THE EFFECT OF MAGNETICALLY ALIGNED
POWDER ON THE MAGNETOSTRICTION OF
SINTERED RARE EARTH-IRON LAVES PHASE COMPOUNDS

M. Malekzadeh and M. R. Pickus

October 1977

Prepared for the U. S. Department of Energy
under Contract W-7405-ENG-48

TWO-WEEK LOAN COPY

This is a Library Circulating Copy
which may be borrowed for two weeks.
For a personal retention copy, call
Tech. Info. Division, Ext. 5782
LEGAL NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.
THE EFFECT OF MAGNETICALLY ALIGNED POWDER
ON THE MAGNETOSTRICTION
OF SINTERED RARE EARTH – IRON LAVES PHASE COMPOUNDS

M. Malekzadeh and M. R. Pickus

Materials and Molecular Research Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

ABSTRACT

A powder metallurgical approach is utilized to prepare
grain oriented Laves phase compounds of Tb$_x$Dy$_{1-x}$Fe$_2$. The
magnetostrains observed in the oriented compounds, though con-
taining ~20% porosity as presently prepared, are far superior
to those of arc cast and highly dense liquid phase sintered
materials. Also, it is shown that the alignment achieved is
strongly dependent on the Tb/Dy ratio.
Recently, a powder metallurgical approach\textsuperscript{1,2,3} has been used successfully in this laboratory to obtain the brittle rare earth-iron Laves compounds in suitable sizes and shapes. Their huge magnetostrictions have been reported elsewhere\textsuperscript{1-4}.

In this study, it is shown that the magnetostrictive properties can be greatly enhanced by magnetically aligning the powder particles prior to solid-state sintering. For this study, the pseudobinary system (Tb\textsubscript{x}Dy\textsubscript{1-x}Fe\textsubscript{2}) with x close to \(x = 0.3\) was selected because of this composition being particularly well suited for a wide range of device applications. Spin orientation diagrams show that this compound, at room temperature, has its axis of easy magnetization along the major cubic symmetry [111] direction.\textsuperscript{5} A powder metallurgical approach can utilize the direction of easy magnetization by alignment of the powder compound in a magnetic field.

The compound preparation consisted of arc melting elemental rare earth metals and iron, all of 99.9\% purity, on a water cooled copper hearth under a Zr gettered argon atmosphere. After a homogenizing anneal at 1000°C in evacuated quartz capsules, the buttons were crushed and pulverized by ball milling under toluene in a steel planetary mill for 20 minutes. The resulting powder (in 35-15 \(\mu\)m range) was rinsed with acetone and vacuum dried. Rubber tubing, 0.6 cm I.D. and 3.8 cm long, was manually filled with powder. Retention of magnetic alignment of the powder particles was the critical processing step.
The objective during the compacting stage was to maintain a good degree of alignment in the compound by mechanically interlocking the powder particles. For this purpose, after alignment of the powder in DC fields up to 20 kOe, the volume was reduced by evacuating the rubber tube to lock the particles in position. The tube was subsequently hydrostatically compressed at 70 Kg/mm$^2$. Although in the first set of experiments X-ray examination showed only a small degree of alignment, nevertheless, a definite improvement in magnetostriction was observed. A substantially higher degree of alignment was achieved when an alternating field of approximately 1000 Oe peak-to-peak at frequencies up to 500 Hz was superimposed on the DC field. A field of this type produced sufficient particle vibration to facilitate orientation of the loose powder. While in the magnetic field, the powder was compacted by hand applied end compression with a plunger before it was isostatically compressed. The cold pressed samples were subsequently wrapped in Ta foils and solid state sintered in a dynamic vacuum of $3 \times 10^{-6}$ mm Hg in the 950-1050°C temperature range for durations up to 12 h. The samples were evaluated by optical microscopy, X-ray diffraction patterns, X-ray pole figures and magnetostriction measurements. Samples measuring approximately 2 cm$^2$ by 0.3 cm thick were polished through a 1 μm diamond wheel and, after etching, were used to obtain X-ray diffraction patterns and pole figures. Plane orientations were determined by the
Schulz method⁶ using a Picker diffractometer with an X-ray monochromator. Intensities of (440) reflections were recorded as ω (the angle of reflecting plane with sample surface), and φ the azimuth angle from the longitudinal directions of the sample) were changed. A temperature-compensated circuitry with commercially available strain gauges from Micro-Measurements was used to determine the magnetostrains.

