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NOV 80 H T SAVAGE, R J ABBUNDI, A E CLARK
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MAGNETOSTRICTION AND CHARACTERIZATION OF BRIDGMAN GROWN RARE-EARTH IRON ALLOYS

BY H. T. SAVAGE, R. J. ABBUNDI,
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RESEARCH AND TECHNOLOGY DEPARTMENT

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We have prepared large samples of $Tb_{.27}Dy_{.73}Fe_2$ and $Tb_{.2}Ho_{.58}Dy_{.22}Fe_2$ by a Bridgman-like technique developed at United Technology. The samples have much larger values of saturation magnetostriction than vertically zoned materials. The materials have been characterized by metallographic x-ray and TEM studies.

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FOREWORD

The magnetic and magnetomechanical properties reported here are developments of a program to develop magnetostrictive materials for high power sonar. The materials are an alloy of terbium-iron and dysprosium-iron Laves phase compounds. The magnetoelastic strains are very large and very anisotropic.

Results of the first attempt to prepare large sample crystals of the highly magnetostrictive Tb compounds utilizing high temperature-gradient furnaces are reported here. This method has been identified as most promising to yield large single crystals (3" x ½" dia.) for sonar transducer elements. Nine crystals were prepared. Plane front conditions were identified. Part of these results will be reported on at the Fifteenth Rare Earth Research Conference (Rolla, Missouri).

This study was carried out in the Solid State Branch of the Radiation Division. The materials development was sponsored by the NRL Material Program under the direction of Howard Lessoff. Magnetic measurements and fabrication of prototype transducer components utilizing these materials is being carried out under the sponsorship of the NOSC Sonar Transducer Sciences Program. Research on the magnetoelastic properties of highly magnetostrictive rare earths is sponsored by the Office of Naval Research and the NSWC Independent Research Fund.

H. R. RIEDL
By direction

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CONTENTS

INTRODUCTION

The cubic Laves-phase rare-earth Fe_2 compounds have huge values of magnetostriction constants (λ) and anisotropy energy (E_k). However, with an appropriate alloying of binary compounds, ternaries of the form $R_xR_{1-x}Fe_2$ may be obtained with large values of λ but with E_k reduced by two orders of magnitude.[1] Large values of the magnetomechanical coupling factor, k_{33}, have been observed in $Tb_{.27}Dy_{.73}Fe_2$ (Terfenol). The magnetostriction is quite anisotropic with $\lambda_{100} << \lambda_{111} \approx 1.6x10^{-3}$. Because the large, anisotropic magnetostriction introduces large inhomogeneous strains in random polycrystals (RPC), the permeability is quite low.[2] This restricts magnetomechanical activity.

We previously reported efforts in grain orienting to enhance magnetomechanical activity.[3] Bridgman growth and horizontal zoning methods were examined during FY78. During FY79, methods were developed to prepare high coupling oriented samples by a vertical zoning technique.[4] This material is prepared by passing a vertical molten zone (VMZ) along a vertical rod. We achieved considerable improvement with this over polycrystalline material. Coupling factors larger than .7 were achieved in long rods. Random polycrystalline materials have values of .55. Raytheon Corporation is now building a transducer using long rods of vertically zoned material.

This report covers our first extensive attempt to obtain single crystal material in a manner suitable for large scale production of transducer rods. The method has been developed by United Technology for single crystal turbine blades. We wish to adapt it to the production of single crystal $Tb_{.27}Dy_{.73}Fe_2$. The method is basically a Bridgman growth technique. A boron nitride crucible is lowered through a temperature gradient. Since a large temperature gradient is necessary the upper part of the crucible is held well above the melting point (called "superheat"). The ampule can be lowered into liquid to enhance the temperature gradient. In this report we compare the VMZ material to material prepared by the United Technology (UT) method.

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[1] A. E. Clark, AIP Conf. Proc. 18, 1015 (1974).

[2] H. T. Savage, A. E. Clark, and J. M. Powers, IEEE Trans. on Magnetics, MAG-11, 1355 (1975).

