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Discovery of Giant Magnetostriction in Amorphous RFe_2B (R = Sm, Tb) Alloys

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Compared with the conventional magnetostriction in Ni alloys which are in the order of several tens ppm (Parts Per Million $=10^{-6}$), RFe_2 (R = rare earth element) Laves Phase intermetallic compounds show large saturation magnetostriction in the range of a few thousands ppm. However, the large external magnetic field necessary to obtain saturation magnetostriction has due to large magnetocrystalline anisotropy energy restrained the application of magnetostriction materials in RFe_2 intermetallic compounds. As a result of its solution, the largest published value of effective giant magnetostriction in a low external magnetic field (less than a few hundred Oe) is reported in this paper by means of amorphisation of RFe_2 intermetallic compounds with the addition of boron, as a half metal. For the amorphous $(SmFe_2)_{0.97}B_{0.03}$ alloys, the effective magnetostriction of -545 and -610×10^{-6} is obtained at 400 and 1,000 Oe, respectively. Moreover, the effective magnetostriction of 590 and 630×10^{-6} in the amorphous $(TbFe_2)_{0.98}B_{0.02}$ alloys is also found at 400 and 1,000 Oe, respectively. This result will provide a clue to understanding the effect of half metal on anomalous increase of the effective giant magnetostriction and attract the great attention for magnetostriction applications.

1. Introduction

It is well known that RFe_2 Laves phase intermetallic compounds exhibit very large magnetostriction in the order of a few thousands ppm at room temperature, referred to as giant magnetostriction(1)(2). These intermetallic compounds are one of the promising materials for magnetostrictive applications, as pointed by Clark *et al*(2). However, because RFe_2 intermetallic compounds have large magnetostriction constants as well as large crystalline anisotropy constants, a large external magnetic field is necessary to reach saturation magnetostriction (λ_S)(3)(4). This has hindered the application of such components. Hence main researches on the giant magnetostriction have been concentrated on producing materials exhibiting high magnetostriction in a low magnetic field(5). The conventional research trend for obtaining a large effective magnetostriction in a low magnetic field has been to substitute rare earth elements for others in the intermetallic compounds(6) and consequently reducing the magneto crystalline anisotropy energy in the crystalline state. Terfenol-D, $Tb_{0.27}Dy_{0.73}Fe_2$ intermetallic compound, is one of the results obtained from such research. Also, there has been the research on amorphous RFe_2 alloy which can theoretically eliminate the magnetocrystalline anisotropy energy(7)(8). Although both of the research trends have yielded good results, large effective magnetostriction has not been successfully achieved under the magnetic field of conventional Ni alloy which is less than a few hundreds Oe.

In the present work, to induce the large effective giant magnetostriction at less than a few hundreds Oe, addition of half metal into the RFe_2 intermetallic compounds and amorphisation of them are carried out. The reason for selecting $SmFe_2$ and $TbFe_2$, and boron as matrix and additive, respectively in this work are,

(1) among RFe_2 intermetallic compounds, $SmFe_2$ and $TbFe_2$ show the largest negative and positive saturation magnetostriction, respectively. Moreover, it is possible to prepare the amorphous $SmFe_2$ and $TbFe_2$ alloys, which significantly reduce the long range order of magnetocrystalline anisotropy energy.

(2) Addition of boron into amorphous $SmFe_2$ and $TbFe_2$ alloys was aimed for the purpose of increasing the saturation magnetization(9) and reducing the localized magnetic anisotropy energy(10).

2. Experimental Procedure

Targets of $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}B_x$ intermetallic compounds were prepared by arc melting in an Ar atmosphere. The prepared targets were placed in a high rate DC-triode sputtering apparatus(11) to produce amorphous bulk specimens of $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}B_x$ alloys in the form of a disk, 15 mm diameter and 0.3 mm thickness, on Cu substrates. To eleminate the effect of Cu on measurement, the substrates were then dissolved in a mixture of chromium trioxide and sulphuric acid solution at 350 K for 24 hours.

The substrate-free specimens were examined by X-ray diffraction (XRD) measurements using Fe-Kα radiation at room temperature to confirm their amorphous state. Magnetic and magnetostrictive hysteresis loops were measured at room temperature using a VSM (Vibrating Sample Magnetometer) and an electrical resistance strain gauge meter with a precision of 10^{-6}, respectively. The magnetostriction samples were measured in parallel ($\lambda\|$) and in perpendicular ($\lambda\perp$) to the direction of the magnetic field. Measurements of Curie (T_C) and crystalisation (T_X) temperatures were carried out using the magnetic balance while the samples were heated at the rate of 10 ℃/sec.

