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Magnetic properties and magnetostriction of $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys with the $MgCu_2$-type Laves structure

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Abstract

The crystal structure, magnetic properties and magnetostriction of arc-melted polycrystalline $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ (x from 0.2 to 0.7) alloys were investigated by X-ray diffraction, vibrating sample magnetometer and standard strain gauge techniques. It was found that all samples possess $MgCu_2$-type cubic Laves structure. The lattice parameter exhibits positive deviation from Vegard's law. The Curie temperature decreases with increasing Ce concentration. The magnetostriction at room temperature decreases with increasing Ce concentration, when $0.2 \leq x < 0.4$ and $x > 0.6$, while increases with increasing Ce concentration from 0.3 to 0.6. The anomalies of lattice parameter and magnetostriction are attributed to the valence fluctuation of Ce ions.

Keywords: Magnetic properties; Magnetostriction; $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$; Valence fluctuation; Laves phase

1. Introduction

In the 1960s, it was found that the rare earth possesses giant magnetostriction at 4.2 K [1]. In order to obtain the large magnetostrictive materials at room temperature, many alloys for the rare earth were investigated. It was found that RFe_2 (R = rare-earth) alloys with Laves structure, such as $TbFe_2$, $DyFe_2$ and $SmFe_2$, have large magnetostriction not only at the low temperature but also at room temperature, so plenty of studies on these RFe_2 alloys have been done [2–5]. However, according to the single-ion model, $CeFe_2$ and $PrFe_2$ alloys possess larger magnetostriction than $TbFe_2$, $DyFe_2$ and $SmFe_2$ alloys [6]. In fact, the experiments showed that $CeFe_2$ alloy possesses 60 ppm at 4 K and nearly 0 ppm magnetostriction at room temperature, which is attributed to the mixed valence of Ce ions [1]. Furthermore, it was also found that it is difficult to get $PrFe_2$ alloys at ambient pressure because of the considerable discrepancy from the ideal atomic radius ratio 1.225 for the Laves phase compound [7]. In order to find the new giant magnetostrictive material at room temperature, many studies on Ce and Pr compounds were made [8–13]. In this paper, we will report the phase formation, magnetic properties and magnetostriction of $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys.

2. Experimental

$Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ (x from 0.2 to 0.7) alloys were prepared by arc melting the appropriate amounts of Fe (purity better than 99.99 wt.%) and rare-earth metals (99.9 wt.%) under a purified argon atmosphere. The buttons were turned over and remelted three times in order to ensure homogenization of the compounds. The as-cast ingots were wrapped in tantalum foils and put into an evacuated quartz capsule, then annealed at 850°C for 4 days.

All the polycrystalline samples were investigated by X-ray diffraction measurement using Cu Kα radiation. The lattice parameters of the polycrystalline samples were determined by least-squares fitting to the X-ray powder pattern and the accuracy was estimated to be ±0.002 Å. The Curie temperature was derived from measurement of the magnetization as a function of temperature. The magnetostriction in directions

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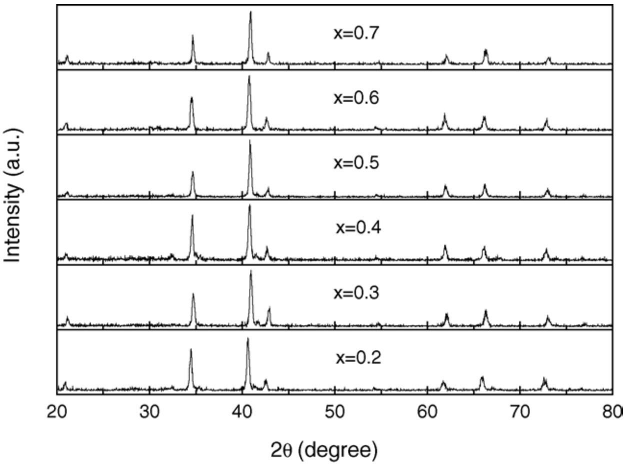

Fig. 1. XRD patterns of homogenized $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys.

of parallel ($\lambda_{//}$) and perpendicular ($\lambda_{\perp}$) to the magnetic field was measured using a standard strain gauge in an applied field up to 1000 kA/m at room temperature.

3. Results and discussion

The room temperature X-ray patterns for $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ ($x = 0.2$–0.7) alloys are shown in Fig. 1. It can be seen from Fig. 1 that all samples exhibit a perfect pure cubic Laves phase of $MgCu_2$-type structure after 850 °C for 4 days.