[3] H. T. Savage, R. Abbundi, A. E. Clark, "Permeability, Magnetomechanical Coupling and Magnetostriction in Grain-Oriented Rare Earth-Iron Alloys," NSWC/WOL TR 78-197.

[4] H. T. Savage, R. Abbundi, A. E. Clark, "Magnetomechanical Coupling and Magnetostriction in Vertically Zoned $Tb_{.27}Dy_{.73}Fe_2$," NSWC TR 79-463.

PREPARATION AND SAMPLE CHARACTERIZATION OF UNITED TECHNOLOGY SAMPLES

We have grown nine samples. They are listed in Table I. The super heat required for a high temperature gradient causes rare-earth loss and interaction with the boron nitride crucible. Without a high temperature gradient, a large amount of Widmanstatten precipitate (WSP) is obtained. Fortunately our previous work has shown how to eliminate WSP. Material from Sample 2 was used to determine adequate heat treatment. Most WSP was removed by holding at 1000 C for 15 minutes and then air or water quenching. The samples showed no damage due to quenching. Unless otherwise noted in Table I, we followed a procedure of starting the growth at 1 cm/hour for about 2 cm of growth and then accelerating the rate linearly to 10 cm/hour. We wished to establish how fast we could grow and still obtain plane front growth. Our work shows that plane front growth can be achieved up to 3 cm/hour growth rate. Samples 9 and 10 were grown to see if a fast growth rate or high Dy content ($DyFe_2$ is much less peritectic than $TbFe_2$) would suppress WSP. They did not.

Sample No. 5 was our first truly successful effort. It will be discussed in detail. As mentioned above, plane front growth was achieved at a growth rate of 3 cm/hour. A 2 cm piece of the tip end of the boule was single by ordinary x-ray analysis. The boule is shown in Fig. 1. The arrow marks the transition to dendritic growth. The plane front end had an orientation of about 9 deg from <111>. Since <111> is the best possible direction we consider this a very good growth direction when compared with the vertically zoned material. Sample 5 was split in half. One-half was subsequently annealed at 650 C for 3 days. The unannealed half has maintained its physical integrity. The annealed half flaked gradually away. The 650 C anneal has apparently consolidated defects in regular pervasive ways. There may be an undetermined defect structure in the United Technology material that is not present in the vertically zoned material. The difference in growth kinetics has influenced the defect structure. The magnetostriction in this sample will be discussed in the next section.

We initiated a scanning transmission electron microscope (STEM) study to identify the defect structure (and look at domain walls). Sample preparation is difficult. By private communication with others who have tried, we think we are the first to successfully prepare a sample. A tiny hole must be in the sample. At the hole edge, stresses cause cleavage. The thin edge will, in general, oxidize away if exposed to air. We succeeded in preparing a sample by chemical attack. The sample is kept under alcohol and then the alcohol is pumped off in-situ. Although personnel problems at United Technology hindered the

TABLE 1

Sample No.	Composition*	Comments
1	$Tb_{.27}Dy_{.73}Fe_{1.95}$	Sample used to try out equipment. Result showed considerable Widmanstatten precipitate (WSP). Probably some rare-earth loss due to superheat. Interaction with crucible.
2	$Tb_{.27}Dy_{.73}Fe_{1.95}$	Superheat reduced 100 F, sample used to determine heat treatment for WSP removal, large amounts of WSP
3	$Tb_{.27}Dy_{.73}Fe_{1.90}$	Rare-earth content increased. Evidence of plane front growth near tip end where growth rate was slow.
4	$Tb_{.20}Dy_{.22}Ho_{.58}Fe_{1.95}$	Considerable interaction with crucible. Auger analysis showed migration of Ho to surface.
5	$Tb_{.27}Dy_{.73}Fe_{1.95}$	First relatively successful effort - discussed in text.
6	$Tb_{.27}Dy_{.73}Fe_{1.90}$	Sample showed dendritic structure throughout although pulled at constant rate of 1.5 cm/hour. Considerable reaction with crucible.
7	$Tb_{.20}Dy_{.22}Ho_{.58}Fe_{1.95}$	Did not react with crucible as much as previous quatenary, but extremely fragile.
8		No data
9	$Tb_{.15}Dy_{.85}Fe_{1.95}$	Solidified at 15 cm/hour, considerable WSP throughout
10	$DyFe_{1.95}$	Solidified at 15 cm/hour, considerable WSP throughout

*Composition of starting charge. Prepared at Iowa State University.

