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THE MAGNETOSTRICTIVE PROPERTIES OF Dy-Fe-B ALLOYS WITH NANOCRYSTALLINE GRAIN STRUCTURE

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Abstract-The magnetostriction versus field (λ-H) curves for the melt-spun ribbons of $Dy_x(Fe_{1-y}B_y)_{1-x}$ (x=0.2, 0.25, 0.3; y=0, 0.05, 0.1, 0.15, 0.2) alloys are measured systematically at various wheel speeds ranging from 10 to 50 m/sec. The λ-H curves in most cases vary sensitively with the wheel speed and, in the wheel speed range where no amorphous phase is formed, the magnetic softness improves rather continuously with the wheel speed. This result is considered to be due to the reduced grain size with increasing wheel speed, which was confirmed by X-ray diffraction and transmission electron microscopy. In particular, homogeneous and ultrafine grains with size of about 10 nm are formed even in the as-spun state when the $Dy_{0.3}(Fe_{1-y}B_y)_{0.7}$ alloys are quenched at the wheel speed of 30 m/sec (for the alloy with y=0.2) or 40 m/sec (for the alloys with y≤0.15) and the ribbons having the nanocrystalline grain structure exhibit good magnetostrictive characteristics.

I. INTRODUCTION

The magnetostriction is a phenomenon related to a change in dimension when a substance is exposed to a magnetic field altering its state of magnetization. This phenomenon is observed in any types of magnetic materials but only ferri- and ferromagnetic materials exhibit large magnetostrains which are of practical importance. Among these materials, rare earth (R) - transition metal (TM) based alloys, which were developed more than two decades ago, are known to exhibit the largest magnetostrains [1-3]. The saturation magnetostriction, λ_s, of cubic Laves RFe_2 is as large as about 2000×10^{-6} (or 2000 ppm). This value is roughly 40 times larger than that of previously developed Ni-Fe, Ni-Co and Fe-Co alloys and also it is more than 10 times larger than that of piezoelectric materials [3]. However, since the magnetocrystalline anisotropy of R-TM alloys is very large, a large magnetic field is required to obtain a large magnetostrain, which is not desirable in the practical point of view. Much work has been done to achieve a large strain at a low field. An early main attempt is to develop alloys with low magnetocrystalline anisotropy. One notable example is the investigation of pseudobinary alloys such as $(R_1,R_2)Fe_2$ alloys [4] and $R(Fe,TM)_2$ alloys [5]. The main outcome of the investigation is the development of the well known $Tb_{0.3}Dy_{0.7}Fe_2$ alloy (Terfenol-D) [6].

Recently, active research is currently being conducted to reduce the magnetocrystalline anisotropy by controlling the microstructure, particularly, the grain size [7,8]. The theoretical background for this is that the apparent or effective magnetocrystalline anisotropy is reduced when the size of grains (D) is smaller than the ferromagnetic exchange length (L_{ex}) [9];

$$K_{eff} = K_1 / \sqrt{N} \quad \text{for } D < L_{ex} \qquad (1)$$

where K_1 is the (intrinsic) magnetocrystalline anisotropy and N is the number of grains within the ferromagnetic coupling region. The main routes to microstructural modification include; (1) the direct fabrication of microcrystalline structure by quenching the melt at a suitable cooling rate and (2) the production of an amorphous phase by rapid-quenching and a subsequent annealing of the amorphous phase to form microcrystalline structure. It has been reported by many workers that the magnetostrictive characteristics are improved by the microstructural modifications [7], although no systematic work has been done yet.

