Structural, Magnetic, and Magnetostrictive Properties of $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$ Alloys

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***Abstract*—The crystal structure, magnetization, and spontaneous magnetostriction of $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$ alloys have been investigated. X-ray diffraction (XRD) analysis shows that the alloys consist predominantly of the cubic Laves phase with a $MgCu_2$-type structure for $x \leq 0.55$. Increasing in lattice parameter and decreasing in Curie temperature with increasing x from 0 to 0.55 reveals an increment in the Nd content in the Laves phase. The (440) XRD line of the Laves phase splits doubly when $x \leq 0.7$ because of a large spontaneous magnetostriction along its $\langle 111 \rangle$ easy magnetization direction. The magnetostriction coefficient λ_{111} decreases significantly with increasing x when $0 \leq x \leq 0.55$. For $x \geq 0.7$, it is difficult to achieve saturation magnetization at room temperature due to increased amount of the $Nd_2Fe_{14}B$-type phase with large magnetocrystalline anisotropy.**

***Index Terms*—Crystal structure, Laves phase, magnetization, magnetostriction.**

I. INTRODUCTION

TERFENOL-D ($Tb_{0.30}Dy_{0.70}Fe_{1.92}$), a rare-earth-iron alloy possessing the well-known giant magnetostriction and low magnetocrystalline anisotropy at room temperature, has been an important magnetostrictive transducer material since its discovery in the 1970s [1], [2]. However, the main raw materials of Terfenol-D are expensive: Tb and Dy. It would be beneficial to applications if we would find a novel magnetostrictive compound based on the low-cost light rare-earths, like Pr or Nd. Moreover, the $NdFe_2$ compound has a large theoretical magnetostriction at 0 K [3], and its easy-magnetization direction (EMD) lies along the $\langle 100 \rangle$ axis at room temperature [4]. The composition anisotropy compensation should be achievable in the pseudobinary $Tb_{1-x}Nd_xFe_2$ system. However, little information has been available on this system. The reason is mainly attributed to difficulties in synthesizing the Laves phase with a high Nd content under an atmospheric pressure [5].

We have recently demonstrated in a pseudobinary $Tb_{1-x}Pr_xFe_2$ system and a multicomponent pseudobinary $Tb_xDy_{1-x-y}Pr_yFe_2$ system that the introduction of a small amount of B effectively restrains the emergence of the $PuNi_3$-type phase, thereby promoting the formation of the cubic Laves phase with a high Pr content [6]–[8]. In this paper, a small amount of B is introduced into the pseudobinary $Tb_{1-x}Nd_xFe_2$ system to stabilize the Nd-containing Laves phase. The structural, magnetic, and magnetostrictive properties of the resulting $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$ alloys are studied.

II. EXPERIMENT

Polycrystalline samples of $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$ alloys with $x = 0, 0.2, 0.4, 0.55, 0.6, 0.7, 0.8,$ and 1.0 were prepared by arc-melting the appropriate constituent metals in a high-purity argon atmosphere. The purities of the constituents were 99.9% for Tb, Nd, and B, and 99.8% for Fe. The ingots were homogenized at 700 °C for seven days in a high-purity argon atmosphere. X-ray diffraction (XRD) data was recorded at room temperature with $\mathrm{Cu}K_\alpha$ radiation in a Riguku D/max-2500pc diffractrometer equipped with a graphite crystal monochromator. A high-precision XRD step scanning with Cu K_α radiation was performed on the samples in powder form for the (440) peaks of the Laves phase so as to investigate the spontaneous peak splitting induced by magnetostriction. The magnetostriction constant λ_{111} was determined from the split distance of the double-splitted (440) lines of the Laves phase after the effect of the $K_{\alpha 2}$ radiation was removed [6], [9]. The Curie temperature was deduced from the temperature dependence of ac initial susceptibility (χ_{ac}) at a magnetic field of 160 A/m and a frequency of 1.13 kHz. The magnetization versus field $(M - H)$ curve was measured with a PAR155-type vibrating sample magnetometer at room temperature for magnetic fields up to 600 kA/m.

III. RESULTS AND DISCUSSIONS

Fig. 1 shows the XRD patterns of some of the homogenized $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$ alloys. It is found that the homogenized samples consist predominantly of the cubic Laves phase with a $MgCu_2$-type structure and a small amount of rare earth as the impurity phase when $x \leq 0.55$. In the region of $0.55 < x \leq 0.8$, the cubic Laves phase coexists with the $Nd_2Fe_{14}B$-type phase and some amount of the rare-earth phase in such a way that the amount of the cubic Laves phase