FIGURE 1 SAMPLE NO. 5. THE ARROW POINTS TO THE TRANSITION FROM PLANE FRONT TO DENDRITIC GROWTH. THE TIP END IS THE PLANE FRONT END.

STEM work, we do have a TEM picture of one sample (Fig. 2). The second phase has cracks which propagate into the parent phase. The sample is from our second run which had a lot of WSP present. The WSP is apparently quite deleterious.

MAGNETOSTRICTION

Magnetostriction measurements were made in Samples 2 and 5. Since the results in 5 are much better, we will discuss them. Figures 3 and 4 show the magnetostriction in the plane front and dendritic regions of sample 5. The large value of magnetostriction is in agreement with the x-ray data. The dendritic region does not show as high a magnetostriction thus showing that the preferred axis is "wandering." The slope of the perpendicular magnetostriction is limited due to demagnetizing effects. The slope of the parallel magnetostriction is also limited by demagnetizing, but in addition by defects.

After a 650 C anneal the magnetostriction at 400 Oe. was studied as a function of temperature, Fig. 5. The easy axis of magnetization is set by stoichiometry. The maximum magnetostriction occurs at minimum anisotropy. Minimum anisotropy occurs at the easy axis transition temperature. The maximum magnetostriction (400 Oe.) occurs at 40 C. This implies a Dy rich condition, the opposite of our expectations. The reasons for this are unknown. The value of the saturation magnetostriction is $\sim$1500 ppm as compared with $\sim$1000 ppm in the vertically zoned material. Since the figure of merit goes as the magnetostriction squared, this is a considerable improvement over the vertically zoned material.

CHARACTERIZATION AND COMPARISON OF THE VERTICALLY ZONED SAMPLES

We have studied the microstructure of the vertically zoned material (VZM) before and after annealing. A rod with a coupling factor of .6 before annealing was split down the middle. One half was then annealed. The microstructure that developed after annealing is shown in Fig. 6. A featureless region that looks like plane front growth is interspersed with a dendritic region that has long black "stringers" (Fig. 7). The "stringers" have been identified as rare earth oxide. A significant difference in permeability was found in different sections of the rod. Different admixtures of the regions could be responsible for the changes in permeability. This would result in a lower coupling factor (CF) than might otherwise be possible. The stringers could be caused by constitutional super cooling (CSC). CSC can be eliminated by a slower growth rate. X-ray studies show the growth axis considerably further from <111> than the United Technology samples. This result is also confirmed by our magnetostriction measurements. We finally note that no Widmanstatten is present before or after annealing, probably because of the high temperature gradient. We apparently need a higher temperature gradient in the United Technology method.

FIGURE 2 TRANSMISSION ELECTRON PICTURE OF WIDMANSTATTEN PRECIPITATE IN SAMPLE 2. THE PRECIPITATE HAS CRACKS WHICH EXTEND INTO THE PARENT PHASE.

