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Structure and magnetostriction of $Ho_{1-x}Pr_xFe_{1.9}$ alloys

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ABSTRACT

Polycrystalline $Ho_{1-x}Pr_xFe_{1.9}$ ($0 \le x \le 1$) cubic Laves alloys were synthesized by arc-melting and subsequent annealing under high stress. Their structure, magnetic properties and magnetostriction are investigated using X-ray diffraction, vibrating sample magnetometer and standard strain gauge technique, respectively. It was found that $Ho_{1-x}Pr_xFe_{1.9}$ single cubic Laves phase cannot be obtained when $x > 0.2$ by a traditional vacuum annealing method. In contrast, the cubic Laves phase can be stabilized over the whole studies range in the samples annealed under high stress. The saturation magnetization for $Ho_{1-x}Pr_xFe_{1.9}$ decreases with the increase of x and reaches a minimum at $x=0.4$, then increases with further increase of x, which indicates the antiparallel magnetic moment alignment between Ho and Pr sublattice. The magnetostriction of $Ho_{1-x}Pr_xFe_{1.9}$ does not linearly increase with increasing x, but presents a minimum at $x=0.4$.

1. Introduction

RFe_2 (R=rare earths) C15 cubic Laves phase alloys were extensively investigated due to their giant magnetostriciton at room temperature. Terfenol-D ($Tb_{0.27}Dy_{0.73}Fe_2$), with low magnetic anisotropy and large low-field magnetostriction, has been widely applied in sonar transducer, sensors, actuators, etc. [1,2]. However, the raw materials of Terfenol-D are mainly expensive heavy rare earths Tb and Dy. According to the single-ion model, $PrFe_2$ possesses a larger calculated magnetostriction constant than $TbFe_2$ and $DyFe_2$ at 0 K due to its large second-order Stevens' factor α_J, ground state angular momentum J and average radius $\langle r_{4f}^2 \rangle$ of the 4f electron shell of the Pr^{3+} ion [1]. In addition, a magnetostrictive material with high-Pr content should have a good practical prospect because it is much cheaper than the heavy rare earths Tb or Dy. However, former studies showed that $PrFe_2$ cubic Laves phase could not be synthesized by conventional annealing at ambient pressure [3]. The unwanted non-cubic phase appears and the single phase materials cannot be synthesized in (R,Pr)Fe_2 alloys when Pr is over a certain concentration in rare earth sublattice [4–7]. Therefore, much attention has been paid to increase the concentration of Pr in (R,Pr)Fe_2 cubic Laves alloys. For example, Ren et al. recently reported the structure, magnetic and magnetostrictive properties in $Tb_{0.1}Ho_{0.9-x}Pr_xFe_{0.9}B_{0.1}$ compounds. They found that the introduction of a small amount of boron is beneficial to stabilize high-Pr content cubic Laves phase. But the single cubic Laves could not be obtained in $Tb_{0.1}Ho_{0.9-x}Pr_xFe_{0.9}B_{0.1}$ alloys when the Pr concentration exceeds 40 at% in rare-earth sublattice [6]. A similar result was also reported by Hari Babu et al. [7]. Up to present, $Ho_{1-x}Pr_xFe_2$ cubic Laves alloys have not yet been synthesized and their magnetic properties remain unknown. Recently, we reported that the structure and magnetic properties of $PrFe_x$($1.5 \le x \le 3$) alloys synthesized by annealing as-cast ingots under 6 GPa and the single cubic Laves phase was realized in $PrFe_{1.9}$ [8]. In this paper, polycrystalline alloys $Ho_{1-x}Pr_xFe_{1.9}$ ($0 \le x \le 1$) with cubic Laves phase have been successfully synthesized by the same method. The crystal structure, magnetic properties and the magnetostriction of the alloys are investigated.

