High Pr-content \((\text{Tb}_{0.2}\text{Pr}_{0.8})(\text{Fe}_{0.4}\text{Co}_{0.6})_{1.93-x}\text{B}_x\) magnetostrictive alloys

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High Pr-content \((\text{Tb}_{0.2}\text{Pr}_{0.8})(\text{Fe}_{0.4}\text{Co}_{0.6})_{1.93-x}\text{B}_x\) magnetostrictive alloys \((0.05 \leq x \leq 0.15)\) with a single cubic Laves phase were synthesized by arc-melting and subsequent annealing. The Curie temperature, \(T_c\), and the lattice parameter, \(a\), of the Laves phase in the alloys increase with the boron content up to \(x = 0.10\), which is ascribed to the preferential occupation of boron in the interstitial sites of the Laves phase. The addition of a small amount of boron to the \((\text{Tb}_{0.2}\text{Pr}_{0.8})(\text{Fe}_{0.4}\text{Co}_{0.6})_{1.93}\) alloy reduces the magnetocrystalline anisotropy constant \(K_1\) and improves the magnetostriction \(\lambda_p = \lambda_{\parallel} - \lambda_{\perp}\) at relatively low magnetic fields at room temperature. The composition dependence of the ratio \(\lambda_p/K_1\) for \((\text{Tb}_{0.2}\text{Pr}_{0.8})(\text{Fe}_{0.4}\text{Co}_{0.6})_{1.93-x}\text{B}_x\) alloys reaches a maximum value at \(x = 0.10\), suggesting that the \((\text{Tb}_{0.2}\text{Pr}_{0.8})(\text{Fe}_{0.4}\text{Co}_{0.6})_{1.8}\text{B}_0.1\) alloy should be a good candidate material for magnetostrictive applications.

The C15 cubic Laves phase compounds \((R, R' \text{Fe}_2 \quad (R, R' \text{Fe}_2 \quad \text{rare} \text{earths}), \text{such as} \text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2 \quad (\text{Terfenol-D}), \text{have been widely applied as magnetostrictive} \text{materials} \text{in} \text{acoustic} \text{transducers,} \text{sensors,} \text{actuators,} \text{etc.}^{1,2} \) The single-ion model indicated that the giant magnetostrictive effect between the electron spin and spatially anisotropic \(4f\) charge, and predicted that the \(\text{PuNi}_3\)-type phase and that boron is helpful for the formation of Cu K\(_{\alpha}\) radiation in a Rigaku D/max-2500pc diffractometer with a graphite crystal monochromator. High-hold precision XRD step scanning was performed for the \((440)\) line of the Laves phase and then the effect of the K\(_{\alpha2}\) radiation was removed with a standard method, in order to investigate its spontaneous magnetostriction coefficient \(\lambda_{111}\). Magnetization curves at room temperature were measured by a quantum design superconducting quantum interference device (SQUID) magnetometer at fields up to 50 kOe. The magnetostriction at room temperature was measured either parallel or perpendicular to the applied field using a standard strain gauge technique.

XRD patterns of homogenized \((\text{Tb}_{0.2}\text{Pr}_{0.8})(\text{Fe}_{0.4}\text{Co}_{0.6})_{1.93-x}\text{B}_x\) alloys are shown in Fig. 1. It is seen that the matrix of \((\text{Tb}_{0.2}\text{Pr}_{0.8})(\text{Fe}_{0.4}\text{Co}_{0.6})_{1.93}\) alloy consists predominantly of the cubic Laves phase with \(\text{MgCu}_2\)-type structure, coexisting with a small amount of impurity phase with \(\text{PuNi}_3\)-type structure. Homogenized B-containing alloys, up to \(x = 0.15\), are almost single Laves phase. As the boron content further increases, some weak peaks emerge in the XRD patterns, which belong to the excessive rare earth phase. No trace of the noncubic \(\text{PuNi}_3\)-type phase is found in all the boron-containing alloys. This result shows that a small amount of boron can effectively inhibit the appearance of the \(\text{PuNi}_3\)-type phase and that boron is helpful for the formation

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of the (Tb, Pr)Fe₂ Laves phase, in consistent with the previous report.⁸

The concentration dependence of the lattice parameter a of the Laves phases in (Tb₀.２Pr₀.８)(Fe₀.４Co₀.６)₁.₉₃−ₓBₓ alloys is shown in Fig. 2(a). The lattice parameter increases with increasing the boron concentration up to x=0.10 and then decreases in the range of 0.15≤x≤0.40. This suggests that the boron atoms can occupy either the substitutional or interstitial site. Because of its small size, the boron occupies preferentially the interstitial sites until its concentration reaches the solubility limit x=0.10 (about 3.41 at % in this system). Above this value, the boron atom starts to occupy the Fe/Co sites in the lattice, which leads to the lattice contraction.

