictive materials. Presently he is a postdoctoral research engineer at the same Laboratory doing research on development of multifilamentary superconductors based on A15 compounds. He is a member of ASME and the Metallurgical Society of AIME.

Milton R. Pickus received his B.S. and Ph.D. degrees from Yale University in 1934 and 1938 respectively. After serving as chief metallurgist for the Parker Pen Co. for 10 years, he came to the University of California at Berkeley where he is now Professor of Mechanical Engineering and a Principal Investigator at the Materials and Molecular Research Division of the Lawrence Berkeley Laboratory. He is a member of the American Society for Metals and of the American Powder Metallurgy Institute.

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