2. Experiment

Ingots with $Ho_{1-x}Pr_xFe_{1.9}$ ($x=0.0$, 0.2, 0.4, 0.6, 0.8 and 1.0) stoichiometry were prepared by melting the appropriate constituent metals in a magneto-controlled arc furnace under a high-purity argon atmosphere. The as-cast ingots were pressed to 6 GPa by a hexahedral anvil press and heated to 900 °C for 30 min. Conventional X-ray diffraction (XRD) analysis was carried out using Cu Kα radiation with a Rigaku D/Max-gA diffractometer. The lattice parameters were calculated using the Jade 5.0 XRD analytical software (Materials Data, Inc., Livemore, CA). The magnetization measurements were carried out using a vibration sample magnetometer (7300, Lakeshore) under a magnetic field up to 10 kOe. The shape of sample for magnetostriction measurement was mainly disk-like with a diameter of 10 mm and a height

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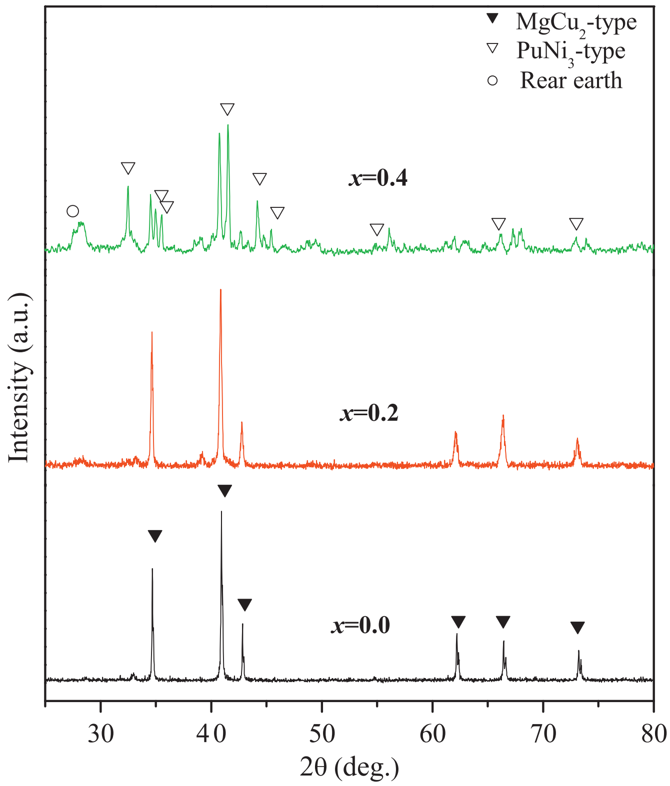

Fig. 1. XRD patterns of $Ho_{1-x}Pr_xFe_{1.9}$ alloys prepared by vacuum annealing at 900 °C for 30 min.

of 2 mm. The linear magnetostriction was measured using standard strain-gauge technique in directions parallel ($\lambda_{\parallel}$) and perpendicular ($\lambda_{\perp}$) to applied magnetic fields at room temperature.

3. Results and discussion

In order to study the structure of $Ho_{1-x}Pr_xFe_{1.9}$ alloys prepared by traditional high-temperature annealing method, the as-cast samples were vacuum annealed at 900 °C for 30 min. The XRD patterns for the samples with $x=0.0$, 0.2 and 0.4 are shown in Fig. 1. It can be observed that the single cubic Laves phase with $MgCu_2$-type structure can only be stabilized with $x\leq 0.2$. A multiphase structure appears when the Pr concentration further increases. For the sample with $x=0.4$, $(Ho,Pr)Fe_3$ with $PuNi_3$-type structure becomes the main phase and $(Ho,Pr)Fe_2$ with $MgCu_2$-type is the secondary phase, coexisting with a small amount rare earth phases. This is consistent with the structure of $Tb_{0.1}Ho_{0.9-x}Pr_xFe_{0.9}B_{0.1}$ alloys, in which the single cubic Laves phase cannot be stabilized when the Pr concentration is higher than 40 at% in rare-earth sublattice even with the help of Tb and B [6,7].

