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SINTERING STUDIES OF RARE-EARTH IRON LAVES PHASE
MAGNETOSTRICTIVE COMPOUNDS

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ABSTRACT

Powder metallurgical techniques were used to prepare high density rare-earth iron Laves phase based magnetostrictive alloys. The rare-earth content of the alloys must be in excess of the stoichiometric composition in order to prevent the formation of oxygen stabilized intermetallic compounds. Solid state sintering leads to low density products. High density can be achieved by employing a liquid phase sintering approach. The magnetostrictive properties of the sintered products are comparable to those reported on cast samples.

The various magnetic properties of the rare-earth iron Laves phases have been subject to extensive studies in recent years. Of particular interest are the exceedingly high room temperature magnetostrictions which have been observed in several of these compounds (1-3). Recent investigations have shown that some of the rare-earth iron Laves phases show promise as compared with conventional energy conversion materials for underwater sound generation purposes (4,5). An inherent feature of these intermetallic compounds is their brittleness which precludes the use of common forming methods such as those employed for nickel based magnetostrictive alloys. The objective of the present study was to examine the possibility of employing powder metallurgical processing techniques in order to fabricate the Laves compounds in desirable shapes and sizes. Such methods are currently used in the fabrication of the structurally and compositionally related SmCo_5 -type permanent magnet materials.

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Throughout our study, we have used an alloy of approximate composition corresponding to the formula $(\text{Dy}_{0.7}\text{Tb}_{0.3})\text{Fe}_2$. This ternary Laves phase compound displays a relatively high magnetostriction associated with a minimal magnetic anisotropy. Various properties of this material, relevant for device application purposes, have been reported in the literature (6).

The initial procedure consisted of arc-melting 99.9% pure elemental rare-earth metals and iron under a zirconium gettered argon atmosphere. After a homogenization anneal at 1000°C for 48 h, the buttons were crushed and pulverized by ball milling under toluene in a steel planetary mill for approximately 30 minutes. The resulting 20-50 μm particle size powder was rinsed with acetone and vacuum dried. Rubber tubing, up to 1/4" diameter, was manually filled with the powder in room atmosphere and isostatically compressed at 75,000 psi. The cold pressed samples were wrapped in Ta foils and sintered in a dynamic vacuum furnace. In order to allow degassing of the samples, the temperature was increased gradually. Various sintering cycles in the 1050-1250 $^\circ\text{C}$ temperature range and for durations up to 48 h were tried. The sintered products were examined by optical and scanning electron microscopy.

FIG. 1

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Low density compacts prepared by solid state sintering. Black areas are mainly pores.

Powder diffraction patterns were taken using a Picker diffractometer in conjunction with an X-ray monochromator. A fluid displacement method was used for the density determination. Care was taken to prevent the penetration of the fluid into the pore network by coating the sample surface with a thin resin layer.

Samples prepared using the above described method exhibited very poor sinterability (Fig. 1). Changes in the various processing parameters such as milling time, compacting pressure, sintering temperature and duration did not lead to densities higher than 82%

of the theoretical density. Our findings confirmed the observation previously made in connection with the processing of SmCo_5 -type materials, that an initial rare-earth metal content in excess of the stoichiometric composition was necessary. In samples which had an initial rare-earth content less than 36 at.% - as compared to 33.3 at.% corresponding to the stoichiometric composition - the presence of an appreciable fraction of a foreign phase was detected. This phase was identified as an oxygen stabilized intermetallic compound of composition $\text{R}_{12}\text{Fe}_{32}\text{O}_x$ with R a rare-earth element and $x \leq 2$ (7,8). The relatively low oxygen content of this compound results in its large volume fraction. The absorbed surface layer on the milled powder particles provides the oxygen necessary for the formation of this intermetallic. Even by using purified argon in a glove box, it was not possible to suppress its formation. Only by adding 3-4 at.% excess rare-earth could its presence be eliminated. The excess

rare-earth reacted with the available oxygen to yield a dispersion of sesquioxides. The oxygen content of these oxides being high, their volume fraction was relatively low. An iron content in excess of the stoichiometric composition led to the precipitation of the RFe_3 phase, in samples which were slowly cooled from the sintering temperature. The appearance of these precipitates is attributed to a solubility range of the Laves phase, extending toward the iron rich composition at elevated temperatures. The presence of the oxygen stabilized compounds and of the RFe_3 precipitates is shown in Figure 2.

The low densities have an adverse effect on both the mechanical and the magnetic properties of the sintered compacts. The presence of pores and of foreign phases (the oxygen stabilized compounds are paramagnetic at room temperature) decreases the permeability of the sintered products by increasing the internal demagnetizing fields. The foreign phases (other than the sesquioxides) were eliminated by using an excess of rare-earth metals; the porosity was drastically reduced by employing a liquid phase sintering approach.

FIG. 2

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Oxygen stabilized intermetallics (rounded grains) in the Laves phase matrix. Plate like RFe_3 precipitates are aligned according to the orientation of the parent Laves phase grains.

The binary Laves compounds $TbFe_2$ and $DyFe_2$ are formed peritectically as a result of a reaction between iron rich phases and a rare-earth rich, low melting liquid (9,10). Even though the ternary Dy-Tb-Fe phase diagram is not available, it is reasonable to assume that the pseudobinary $(Dy_{0.7}Tb_{0.3})Fe_2$ compound is formed in a similar way. In order to provide a liquid component in the course of the sintering, the procedures described above were followed except that the starting material consisted of two different alloys containing 55 and 66.7 at.% Fe respectively. These initial compositions were chosen in such a manner that by taking 25% of the iron poor alloy, the final homogenized composition

of the product corresponded to 64 at.% Fe, necessary for the elimination of the oxygen stabilized phase. The iron deficient component of the mixture constituted the liquid phase at the sintering temperature. Higher than a 25% liquid fraction led to loss of shape of the compact during sintering. The composition of the iron poor phase, its relative fraction, the sintering temperature and the rate of heating are critical parameters of the process. The cold pressed mixture of the two powders was heated gradually to $800^\circ C$, and then, at the highest possible rate (50 deg/min) to the sintering temperature of $1130 \pm 5^\circ C$. At lower heating rates in this upper temperature range, the

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FIG. 3

Optical micrograph of $\text{Dy}_{0.7}\text{Tb}_{0.3}\text{Fe}_2$ sample. Liquid phase sintered 12 h at 1130°C ; 95% of theoretical density.

XBB 768-6887

FIG. 4

Scanning electron micrograph of liquid phase sintered sample, 97% theoretical density. Pores appear as black and oxides as white areas.

increased mobility of the atoms leads to rapid homogenization, thereby reducing the liquid phase fraction to values at which it is not effective.

Using the liquid phase sintering procedure, samples with 97% of the theoretical density were prepared. Beside the increased density, this method

XBL 768-7302

FIG. 5

Room temperature magnetostriction
of sintered $\text{Dy}_{0.7}\text{Tb}_{0.3}\text{Fe}_2$.

of preparation also resulted in a large grain size (Figs. 3 and 4). Since grain boundaries are effective barriers to domain wall motion, large grain size samples are preferred for their higher permeability.

The static magnetostrictive behavior of the sintered compacts was measured using a conventional strain gauge technique. The results obtained thus far (Fig. 5), indicate that both with regard to the magnetostrains attained, and the rate of approach to saturation, the sintered material is comparable, though slightly inferior to cast materials (6).

Further work and, in particular, a study of the dynamic magnetostrictive properties of the sintered products is in progress.

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