In this research scheme, effects of the microstructure of melt-spun R-Fe based alloys are investigated systematically on the magnetostrictive characteristics. The control of microstructure was done (1) by varying the cooling rate during melt-quenching and (2) by adding a small amount of metalloid to the alloys. The metalloid was added, since it affects the microstructure of melt-spun ribbons and reduces local magnetocrystalline anisotropy [10]. As a first attempt, the magnetostriction of melt-spun $Dy_x(Fe_{1-y}B_y)_{1-x}$ (x=0.2, 0.25 and 0.3; y=0, 0.05, 0.1, 0.15 and 0.2) was investigated. A preliminary result shows that melt-spun ribbons of the alloy with x=0.3 and y=0.2 exhibit good magnetostrictive characteristics when the ribbons are produced at suitable cooling rates so that grains of 10-20 nm are formed [11]. In this paper, more systematic and comprehensive results are presented on the magnetostrictive characteristics of $Dy_x(Fe_{1-y}B_y)_{1-x}$ (x=0.2, 0.25 and 0.3; y=0, 0.05, 0.1, 0.15 and 0.2) alloys which are fabricated at various cooling rates. Similar work on Tb-based alloys are currently under way and will be reported in the future.

II. EXPERIMENTAL

Dy-Fe-B alloys were arc-melted in an Ar atmosphere. In order to reduce the compositional segregation, the ingots were remelted several times. Subsequent melt-spinning was carried out also in an Ar atmosphere. In the present melt spinning experiments, all fabrication parameters are fixed except for the wheel speed which was varied widely from 10 to 50 m/sec. The melt temperature was maintained just above the respective alloys. The orifice diameter was about 0.5 mm and the chamber and ejecting pressures were 500 mmHg and 8.5×10^{-2} MPa, respectively. The ribbon width was 1-2 mm. The length varied widely depending on fabrication parameters and the alloy composition, particularly B content; the length in general increases with increasing B content and decreasing wheel speed. The magnetostriction was measured by a three terminal capacitance method at room temperature and at the magnetic field (H) up to 8 kOe. The microstructure was mainly examined by X-ray diffraction. The grain size was determined from full width at half maximum (FWHM) by using the Scherrer equation. Also, in some cases, the X-ray results were supplemented by transmission electron microscopy (TEM).

III. RESULTS AND DISCUSSION

$Dy_{0.2}(Fe_{1-y}B_y)_{0.8}$ alloy system

In Fig.1(a) are shown the λ- H results for the alloys y=0 and y=0.2 for the wheel speeds of 30 and 50 m/sec. It is seen from Fig.1(a) and the results for the allloys with the intermediate B contents, not shown here to avoid complexity, that

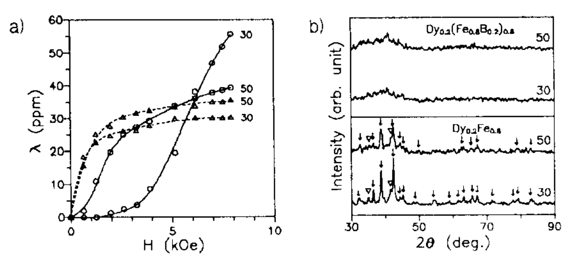

Fig.1 (a) The λ-H plots and (b) the X-ray diffraction patterns for the alloys $Dy_{0.2}Fe_{0.8}$ (circles) and $Dy_{0.2}(Fe_{0.8}B_{0.2})_{0.8}$ (inverse triangles). The numbers on the curves denote the wheel speed in m/sec and, the Dy_6Fe_{23} and $DyFe_3$ phases are indicated by the arrows and open triangles respectively.

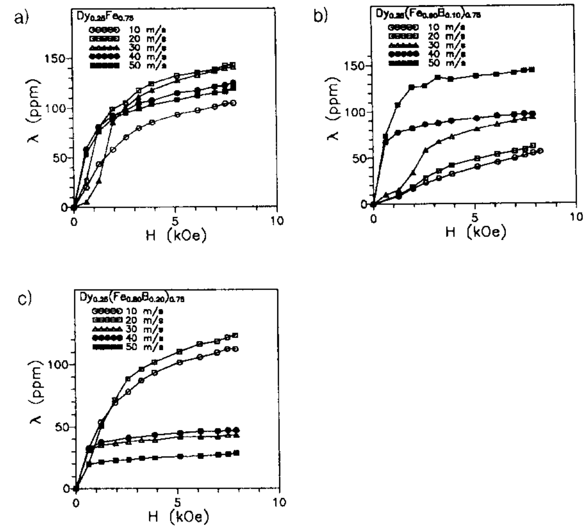

Fig.2 The λ-H plots for the alloy (a) $Dy_{0.25}Fe_{0.75}$, (b) $Dy_{0.25}(Fe_{0.90}B_{0.10})_{0.75}$ and (c) $Dy_{0.25}(Fe_{0.80}B_{0.20})_{0.75}$.