In contrast with Fig. 1, the samples prepared by annealing under high stress (Fig. 2) exhibit almost single cubic Laves phase with $MgCu_2$-type structure, coexisting with minor impurities, i.e. rare earth phases. Generally, the atomic size plays an important role in the formation of RFe_2 cubic Laves alloys. It was estimated that the ideal radius ratio between R and Fe for a cubic Laves phase is 1.225 [9]. As a result, it is difficult to obtain high-Pr content cubic Laves phases under normal pressure due to the large radius ratio between Pr and Fe. By the method of annealing at ambient pressure, the element of Ho with its smaller radius can stabilize $Ho_{1-x}Pr_xFe_{1.9}$ cubic Laves phase with Pr up to 20 at% in rare-earth sublattice, but cannot hold when the Pr concentration goes much higher. Therefore, the obtained stabilization of cubic

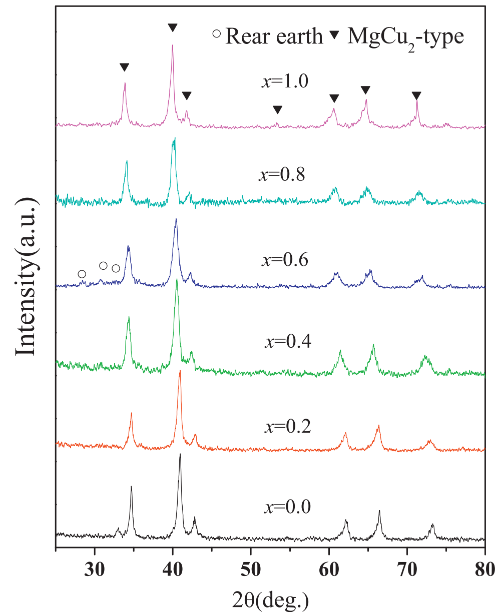

Fig. 2. XRD patterns of $Ho_{1-x}Pr_xFe_{1.9}$ alloys prepared by annealing under 6 GPa at 900 °C for 30 min.

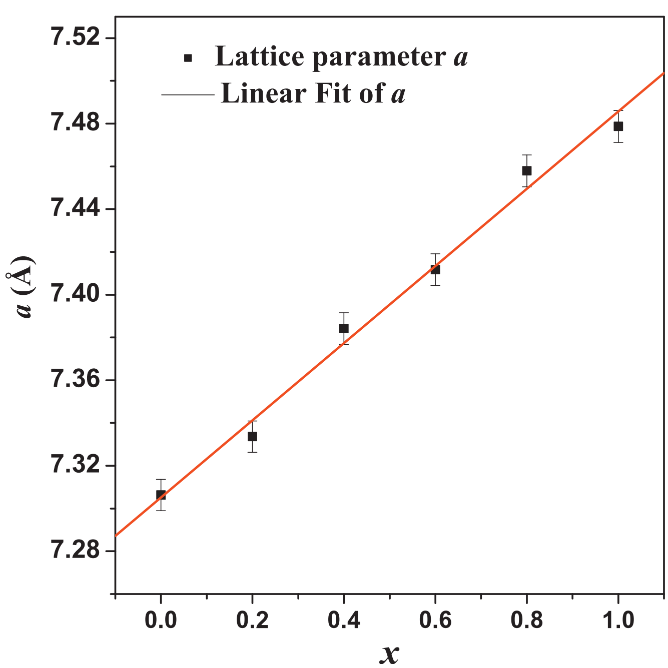

Fig. 3. The lattice parameter a of $Ho_{1-x}Pr_xFe_{1.9}$ Laves phase synthesized by annealing under 6 GPa at 900 °C for 30 min.

Laves phase with high-Pr content in $Ho_{1-x}Pr_xFe_{1.9}$ should be attributed to the effects of high stress.

The lattice parameter (a) of $Ho_{1-x}Pr_xFe_{1.9}$ Laves phase synthesized under high stress is shown in Fig. 3. An approximate linear increase with Pr-concentration from 0.0 to 1.0 is found, as expected from Vegard's law: $a=xa_1+(1-x)a_2$, where a_1 and a_2 are the lattice parameters of $PrFe_{1.9}$ and $HoFe_{1.9}$, respectively. The increase of lattice parameter with increasing Pr concentration is due to the larger ionic radius Pr^{3+} than that of Ho^{3+}.

Magnetic field dependence of magnetization at 300 K for the $Ho_{1-x}Pr_xFe_{1.9}$ is shown in Fig. 4(a). We can note that each sample shows a tendency to saturation at the field of 10 kOe. For clarity, the magnetization at the maximum available 10 kOe, denoted as