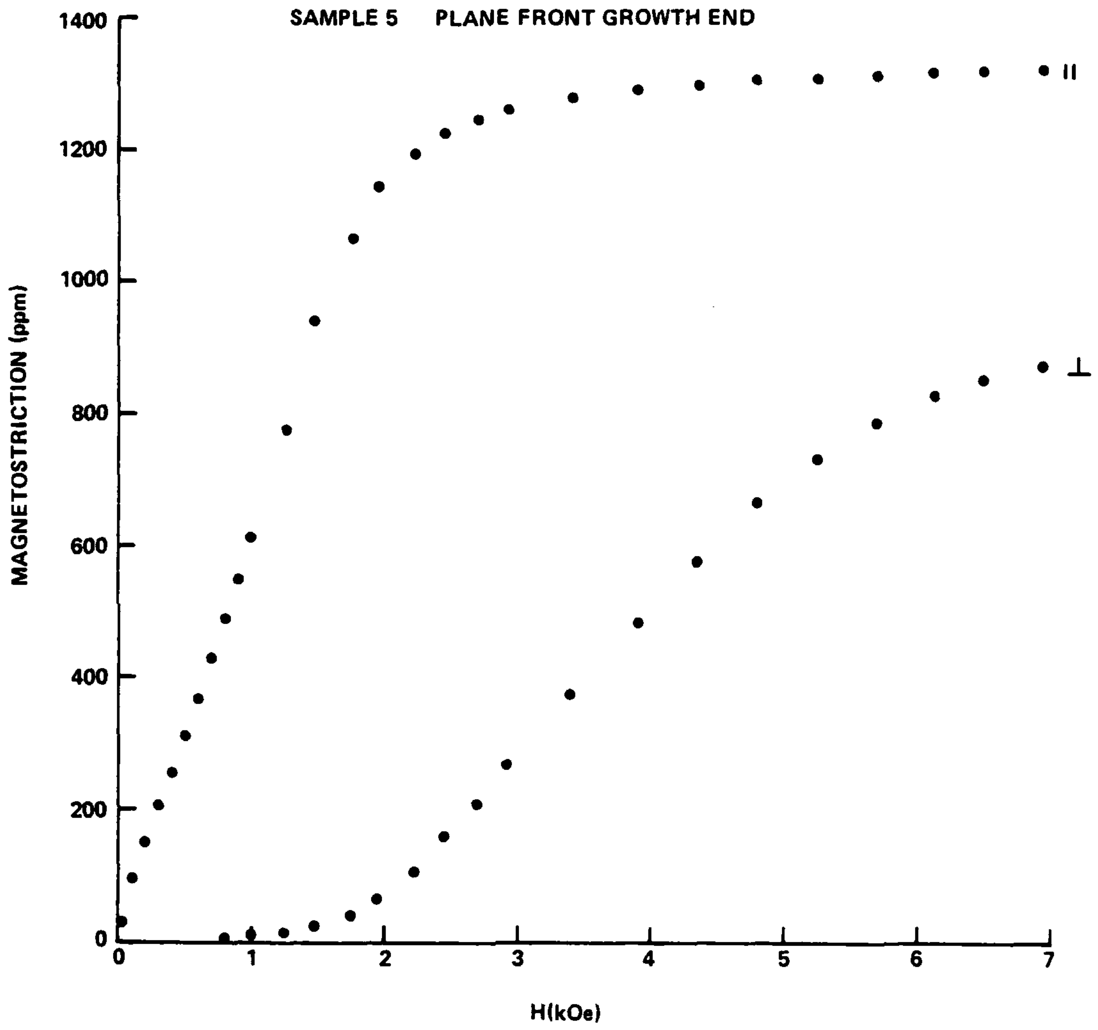

FIGURE 3 MAGNETOSTRICTION IN PLANE FRONT REGION OF SAMPLE 5. THE || MEASUREMENT IS WITH THE FIELD PARALLEL TO THE LONG SAMPLE AXIS. THE ⊥ MEASUREMENT IS WITH THE FIELD PERPENDICULAR TO THE SAMPLE AXIS.