λ increases very slowly with H at low fields when ribbons of low B alloys are fabricated at low wheel speeds indicating wheel speed of 30 m/sec or less. However, λ increases substantially with H even at low magnetic fields when the ribbons of the same low B alloys are fabricated at wheel speed of 40 or 50 m/sec. This indicates that the magnetic softness improves with the wheel speed. The magnetic softness of the high B alloy is good at all wheel speeds investigated in this work and is better than that of the low B alloy. These λ-H results are well explained by the X-ray diffraction results shown in Fig.1(b). In the low B alloy, sharp diffraction peaks are seen at low wheel speeds indicating the existence of coarse grains, but the peaks are becoming broad at high wheel speeds indicative of grain refinement. In the high B alloy, amorphous-like broad pattern is observed even at that the glass forming ability is improv0ed by B addition. The value of λ is low in this alloy system; 30 to 60 ppm at 8 kOe. The crystalline phase formed is mainly Dy_6Fe_{23}. A small amount of $DyFe_3$ is also idenfied from the X-ray results.

$Dy_{0.25}(Fe_{1-y}B_y)_{0.75}$ alloy system

In Figs.2(a)-(c) are shown the λ-H results for the alloys y=0, 0.1 and 0.20, respectively. The λ-H curves of the alloys y=0.05 and 0.15, not shown here, are similar to those shown in Fig.2(b) for the y=0.1 alloy. The wheel speed was varied from 10 to 50 m/sec. It is seen from the figures that the magnetic softness improves continuously with the wheel speed, except for the alloys y=0 and 0.2.

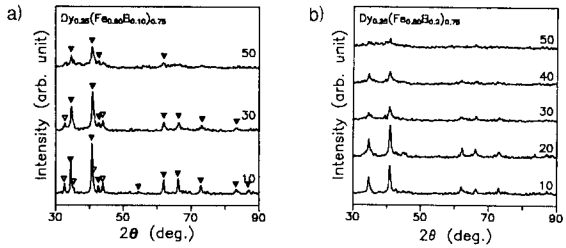

Fig.3 The X-ray diffraction patterns for the alloy (a) $Dy_{0.25}(Fe_{0.9}B_{0.1})_{0.75}$ and (b) $Dy_{0.25}(Fe_{0.8}B_{0.2})_{0.75}$. The $DyFe_2$ and $DyFe_3$ phases are indicated by the closed and open inverse triangles, respectively. The numbers on the curves denote the wheel speed in m/sec.

These λ-H results are well matched with the X-ray results shown in Fig.3(a) for the alloy y=0.1. The X-ray results of the other alloys except for the y=0.2 alloy, not shown due to space limitation, are similar to those shown in Fig.3(a). The X-xay results clearly show that the peak intensity decreases and the FWHM increases with the wheel speed, indicating that the grains are refined and hence the magnetic softness inproves as the wheel speed increases. The wheel speed dependence of λ -H behavior of the B free alloy is rather different from that of the B containing aloys; in the case of the B free alloy, the wheel speed dependence of λ -H behavior is small and, at low magnetic fields, the magnetic softness of the ribbons fabricated at the wheel speed of 30 m/sec is even poorer than that of the ribbons produced at the wheel speed of 10 or 20 m/sec. In the case of the y=0.2 alloy, the value of λ at low wheel speeds of 10 and 20 m/sec is higher than that at the higher wheel speeds. The reason for this was originally thought to be due to the fact that, even at the low wheel speeds of 10 and 20 m/sec, fine crystalline grains enough to result in good magnetic softness are formed at the high B content. The observed X-ray results shown in Fig.3(b), however, do not seem to clearly support the assumption, since the peak intensity and the degree of peak broadening for the high B alloy are similar to those for the low B alloys. More experiments are needed to clearly understand these results. The value of λ at 8 kOe is significantly reduced to 40 ppm when the ribbons are produced at the high wheel speeds of 30 m/sec and over. This is considered to be related to the formation of an amorphous phase, although this fact is not fully supported by the X-ray results. The crystalline phase formed changes with B content; mainly $DyFe_3$ and $DyFe_2$ phases are formed at the y=0 and y=0.2 alloys, respectively