The samples were heat treated at 523 K for 3 hours at a pressure of 2×10^{-6} Torr in order to release the internal stress and to homogenize the amorphous state. After the heat treatment, the amorphous state of specimens was again confirmed by the XRD measurement.

3. Results and Discussion

The changes in crystallization (T_X) and Curie (T_C) temperatures of the heat treated amorphous $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}B_x$ alloys as a function of boron content are shown in Fig. 1. As boron is added to the amorphous alloys, T_X is reduced by about 100 K, but T_C remains constant. The effect of the boron on the both amorphous alloys is known to be the destablization of the amorphous structure(12), although it does not effect electronic structure. Magnetic and electric properties of the amorphous alloys are usually determined by their localized electron structures(13)(14). Since giant magnetostriction is defined by the electronic structure, it will be maintained within the relaxed amorphous structure of both amorphous alloys by means of the addition of boron.

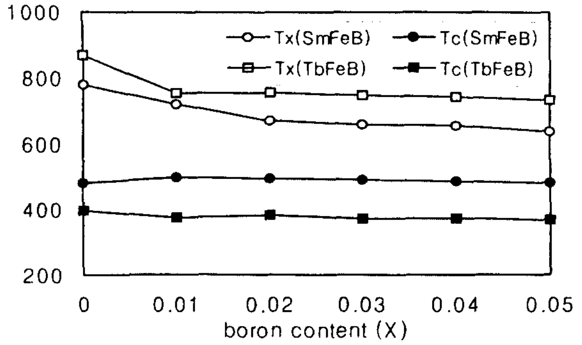


Fig. 1. Crystallization temperature (T_X) and Curie temperature (T_C) for the heat treated amorphous $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}B_x$ alloys with different boron content

Fig. 2 shows changes in the room temperature magnetization (M_S) of heat treated amorphous $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}B_x$ alloys as a function of boron content at 10 kOe. The M_S in both of the amorphous alloys slightly rise with the initial increase of boron content, but then drastically decrease with the increase of the boron content. The boron may be located, either in an interstitial site or a substitutional site. At low concentration, boron probably enters the interstitial site of the DRP (Dense Random Packing) structure in the amorphous alloys. As boron located at the interstitial sites would increase the distance of Fe atoms, there would be an enhancement of saturation magnetization(9). On the other hand, as the boron further increases, some of the excess boron could substitute with Fe atoms in the amorphous alloys and the p-d band exchange coupling between the electrons of boron and Fe could be formed(5)(15). The electrons extracted from the 2p-oribit of the boron acts as an electron donor and transit to the 3d-oribit of the Fe atoms to make the exchange coupling with uncoupled electrons in the 3d-oribit. This means that the sub-magnetic moment of Fe atoms decrease and magnetization in the both of the amorphous alloys, which are ferri-magnetism, decreases. Changes in the magnetization of the alloys will be proportional to those in magnetostriction.

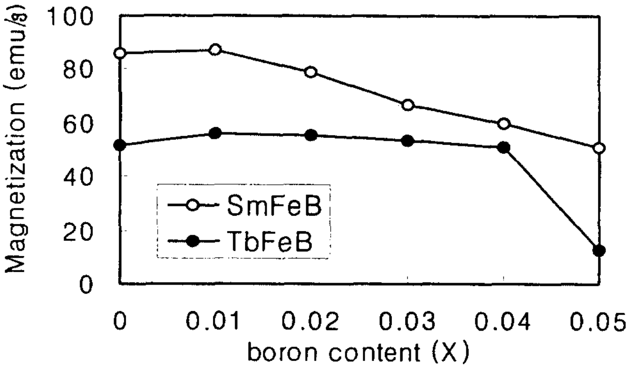
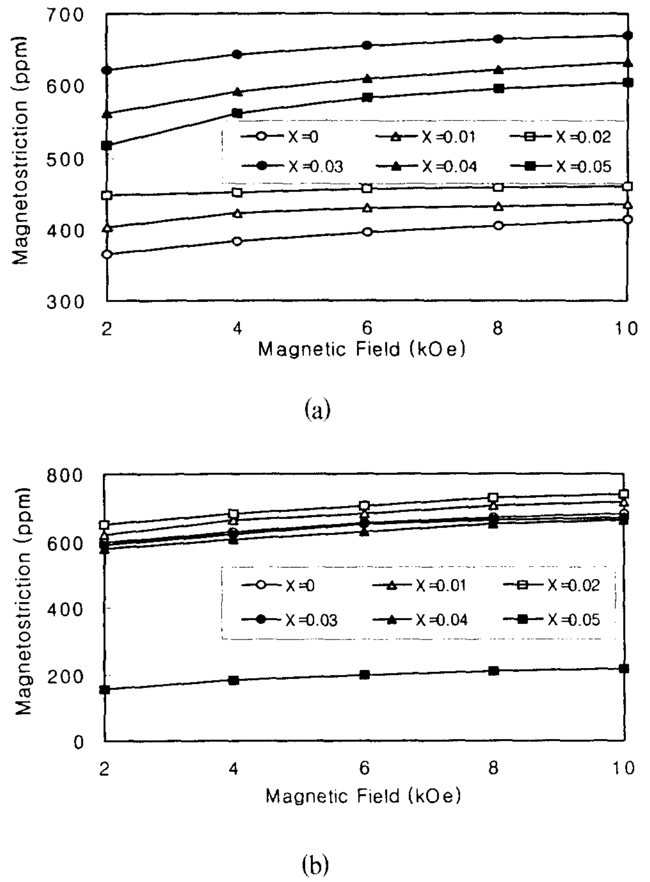