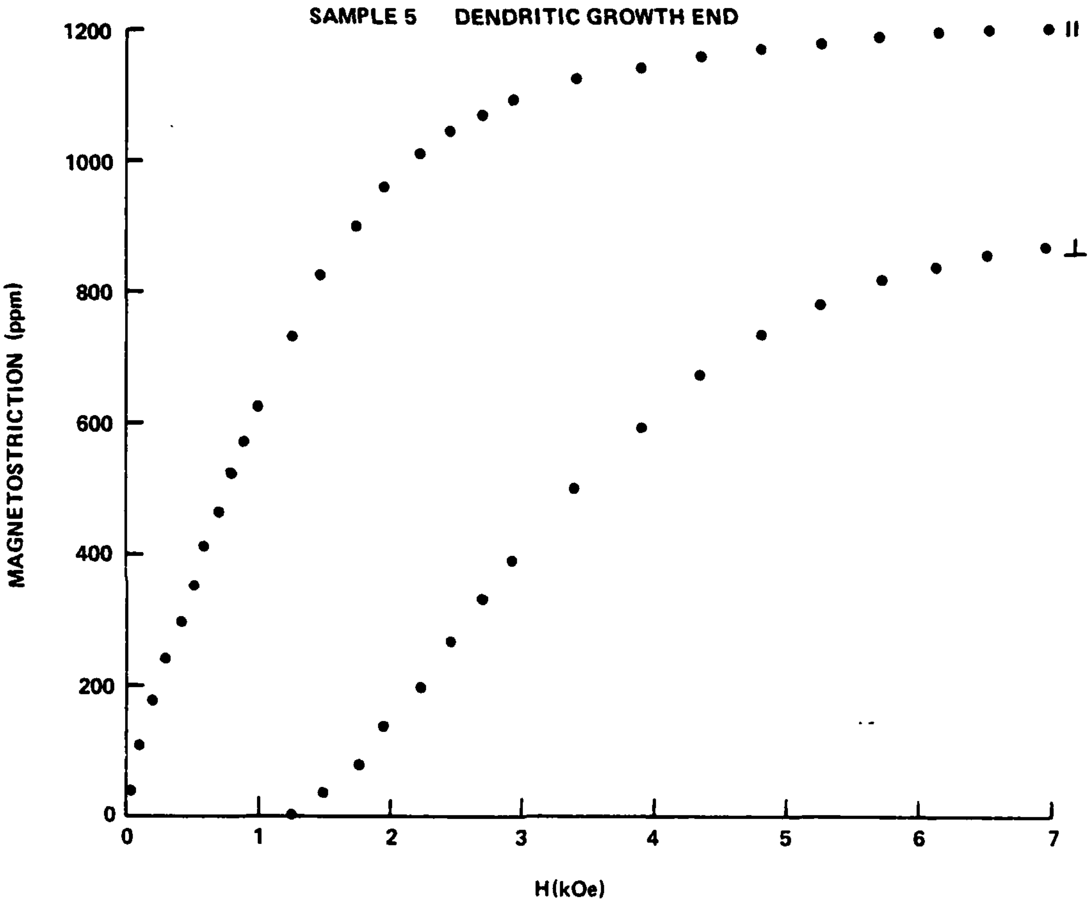

FIGURE 4 MAGNETOSTRICTION IN DENDRITIC REGION OF SAMPLE 5. THE || MEASUREMENT IS WITH THE FIELD PARALLEL TO THE LONG SAMPLE AXIS. THE ⊥ MEASUREMENT IS WITH THE FIELD PERPENDICULAR TO THE SAMPLE AXIS.