Fig. 2 shows the lattice parameter dependence of Ce concentration for $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ ($x = 0.2$–0.7) alloys obtained from X-ray diffraction. It can be seen that the lattice parameter of $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys exhibits positive deviation from Vegard's law. In our pervious studies [8,9,11], it was found that some of the rare earth (such as Tb, Dy, Ho) in $Ce_xR_{1-x}Fe_2$ (there R is rare earth) alloys result in the positive deviation of the lattice parameter from Vegard's law, while the other (such as Pr and Nd) result in the negative deviation. In the Ce-based intermetallic compounds, this deviation is attributed to the valence fluctuation of Ce ions, because the ionic radius of localized Ce ions is larger than that of the delocalized Ce ions, the positive deviation is resulted from the fluctuation of Ce ions from delocalized state to localized state. In contrast, the negative deviation is resulted from localized state to delocalized state. In our studied system, $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys, the Pr concentration is constant, and the valence fluctuation of Ce ion is attributed to the change of Tb concentration, so a similar dependence of the lattice parameter on x has been found for $Ce_xTb_{1-x}Fe_2$ system [11]. It can be seen from Fig. 2 that the deviation from Vegard's law exhibits a maximum value at $x = 0.5$, which is consistent with the anomalies of magnetostriction at this point.

Fig. 3 shows the Curie temperature (T_c) dependence of Ce concentration for $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys. It was found that Curie temperature decrease from 603 to 377 K with increasing Ce concentration from 0.2 to 0.7. It is well known that the Curie temperature of $CeFe_2$ (235 K) is far lower than that of $TbFe_2$ (704 K) [1], therefore the Curie temperature decreases with increasing Ce concentration in $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys.

Fig. 4 shows the magnetostriction ($\lambda_{//}$) in direction of parallel to magnetic field for $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys at room temperature. It can be seen that the magnetostriction does not saturate at the magnetic field of 1000 kA/m when $x \leq 0.5$, while reaches saturation at the low magnetic field when $x > 0.5$. During the measurement, the hysteresis has been observed when $x < 0.4$, especially at $x = 0.3$, while no hysteresis has been observed when $x > 0.4$.

Fig. 5 shows the magnetostriction ($\lambda_{//} - \lambda_{\perp}$) dependence of the applied magnetic field for $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys. The result of magnetostriction at the room temperature showed that the magnetostriction decreases with increasing Ce content in the range of $0.2 \leq x < 0.4$ and $x > 0.6$, but

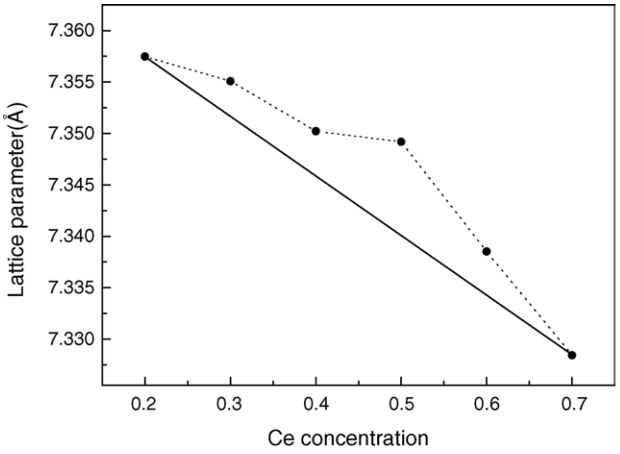

Fig. 2. The dependence of the lattice parameter on Ce concentration for $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys. The solid line represents Vegard's law behavior.

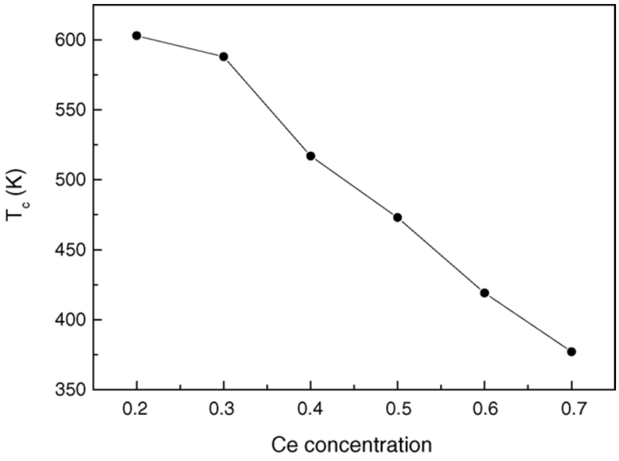

Fig. 3. The Curie temperature (T_c) dependence on Ce concentration for the $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys.