Fig. 2. Room-temperature magnetization (M_S) at 10 kOe of heat treated amorphous $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}B_x$ alloys with respects to boron content
(○ and ● represent for M_S of amorphous $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}B_x$ alloys, respectively)

The tendency of magnetostriction ($\lambda\| - \lambda\perp$) of the heat treated amorphous $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}B_x$ alloys with different boron content with respect to external magnetic fields up to 10 kOe are shown in Fig. 3 (A) and (B), respectively. Magnetostriction of amorphous $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}B_x$ alloys originally shows negative and positive values, respectively, similar to that shown by the SMFe$_2$ and TbFe$_2$ Laves phase intermetallic compounds(3) (6). For an easy of comparison, the absolute values of $(\lambda\| - \lambda\perp)$ in the amorphous $(SmFe_2)_{1-x}B_x$ alloys were used, on the basis that addition of boron and amorphisation of SMFe$_2$ and TbFe$_2$ intermetallic compounds cannot change the electron structure(13) (14), as mentioned in the discussion about Fig. 1.

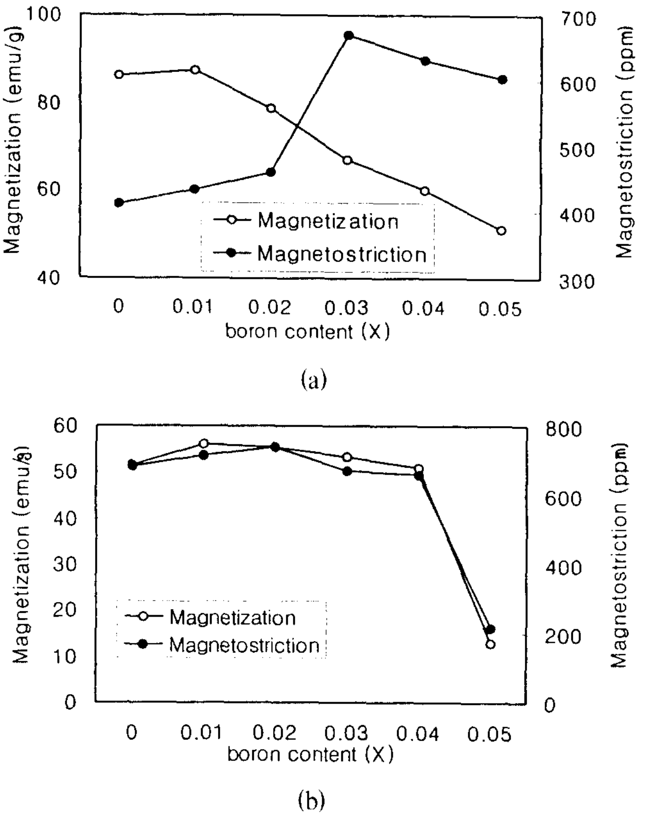


(a)

(b)

Fig. 3. (a) and (b): Magnetostriction $(\lambda\| - \lambda\perp)$ at 10 kOe for the heat treated amorphous $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}B_x$ alloys, respectively in terms of a external magnetic field