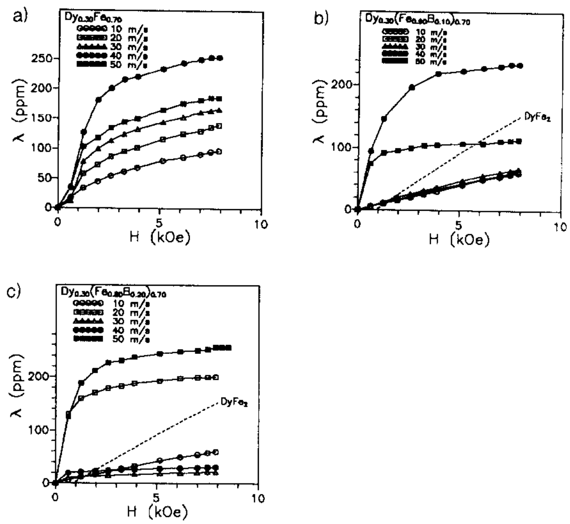

Fig.4 The λ-H plots for the alloy (a) $Dy_{0.30}Fe_{0.70}$, (b) $Dy_{0.30}(Fe_{0.90}B_{0.10})_{0.70}$, and (c) $Dy_{0.30}(Fe_{0.80}B_{0.20})_{0.70}$.

and, at the intermediate B contents, $DyFe_3$ and $DyFe_2$ phases coexist. The value of λ at 8 kOe is 70-160 ppm, which is 2-3 times higher than that for the $Dy_{0.2}(Fe_{1-y}B_y)_{0.8}$ alloy system.

$Dy_{0.3}(Fe_{1-y}B_y)_{0.7}$ alloy system

In Figs.4(a)-(c) are shown the λ-H results for the alloys y=0, 0.1 and 0.2, respectively. The λ-H results of the alloys y=0.05 and 0.15, omitted here, are similar to those of the y=0.1 alloy. The value of λ increases with the wheel speed, exhibits a maximum at the wheel speed of 40 m/sec (30 m/sec for the y=0.2 alloy) and then decreases with the wheel speed. Figs.5(a) and (b) show the X-ray results for the alloys y=0.1 and 0.2, respectively. The diffraction patterns of the other alloys are similar to those shown in Fig.5(a) for the y=0.1 alloy. The way in which the intensity and the width of the peaks vary with the wheel speed is

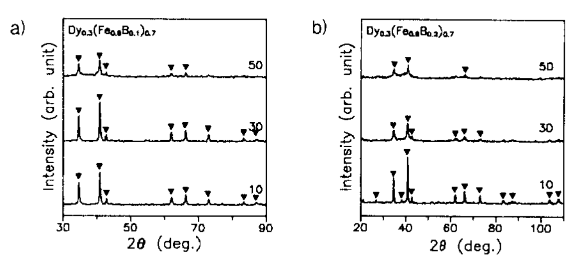

Fig.5 The diffraction patterns for the alloy (a) $Dy_{0.30}(Fe_{0.90}B_{0.10})_{0.70}$ and (b) $Dy_{0.30}(Fe_{0.80}B_{0.20})_{0.70}$. The $DyFe_2$ phase is indicated by the closed inverse triangles. The numbers on the curves denote the wheel speed in m/sec.

similar to that observed for the $Dy_{0.25}(Fe_{1-y}B_y)_{0.75}$ alloy system. The crystalline phase identified is mainly $DyFe_2$ and a small amount of $DyFe_3$ phase is also observed for the alloys y=0 and 0.05 from the X-ray diffraction patterns. Also shown in the figires are the results for a crystalline bulk $DyFe_2$ phase for comparison [4]. In the case of bulk $DyFe_2$, λ remains nearly zero until 1 kOe and then increases linearly with the magnetic field.