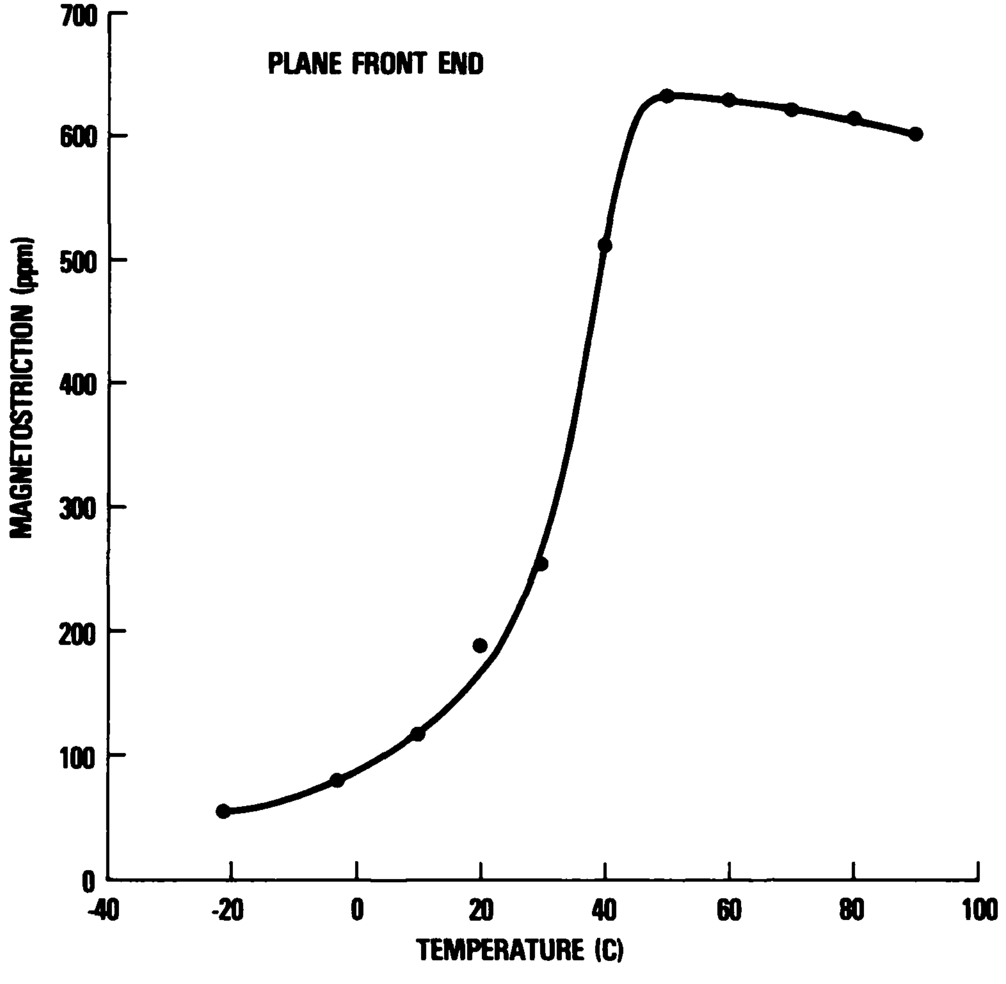

FIGURE 5 LONGITUDINAL MAGNETOSTRICTION AT 400 Oe AS A FUNCTION OF TEMPERATURE IN THE PLANE FRONT REGION OF SAMPLE 5.

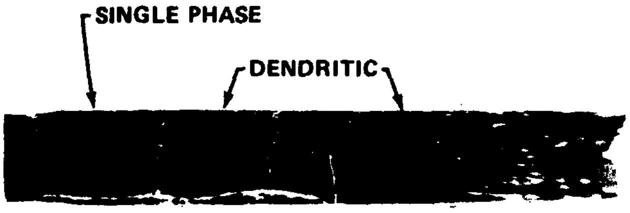

FIGURE 6 MICROGRAPH OF VERTICALLY ZONED, ANNEALED ROD, 2X MAGNIFICATION.

FIGURE 7 "STRINGERS" OF RARE-EARTH OXIDE IN VERTICALLY ZONED MATERIAL OF FIG. 6, 200X MAGNIFICATION.

RECOMMENDATIONS

The goal of this program is to develop mass production techniques to inexpensively produce large samples of highly magnetostrictive rare earth-Fe alloys. The alloys should possess magnetostrictions of greater than 1000 ppm at 500 Oe.

Widmanstatten-free Bridgman growth single crystals offer the highest promise. These samples will possess the highest possible saturation strain (2400 x 10^{-6}). Methods must be found to eliminate defects which limit the magnetostriction obtained at 500 Oe. Our recommendations are: (1) for more complete STEM work to identify defect structures; (2) determination of the phase diagram to eliminate defects by controlled growth kinetics, (3) crucible improvement. A most valuable study is the STEM because it will yield microscopic information on defects and structure of domain walls. X-ray topography will be done under a separate contract. The experiments will complement each other. STEM looks at microstructure, topography at more long range defects. The existence of a phase diagram will help in the choice of starting alloys and growth conditions. It may be possible to prevent the formation of a defect structure, rather than subsequently eliminating it by heat treatment. There is a serious problem with crucible interaction with the melt. The last several runs have been damaged by crucible breakdown. Alternate crucible materials calcia or calcia-zirconia should be investigated.

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