In the case of the amorphous $(SmFe_2)_{1-x}B_x$ alloys, the values of $(\lambda\| - \lambda\perp)$ increase with the increasing boron content up to X = 0.02, constantly. For X = 0.02, the magnetostriction curve seems to indicate the saturation magnetostriction of the amorphous SMFe$_2$ alloy. However, further increasing in boron content beyond X = 0.02 results in the anomalous increment of magnetostriction, especially for X = 0.03. Comparison the magnetostriction with the magnetization of the amorphous $(SmFe_2)_{1-x}B_x$ alloys is shown in Fig. 4 (A). The initial constant increment of the magnetostriction is proportional to that of magnetization, resulted from boron filling the interstitial site as shown in Fig. 2. However, anomalous increase of magnetostriction at X = 0.03 cannot simply explain by decrease of the magnetization. It is reasonable to describe that the changes of the magnetostriction in the amorphous $(SmFe_2)_{1-x}B_x$ alloys behaves discretely with the magnetization, depending on whether boron site is interstitial or substitutional. This will be demonstrated in Fig. 6. In the substitutional site of boron content at X = 0.03, magnetostriction proportionally decreases to magnetization, similar to interstitial site behaviors.



(a)

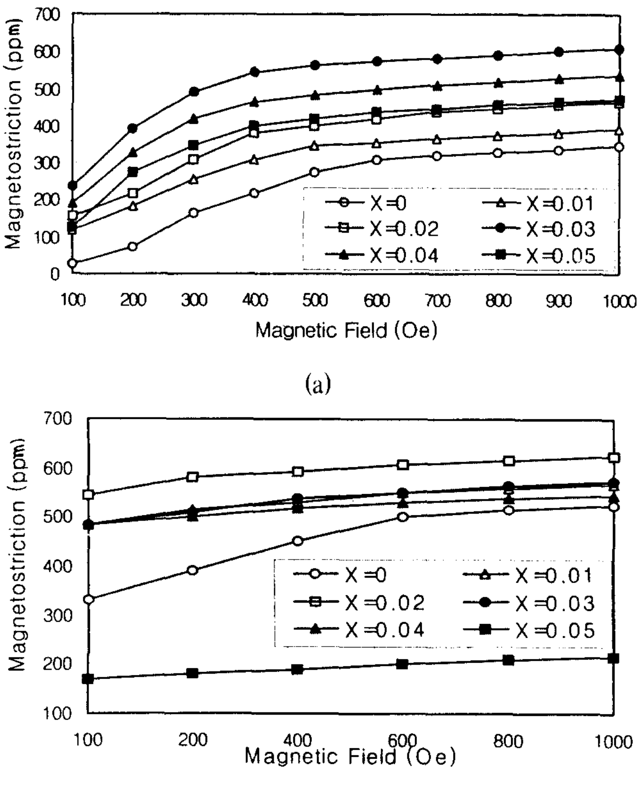
(b)

Fig. 4. (a) and (b): Comparison of magnetostriction $(\lambda\| - \lambda\perp)$ with magnetization (M_s) at 10 kOe for the heat treated amorphous $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}B_x$ alloys, respectively with different boron content

The values of $(\lambda\| - \lambda\perp)$ for the amorphous $(TbFe_2)_{1-x}B_x$ alloys as shown in Fig. 3 (B) does not seem to change significantly when compared with boron content, except for X = 0.05 and are larger than those for the amorphous $(SmFe_2)_{1-x}B_x$ alloys, like Laves phase intermetallic compounds(2). In the case of the amorphous $(TbFe_2)_{1-x}B_x$ alloys, the role of boron atoms for interstitial or substitutional role is not clearly distinguished, unlike the amorphous $(SmFe_2)_{1-x}B_x$ alloys shown in Fig. 3 (A). This is due to the difference in atomic sizes of Tb and Sm in short range of Laves phase structure in the amorphous $(RFe_2)_{1-x}B_x$ alloy. The larger Tb atoms allow smaller interstitial space, which will be more quickly filled with boron atoms. So the interstitial boron atoms hardly exist in the amorphous $(TbFe_2)_{1-x}B_x$ alloys. This results in amorphous $(TbFe_2)_{1-x}B_x$ alloys mainly showing anomalous giant magnetostriction as a function of substitutional boron atoms. In substitutional range, magnetostriction is proportional to magnetization as shown in Fig. 4(B), similar to Fig. 4(A).