Figure 1 shows the X-ray diffraction patterns of random and magnetically oriented Tb₃Dy₇Fe₂ compound. In the surface perpendicular to the magnetic field, the diffraction intensity from (220) planes has decreased while the intensity of (222) reflection has increased, to a substantial degree, with respect to the intensities of reflections from these planes in an almost randomly oriented powdered compound. Magnetostriction measurements at room temperature show that with regard to both magnetostrains and rate of approach to saturation, the aligned material, though containing considerable porosity, is much superior to a highly dense (95% theoretical density) liquid phase sintered compound which is not aligned (Fig. 2). This superiority is possibly due to the effect of the preferred grain orientation on reducing the large internal strains at grain boundaries associated with highly magnetostrictive materials. Also, it is noteworthy that in rare earth-iron Laves phase compounds λ₁₁₁ ≫ λ₁₀₀ due to a structural distortion associated with the [111] easy direction of magnetization.⁷
(\(\lambda_{100}\) and \(\lambda_{111}\) are single crystal magnetostriction constants of cubic crystals in direction \(\langle 100 \rangle\) and \(\langle 111 \rangle\) respectively.)

Since for a polycrystalline material with random grain orientation, the saturation magnetostriction can be expressed as

\[
\lambda_s = \frac{2\lambda_{100} + 3\lambda_{111}}{5},
\]

magnetic alignment of these compounds orienting the crystallites along the \([111]\) easy axis, will enhance their saturation magnetostriction drastically. This is also illustrated in Fig. 2, where the saturation magnetostriction for the aligned \(Tb_{0.3}Dy_{0.7}Fe_2\) shows a 20% increase over the highly dense liquid phase sintered material.

It was also found that the alignment improves as the value of \(x\) in \(Tb_xDy_{1-x}Fe_2\) is increased, as is shown in Fig. 3, where the relative intensity of reflections from \((222)\) and \((220)\) planes are plotted against values of \((x)\). This is attributed to the fact that \(Tb_xDy_{1-x}Fe_2\) has a minimal anisotropy near \(x = 0.3\). An increase in \((x)\) will increase the anisotropy making an improvement in the alignment degree possible, which is evident in X-ray diffraction patterns taken from aligned sintered \(TbFe_2\) materials (Fig. 4a). Figure 4b shows a \((440)\) pole figure for an aligned \(TbFe_2\) sintered rod with contours of constant intensity around the \((440)\) pole, at the center. Although a decrease in \((x)\) toward a Dy richer compound also will increase the magnetic anisotropy, it was not considered as beneficial, since \(DyFe_2\) has its easy direction of magnetization along the \([100]\) instead of along the \([111]\) direction, thus not contributing much to the magnetostriction.
This work was supported by the Division of Materials Sciences, Office of Basic Energy Science, U. S. Department of Energy.

The authors wish to express their gratitude to Messrs. D. H. Nelson and W. E. Canady for their assistance in setting up the experimental equipment.
REFERENCES

FIGURE CAPTIONS

Fig. 1: X-ray diffraction patterns for an almost randomly oriented powder and a magnetically aligned specimen of Tb$_{3}$Dy$_{7}$Fe$_2$ compound.

Fig. 2: Room temperature magnetostriction of an aligned and a liquid phase sintered specimen of Tb$_{3}$Dy$_{7}$Fe$_2$ indicating a saturation magnetostriction for the aligned sample approximately 20% greater than that of the highly dense liquid phase sintered specimen.

Fig. 3: Change of X-ray reflection intensity ratios, (220)/(311) and (222)/(311), vs. the concentrations of Tb and Dy in magnetically aligned specimens.

Fig. 4: (a) X-ray diffraction patterns for a magnetically oriented TbFe$_2$ specimen.
(b) The (440) pole figure for the specimen.
Fig. 1
Figure 2

Aligned in magnetic field

Liquid phase sintered

\[ |\lambda_{ii} - \lambda_{zz}| \times 10^6 \]

vs.

H (kOe)
Cu Radiation

Diffraction Angle 2θ

Intensity

Field Direction

a

b

XBL 782-7284

Fig.
This report was done with support from the United States Energy Research and Development Administration. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the United States Energy Research and Development Administration.