Spin reorientation and magnetocrystalline anisotropy of $(\text{Nd}_{1-x}\text{RE}_x)_2\text{Fe}_{14}\text{B}$ (RE=Y,Pr)

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The substitutional effect of Y or Pr for Nd on spin reorientation and magnetocrystalline anisotropy of $(\text{Nd}_{1-x}\text{RE}_x)_2\text{Fe}_{14}\text{B}$ ($0 \leq x \leq 0.75$) has been investigated from magnetization curves of magnetically aligned powders. The experiments have shown that Y- or Pr-substitution for Nd decreases both the spin reorientation temperature (T_{SR}) and spin reorientation angle. The decrement of T_{SR} by Y-substitution was within 15 K. The Pr-substitution, however, considerably decreased T_{SR} with the ratio of $\Delta T_{SR} = -1.35$ K/Pr at.%. The decrement of spin reorientation angle in the composition range of $0 \leq x \leq 0.5$ is about $\Delta SRA = -0.073^\circ/\text{Y}$ at.% and $\Delta SRA = -0.258^\circ/\text{Pr}$ at.%, respectively. The experiments on magnetocrystalline anisotropy expect the disappearance of spin reorientation at about $x > 0.9$ for RE=Y and at about $x > 0.8$ for RE=Pr at 4.2 K.

Key words : spin reorientation, rare earth compounds, magnetocrystalline anisotropy

1. Introduction

$\text{Nd}_2\text{Fe}_{14}\text{B}^{1,2)}$ shows strong uniaxial magnetocrystalline anisotropy with easy magnetization of tetragonal c -axis at room temperature. Below the spin reorientation temperature of 135 K³⁾, however, the easy axis of the compound tilts away from c -axis by certain angle θ within {110} plane, and to show cone anisotropy. The tilting angle increases according to the decrease of temperature and reaches to $\theta \approx 30^\circ$ ^{3) 5)} at 4.2 K. The spin reorientation depends on magnetocrystalline anisotropy, and the effects of anisotropy constants^{6),7)} or rare earth elements and 3d transition metal have been examined⁸⁾.

$\text{Y}_2\text{Fe}_{14}\text{B}$ and $\text{Pr}_2\text{Fe}_{14}\text{B}$ show uniaxial magnetocrystalline anisotropy below Curie temperature (T_c). The substitution of Nd by Y or Pr, therefore, is expected to suppress the spin reorientation by changing the anisotropy. In this paper, we report the effects of Y- and Pr-substitution on spin reorientation and magnetocrystalline anisotropy of $\text{Nd}_2\text{Fe}_{14}\text{B}$ -type

compounds.

2. Experimental details

The alloys of $(\text{Nd}_{1-x}\text{RE}_x)_2\text{Fe}_{14}\text{B}$ (RE=Y,Pr : $x=0.25 \sim 0.75$) and $\text{Nd}_2\text{Fe}_{14}\text{B}$ were prepared by arc melting and induction melting pure metals in argon gas atmosphere, respectively. The alloys were annealed at 1173 K for 7 days, and confirmed to be single 2-14-1 phase by x-ray diffractometry and thermomagnetic analysis. In order to measure the magnetization curves, the compounds were ground into powders smaller than 40 μm in size and aligned in a magnetic field of 1.5 T within paraffin. Spin reorientation temperature was determined from the inflection of the temperature-dependent magnetization curves measured perpendicular to the alignment direction under a constant field lower than 0.004 T. The magnetization curves along parallel and perpendicular to the alignment direction were measured by a high field vibrating sample magnetometer under the maximum applied field of 8 T in the temperature range of 4.2 ~ 250 K.

3. Results and discussion

A. Spin reorientation

Fig.1 shows the compositional dependence of T_{SR} for $(\text{Nd}_{1-x}\text{Y}_x)_2\text{Fe}_{14}\text{B}$ and $(\text{Nd}_{1-x}\text{Pr}_x)_2\text{Fe}_{14}\text{B}$ determined from the inflection of temperature-dependent magnetizations measured perpendicular to the alignment direction. The experiments show that T_{SR} decreases by the substitution of Y or Pr for Nd. For example, Pr-substitution decreases T_{SR} linearly with the ratio of $\Delta T_{SR} = -1.35$ K/Pr at.%. The substitution of Y for Nd, however, is insignificant compared to the Pr-substitution, and the decrement of T_{SR} is within 15 K in the composition range of $0 \leq x \leq 0.75$.