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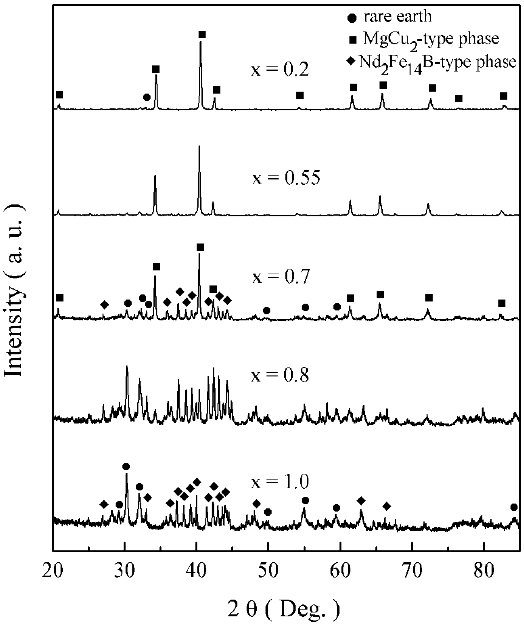

Fig. 1. XRD patterns of homogenized $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$ alloys.

decreases while that of the others increases with increasing x. The cubic Laves phase totally disappears at $x = 1.0$, leaving only the $Nd_2Fe_{14}B$-type phase and some amount of Nd. The $PuNi_3$-type phase is not observed in the alloys studied. This is mainly caused by the substitution effect of B, which effectively extends the cubic Laves phase to high Nd contents. The result is consistent with our previous reports on $Tb_{1-x}Pr_xFe_{1.93}B_{0.15}$ and $Tb_{0.2}Dy_{0.8-x}Pr_x(Fe_{0.9}B_{0.1})_{1.93}$ alloys that B is helpful for the formation of the cubic Laves phase with a high Pr content [6]–[8].

The composition x dependence of the lattice parameter a and Curie temperature T_c of the Laves phase in the alloys is shown in Fig. 2. For $x \leq 0.55$, a increases from 0.7344 nm to 0.7391 nm while T_c decreases from 698 K to 640 K when x is increased from 0–0.55. This is due to the larger radius of Nd^{3+} than that of Tb^{3+} and the lower T_c of $NdFe_2(= 578\ K)$ than that of $TbFe_2(= 704\ K)$. For $0.55 < x \leq 0.8$, the increase in a becomes less pronounced, while T_c almost keeps invariant. This implies that the Nd content in the Laves phase is lower than its nominal composition of the corresponding alloy, owing to the appearance of other Nd-containing phases.

The step-scanned XRD profiles of the (440) line of the Laves phase in the alloys with $x \leq 0.7$ are illustrated in Fig. 3. It is clear that all profiles split doubly and overlap partially. This concludes that the EMD of the Laves phase lies along the $\langle 111 \rangle$ axis, and a large rhombohedral distortion in the crystal structure occurs due to magnetostriction [9]. The EMD of the Laves phase of the $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$ alloys does not change from $\langle 111 \rangle$ to $\langle 100 \rangle$ axis when x is increased. Because the Laves phase with high Nd content whose EMD should lie along $\langle 100 \rangle$ axis has not been attained due to that the composition of the Laves phase remains almost unchanged when $x > 0.55$ (Fig. 2). In other words, the composition anisotropy compensation has not been achieved in $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$ system.

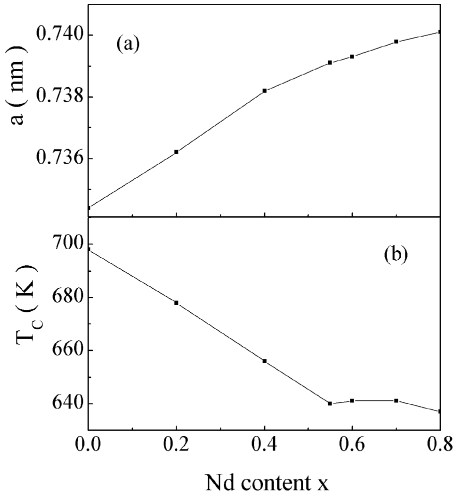

Fig. 2. Composition x dependence of (a) lattice parameter a and (b) Curie temperature T_c of the Laves phase in $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$ alloys.

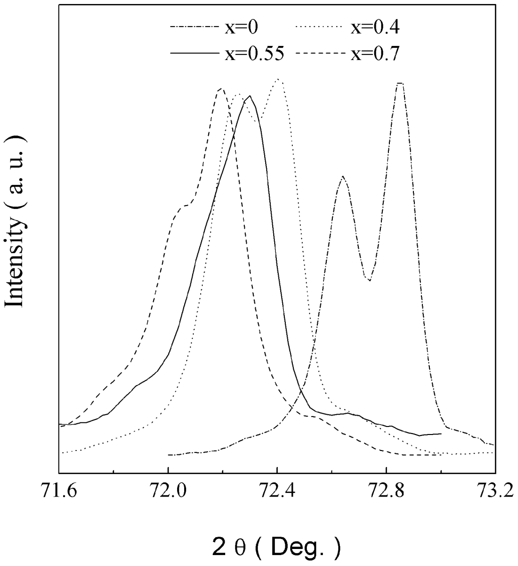

Fig. 3. Step-scanned XRD profiles of the (440) line of the Laves phase in $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$ alloys.