The Curie temperature of all the alloys was detected from the measurements of AC initial susceptibility χAC. Two peaks were observed at 418 and 621 K, in the $\chi_{AC} - T$ curves for the (Tb₀.２Pr₀.８)(Fe₀.４Co₀.６)₁.₉₃ alloy. The former corresponds to the Curie temperature of the Laves phase, while the latter is for the PuNi₃-type phase. As the boron is intro-

duced into the lattice, only does one peak appear in the $\chi_{AC} - T$ curve, which is attributed to the Curie temperature of the Laves phase. It is in consistence with the XRD result that the single phase with the Laves structure forms, free of the PuNi₃-type phase. The concentration dependence of the Curie temperature $T_c$ for the Laves phases in (Tb₀.２Pr₀.８)(Fe₀.４Co₀.６)₁.₉₃−ₓBₓ alloys is represented in Fig. 2(b). The Curie temperature $T_c$ increases with increasing the boron concentration in the alloys when 0≤x≤0.10, indicating that the exchange interaction in the Laves phase enhances since the boron atoms occupy the interstitial sites.⁸,¹¹ Then the Curie temperature $T_c$ decreases when 0.15≤x≤0.40, because of the weakening of the exchange interaction in the Laves phase when the boron atoms substitute the Fe/Co atoms.

The magnetic field dependence of the magnetization at room temperature for the (Tb₀.２Pr₀.８)(Fe₀.４Co₀.６)₁.₉₃−ₓBₓ (0≤x≤0.20) alloys is shown in Fig. 3. The magnetization at the maximum available magnetic field of 50 kOe decreases with increasing the boron content x. It is well-known that in the rare-earth–transition-metal compounds there are parallel (or antiparallel) alignments between the magnetic moments of praseodymium (or terbium) and iron/cobalt atoms. The magnetic moment μ of the (Tb₀.２Pr₀.８)(Fe₀.４Co₀.６)₁.₉₃−ₓBₓ Laves phase can be described as

$$μ = 0.8μ_{Pr} - 0.2μ_{Tb} + (1.93-x)μ_{Fe/Co}. \quad (1)$$

From this formula it would be not difficult to understand why the magnetization decreases with increasing x, if the influence of the small amount of impurities phase on the magnetization were neglected.

The magnetcocrystalline anisotropy constant $K_1$ of the (Tb₀.２Pr₀.８)(Fe₀.４Co₀.６)₁.₉₃−ₓBₓ Laves phase was determined by simulating the M-H curves using the approximate law of the saturation as follows:¹²

$$M = M_s \left(1 - \frac{a}{H} - \frac{b}{H^2}\right) + \chi_B \cdot H, \quad (2)$$

and the relation¹²
The spontaneous magnetostriction constant $\lambda_s$ for the (Tb$_{0.2}$Pr$_{0.8}$)(Fe$_{0.4}$Co$_{0.6}$)$_{1.93}$-B$_x$ alloys is helpful for the formation of the cubic Laves phase. High Pr-content (Tb$_{0.2}$Pr$_{0.8}$)(Fe$_{0.4}$Co$_{0.6}$)$_{1.93}$-B$_x$ (0.05 $\leq$ $x$ $\leq$ 0.15) alloys with a single Laves phase have been synthesized. The addition of the small amount of boron enhances the exchange interaction as well as the Curie temperature $T_c$ of the Laves phase. $K_1$ and $\lambda_{111}$ of the Laves phase in the (Tb$_{0.2}$Pr$_{0.8}$)(Fe$_{0.4}$Co$_{0.6}$)$_{1.93}$-B$_x$ alloys decrease with increasing the boron content. The (Tb$_{0.2}$Pr$_{0.8}$)(Fe$_{0.4}$Co$_{0.6}$)$_{1.83}$B$_{0.1}$ alloy with a single Laves phase has a large $\lambda_s$ at relatively low magnetic field and a large $\lambda_s/K_1$, which should be a potential candidate for use as a magnetostrictive material.

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