The decrease in λ at the high wheel speed of 40 or 50 m/sec may be related to the formation of an amorphous phase. The value of λ_s of amorphous $DyFe_2$ is very low (38 ppm [3]), which is much lower than the value of 433 ppm at 25 kOe for the crystalline counterpart as well as that of the present melt-spun ribbons fabricated at the optimum condition. It is therefore likely that λ is reduced by the formation of an amorphous phase. An amorphous phase can also be formed at the high wheel speed for the $Dy_{0.25}(Fe_{1-y}B_y)_{0.75}$ alloy system. However, since the main crystalline phase formed is $DyFe_3$ whose amorphous phase has rather high λ value (130 ppm [3]), the value of λ will not be affected by the presence of an amorphous phase. The dependence of λ-H behavior on the wheel speed for the alloy with the highest B content in the $Dy_{0.25}(Fe_{1-y}B_y)_{0.75}$ alloy system (very low λ_s at high wheel speeds) may also be explained in a similar to way to that observed for $Dy_{0.3}(Fe_{1-y}B_y)_{0.7}$ alloy system, since the main crystalline phase is $DyFe_2$. The explanation that the decrease in λ at the high wheel speeds is related to the formation of an amorphous phase is well supported by TEM experiments to be discussed later and also by the X-ray diffraction patterns where amorphous-like patterns appear to be seen at high wheel speeds.

In order to examine the microstructure more clearly, TEM experiments were carried out. The samples selected for TEM observations are; (1) the ribbons fabricated at the same wheel speed of 40 m/sec for all the alloys and (2) the ribbons of the alloy y=0.2 fabricated at the wheel speeds of 10, 20, 30, 40 and 50 m/sec. TEM results for the ribbons fabricated at the wheel speed of 40 m/sec shows that the nanocrystalline structure is formed in the case of low B alloys (y=0, 0.05 and 0.1), whilst an amorphous phase is formed for the high B alloys. In Fig.6 is shown the TEM microstructure for the ribbon fabricated at the same wheel speed of 40 m/sec for the y=0.05 alloy. It is seen that very fine grains with the size of about 10 nm are observed. In Fig.7 are shown the TEM results for the y=0.2 alloy ribbons fabricated at the wheel speeds of 10,

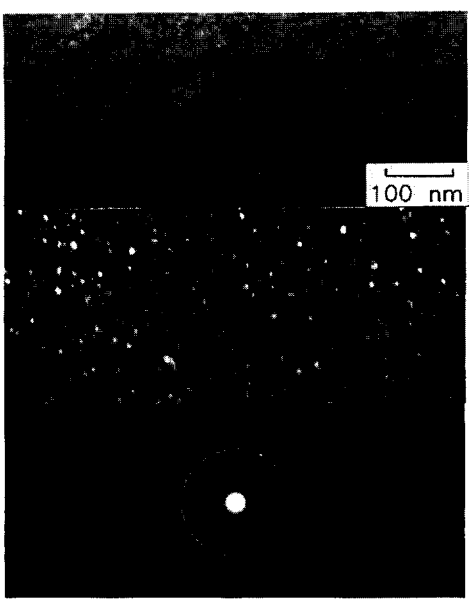

Fig.6 The transmission electron micrographs(the bright and dark field images) and selected area diffraction pattern for $Dy_{0.30}(Fe_{0.95}B_{0.05})_{0.70}$ alloy ribbon fabricated at the wheel speed of 40 m/sec.