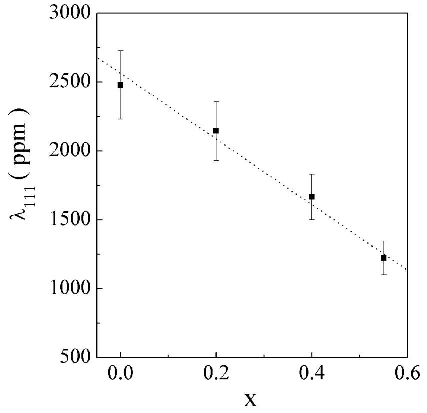

Fig. 4. Composition x dependence of magnetostriction coefficient λ_{111} of the Laves phase in $Tb_{1-x}Nd_x(Fe_{0.9}B_{0.1})_2$ alloys.

The composition x dependence of the magnetostriction coefficient λ_{111} of the Laves phase in the alloys with $0 \leq x \leq 0.55$ is plotted in Fig. 4. λ_{111} decreases monotonically from 2478 ppm to 1224 ppm when x is increased from 0 to 0.55, indicating that λ_{111} of the $Nd(Fe,B)_2$ Laves phase is much smaller than

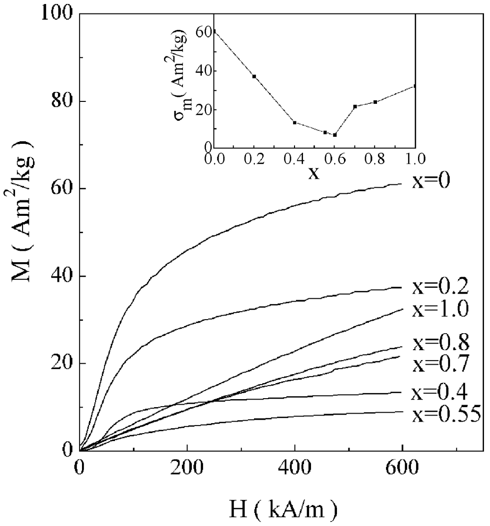

Fig. 5. Magnetization M versus magnetic field H curves for $\mathrm{Tb}_{1-x}\mathrm{Nd}_x(\mathrm{Fe}_{0.9}\mathrm{B}_{0.1})_2$ alloys at room temperature. The inset is the composition x dependence of the maximum magnetization $\sigma_{\max}$.

that of the $\mathrm{Tb(Fe,B)}_2$ Laves phase at room temperature. Thus, NdFe_2 should have a much smaller λ_{111} than TbFe_2 according to the model of Cullen and Clark [10].

The magnetization M versus magnetic field H curves for the alloys at room temperature are shown in Fig. 5. For $0 \leq x \leq 0.55$, M is close to be saturated by H at 600 kA/m but far from saturation for $0.55 < x \leq 1$. The result is in good agreement with those of the structural analysis given above. The main phase of the alloys with $0 \leq x \leq 0.55$ is the $\mathrm{(Tb, Nd)(Fe, B)}_2$ cubic Laves phase, while that of the alloys with $0.55 < x \leq 1.0$ consists of a large amount of the $\mathrm{Nd_2Fe_{14}B}$-type phase with a much larger magnetocrystalline anisotropy than that of the Laves phase. The composition dependence of the maximum magnetization $\sigma_{\max}$ of the alloys is shown as an inset in Fig. 5. $\sigma_{\max}$ decreases with increasing x when $0 \leq x \leq 0.6$. In fact, the coupling between the Nd and Fe moments is parallel, whereas the coupling between the Tb and Fe moments is antiparallel. Usually, the moments of Tb and Nd ions align antiparallelly in the Laves phase. It is well understood that the substitution of Nd for Tb in alloys leads to a decrease in M. When $0.6 < x \leq 1.0$, $\sigma_{\max}$ increases with increasing x, subjected to the increased amount of the $\mathrm{Nd_2Fe_{14}B}$-type phase.

IV. CONCLUSION

The crystal structure, magnetization, and spontaneous magnetostriction of $\mathrm{Tb}_{1-x}\mathrm{Nd}_x(\mathrm{Fe}_{0.9}\mathrm{B}_{0.1})_2$ alloys have been studied. The homogenized alloys comprise mainly the cubic Laves phase with a MgCu_2-type structure when $x \leq 0.55$. The lattice parameter of the Laves phase in the alloys increases, while the Curie temperature decreases with increasing Nd content at $x \leq 0.55$. Practical magnetostrictive materials with low magnetocrystalline anisotropy cannot be achieved in the $\mathrm{Tb}_{1-x}\mathrm{Nd}_x(\mathrm{Fe}_{0.9}\mathrm{B}_{0.1})_2$ system because the composition of the Laves phase in alloys remains almost unchanged when $0.55 < x \leq 0.8$. The magnetostriction coefficient λ_{111} of the Laves phase decreases with increasing x when $0 \leq x \leq 0.55$. Saturation magnetization cannot be achieved at room temperature for $x \geq 0.7$.

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