Fig.2 shows the compositional dependence of spin reorientation angle (SRA) of $(\text{Nd}_{1-x}\text{Y}_x)_2\text{Fe}_{14}\text{B}$ and $(\text{Nd}_{1-x}\text{Pr}_x)_2\text{Fe}_{14}\text{B}$ at 4.2 K. The SRA was calculated from magnetization curves measured along parallel and perpendicular

Fig.1. The compositional dependence of spin reorientation temperature (T_{SR}) of $(Nd_{1-x}RE_x)_2Fe_{14}B$.

to the alignment direction using the formula⁹⁾ which includes basal plane anisotropy effect. In calculation, the aligned powders were assumed to have a Gaussian distribution, and the alignment was evaluated from magnetization curves measured at 250 K according to the method explained in the literature¹⁰⁾. The calculation results show that SRA decreases by Y- or Pr-substitution. As the same tendency of T_{SR} , the decrement is more profound for the case of Pr-substitution than Y-substitution. For example, the ratio of SRA decrement in the composition range of $0 \leq x \leq 0.5$ is about $\Delta SRA = -0.073^\circ/Y \text{ at.}\%$ and $\Delta SRA = -0.258^\circ/Pr \text{ at.}\%$.

B. Magnetocrystalline anisotropy

The magnetocrystalline anisotropy of $(Nd_{1-x}Y_x)_2Fe_{14}B$ and $(Nd_{1-x}Pr_x)_2Fe_{14}B$ was investigated by a fitting method. In calculation, we assumed the Gaussian distribution of powder, and the magnetization was calculated by the energy minimum of free energy which is described by eq. (1);

$$E = K_1 \sin^2 \theta + K_2 \sin^4 \theta - \vec{\sigma}_s \cdot \vec{H} \quad (1)$$

Here, K_n and $\vec{\sigma}_s$ are the anisotropy constant of n th order and saturation magnetization, respectively.

Fig. 3 shows the magnetocrystalline anisotropy constants of $(Nd_{1-x}Y_x)_2Fe_{14}B$ and $(Nd_{1-x}Pr_x)_2Fe_{14}B$. The temperature dependency of the anisotropy constants is very similar to the case of $Nd_2Fe_{14}B$ ¹¹⁾. For example, $K_1 > K_2 > 0$ at high temperature near 250 K. Upon decreasing temperature, K_1 increases slightly to show the maximum then decreases quickly and changes the sign to negative, while K_2 keeps the positive sign at whole temperature. Substitution

Fig. 2 The compositional dependence of spin reorientation angle (SRA) of $(Nd_{1-x}RE_x)_2Fe_{14}B$ at 4.2 K.

of Y or Pr for Nd lowers the temperature at which K_1 changes the sign. This temperature coincides well with the spin reorientation temperature (Compare with Fig.1), as was expected in the literatures^{6) 8)}.

Fig.4 shows the compositional dependence of K_1, K_2 of $(Nd_{1-x}Y_x)_2Fe_{14}B$ and $(Nd_{1-x}Pr_x)_2Fe_{14}B$ at 4.2 K. (The data for $Nd_2Fe_{14}B$ and $Y_2Fe_{14}B$ were cited from ref.12) and $Pr_2Fe_{14}B$ from ref.13)). From anisotropy equation, it is known that the spin reorientation to cone anisotropy occurs under the condition of $K_1 < 0$ while $K_1 + 2K_2 > 0$. Since the condition $K_1 + 2K_2 > 0$ is satisfied in $(Nd_{1-x}Y_x)_2Fe_{14}B$ and $(Nd_{1-x}Pr_x)_2Fe_{14}B$

Fig.3 The change of magnetocrystalline anisotropy constants K_1 ($\circ$) and K_2 ($\bullet$) of $(Nd_{1-x}Y_x)_2Fe_{14}B$ and $(Nd_{1-x}Pr_x)_2Fe_{14}B$ as a function of temperature.

Fig. 4 The compositional dependence of the anisotropy constants of $(\text{Nd}_{1-x}\text{RE}_x)_2\text{Fe}_{14}\text{B}$ at 4.2 K. The values for $\text{Nd}_2\text{Fe}_{14}\text{B}$ and $\text{Y}_2\text{Fe}_{14}\text{B}$ were cited from ref.12) and $\text{Pr}_2\text{Fe}_{14}\text{B}$ from ref.13).

(See Fig.3), the spin reorientation depends on the sign of K_1 . From the experiments, the spin reorientation is expected to disappear at about $x > 0.9$ for RE=Y and at about $x > 0.8$ for RE=Pr.

4. Conclusion

In $\text{Nd}_2\text{Fe}_{14}\text{B}$, Y- or Pr-substitution for Nd decreases both the spin reorientation temperature and spin reorientation angle. The decrement of T_{SR} by Y-substitution was within 15 K. The Pr-substitution considerably decreased T_{SR} with the ratio of $\Delta T_{SR} = -1.35$ K/Pr at.%. The decrement of spin reorientation angle in the composition range of $0 \leq x \leq 0.5$ is about $\Delta \text{SRA} = -0.073^\circ/\text{Y}$ at.% and $\Delta \text{SRA} = -0.258^\circ/\text{Pr}$ at.%, respectively. The experiments on magnetocrystalline anisotropy expect the disappearance of spin reorientation at about $x > 0.9$ for $(\text{Nd}_{1-x}\text{Y}_x)_2\text{Fe}_{14}\text{B}$ and at about $x > 0.8$ for $(\text{Nd}_{1-x}\text{Pr}_x)_2\text{Fe}_{14}\text{B}$ at 4.2 K.

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