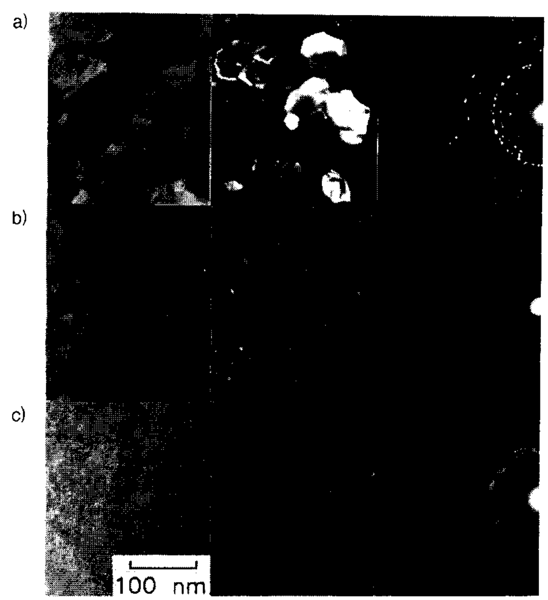

Fig.7 The transmission electron micrographs(the bright and dark field images) and selected area diffraction patterns for $Dy_{0.3}(Fe_{0.8}B_{0.2})_{0.7}$ alloy ribbons fabricated at the wheel speeds of (a)10 m/sec, (b)30 m/sec and (c) 50 m/sec.

Table 1. The grain size (in nm) of $Dy_{0.3}(Fe_{1-y}B_y)_{0.7}$ alloys determined by using the Scherrer formula

y \ Wheel speed (m/sec)	10	20	30	40
0				11.3
0.05				18.9
0.10				18.9
0.15				—
0.20	29.3	22.9	11.6	—

30 and 50 m/sec. Very coarse grains are observed at the wheel speed of 10 m/sec but, at the wheel speed of 30 m/sec, very fine grains of 10 nm are formed. An amorphous phase is seen at the wheel speed of 50 m/sec, as is well expected.

The present λ-H and microstructural results indicate that good magnetostrictive characteristics are obtained when very fine and homogeneous grains of about 10 nm are formed. When grains are coarse, the magnetic softness is poor, whilst the value of λ is very low when an amorphous phase is formed. The observation of very fine and homogeneous grains in the as-spun state is of interest and, to our knowledge, is first, although similar ultrafine grains were previously observed for the Tb-Fe-B alloy system by suitably annealing precusor amorphous alloy ribbons [7].

In order to compare the grain size observed by TEM with that by X-ray diffraction, the grain size obtained by using the Scherror equation is given in Table 1. The values of 11.3 and 11.6 nm obtained from X-ray results for the alloy y=0 and the wheel speed of 40 m/sec and for the alloy y=0.2 and the wheel speed of 30 m/sec, respectively, are well in agreement with those observed by TEM. The grain size of 18.9 nm for the alloy y=0.1 and the wheel speed of 40 m/sec obtained from X-ray diffraction patterns also agrees well with that observed by TEM. The value of 18.9 nm for the alloy y=0.05 and the wheel speed of 40 m/sec obtained from X-ray diffraction patterns is rather larger than that observed by TEM. In the case of the y=0.2 alloy, the grain size determined from the X-ray diffraction patterns decreases with the wheel speed, which agrees well with the TEM observation, but the value of grain size itself at the wheel speeds of 10 and 20 m/sec is substantially small compared to that observed by TEM.

IV. CONCLUSION

The magnetostrictive characteristics of melt-spun $Dy_x(Fe_{1-y}B_y)_{1-x}$ (x=0.2, 0.25 and 0.3; y=0, 0.05, 0.1, 0.15 and 0.2) alloys are systematically investigated as a function cooling rate during melt spinning and the results are summarized as follows;

(1) Homogeneous and ultrafine grains with the size of 10 nm are formed even in the as-spun state when the $Dy_{0.3}(Fe_{1-y}B_y)_{0.7}$ alloys are melt-spinned at a suitable cooling rate which depends on the B content. The ribbons having the nanocrystalline structure exhibit excellent magnetostrictive characteristics.

(2) It is seen from the results for the wheel speed dependence of λ-H curves that, in most cases, the magnetic softness improves with the wheel speed unless an amorphous phase is formed. This result is explained by the grain refinement with the increase of wheel speed and was supported by X-ray diffraction and transmission electron microscopy.

(3) The magnitude of λ is reduced significantly when main crystalline phase $DyFe_2$ is transformed into an amorphous phase. The wheel speed at which an amorphous phase is formed decreases with the B content.

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