Fig. 5 (A) and (B) show the changes in the magnetostriction of the heat treated amorphous $(SmFe_2)_{1-x}B_x$ and $(TbFe_2)_{1-x}$

B$_x$ alloys with varying boron content in a low external magnetic field. For the amorphous (SmFe$_2$)$_{0.97}$ B$_{0.03}$ alloys shown in Fig. 5 (A), the effective magnetostriction of -545 and -610 $\times$ 10^{-6} is obtained at 400 and 1,000 Oe, respectively. Moreover, the effective magnetostriction of 590 and 630 $\times$ 10^{-6} in the amorphous (TbFe$_2$)$_{0.98}$ B$_{0.02}$ alloys shown in Fig. 5 (B) is also found at 400 and 1,000 Oe, respectively. Even though the largest saturation magnetostriction in a high magnetic field (roughly 25 kOe) was obtained from RFe$_2$ Laves phase intermetallic compounds, as reported by A. Clark *et al.*[2], the largest effective magnetostriction in a low magnetic field is reported in this research. These anomalous magnetostrictions phenomena appear as the boron atoms occupy substitutional sites in amorphous (SmFe$_2$)$_{1-x}$ B$_x$ and (TbFe$_2$)$_{1-x}$ B$_x$ alloys. This discovery of the largest effective giant magnetostriction will provide the clue for the development of new magnetostrictive materials with large effective value and a new era for the application of giant magnetostriction.



(a)

(b)

Fig. 5. (a) and (b): Magnetostriction curves of the heat treated amorphous (SmFe$_2$)$_{1-x}$ B$_x$ and (TbFe$_2$)$_{1-x}$ B$_x$ alloys, respectively in a low external magnetic field.

Fig. 6 shows the schematic diagram for the origin of magnetostriction governed by magnetoelastic energy and elastic energy of materials. The former and the latter are proportional to the first order and the second order of lattice strain[16]. The saturation magnetostriction (λ_{S1}) of amorphous SmFe$_2$ and TbFe$_2$ alloys is determined by the minimum sum of magnetoelastic energy without the boron and elastic energy[16] in those alloys. As the magnetoelastic energy with the boron is decreased by the p-d electron band exchange coupling between boron atoms and Fe atoms in the range of the substitutional sites, elastic energy in the alloys increases to reach the new minimum sum energy and leads to the new enlarged saturation magnetostriction (λ_{S2}) in the amorphous (SmFe$_2$)$_{1-x}$ B$_x$ and (Tb Fe$_2$)$_{1-x}$ B$_x$ alloys. This is the hypothetic interpretation for the anomalous giant magnetostriction in the amorphous RFe$_2$ alloys with the substitutional boron addition.

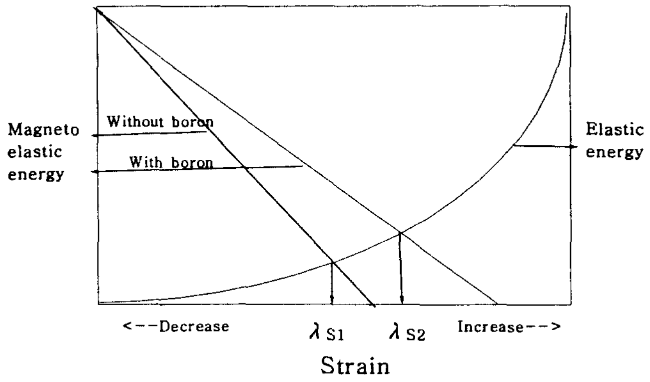


Fig. 6. Schematic diagram for origin of anomalous giant magnetostriction

4. Conclusions

Discovery of effective giant magnetostriction in a low magnetic field can be achieved by the substitutional addition of boron and amorphisation of RFe$_2$ (R = Sm and Tb) Laves phase intermetallic compounds. The effective magnetostriction of -545 $\times$ 10^{-6} and 590 $\times$ 10^{-6} at 400 Oe for amorphous (Sm Fe$_2$)$_{1-x}$ B$_x$ and (TbFe$_2$)$_{1-x}$ B$_x$ alloys is the highest reported values in external magnetic field of a few hundreds Oe. Until now, this phenomena has never been reported in the field of giant magnetostriction. Since p-d exchange coupling does not affect Sm and Tb but affects Fe, the phenomena will have the effect on all amorphous RFe$_2$ alloys. From the application and research point of view, the amorphous (RFe$_2$)$_{1-x}$ B$_x$ alloys should attract great attention as giant magnetostrictive materials.

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