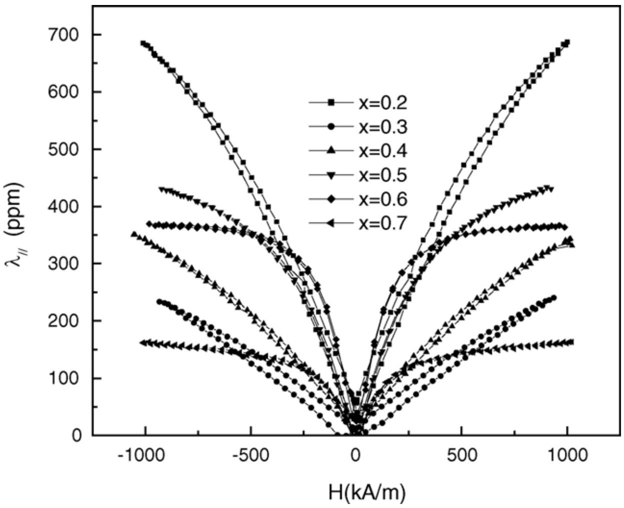

Fig. 4. Magnetostriction ($\lambda_{//}$) of the $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys at room temperature.

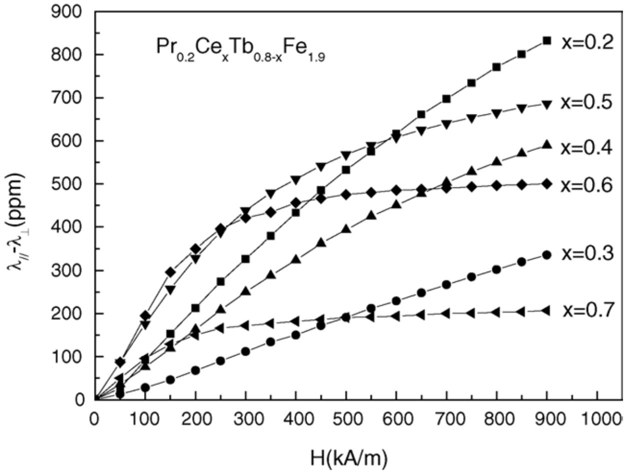

Fig. 5. Magnetostriction ($\lambda_{//}-\lambda_{\perp}$) of the $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys at room temperature.

increases with Ce concentration increasing from 0.3 to 0.5. The rapid decrease in magnetostriction is ascribed to the Ce substitution for Tb when $0.2 \leq x < 0.4$ and $x > 0.6$. The anomalous increase of magnetostriction when $0.4 \leq x \leq 0.6$ is ascribed to the valence fluctuation of Ce ions. According to the single-ion model, the trivalent Ce ions have larger magnetostriction than Tb and Dy ions, and in our previous study on $Ce_xTb_{1-x}Fe_2$ compounds, it was found that Ce ions contributed to the magnetostriction when the valence of Ce ions fluctuate toward trivalence. It has been known from the dependence of the lattice parameter on Ce concentration that the positive deviation from Vegard's law was observed, and exhibit a maximum value at $x = 0.5$, the anomalous increase when $0.4 \leq x \leq 0.6$ further confirms that Ce ions really contribute to the magnetostriction when the valence of Ce ions fluctuate toward trivalence.

4. Conclusion

The structural and magnetic properties of the $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ system were studied. It was found that all samples are pure cubic Laves phase of $MgCu_2$-type structure. The lattice parameter shows positive deviation from Vegard's law, which is resulted from the tendency of Ce toward trivalence. The Curie temperature decreases with increasing Ce concentration. The magnetostriction of the $Pr_{0.2}Ce_xTb_{0.8-x}Fe_2$ alloys decreases with increasing Ce concentration, when $0.2 \leq x < 0.4$ and $x > 0.6$, while increases when $0.4 \leq x \leq 0.6$. The anomalous increase of magnetostriciton between 0.3 and 0.5 can be attributed to the fluctuation of Ce toward trivalence.

Acknowledgements

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