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Materials Science and Engineering A 419 (2006) 39–44

MATERIALS SCIENCE & ENGINEERING A

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Study on the melt-textured technique in a magnetic field for giant magnetostrictive materials R–Fe alloy

Peiran Deng, Jianguo Li *, Zhenming Xu

School of Materials Science and Engineering, Shanghai Jiao Tong University, 1954 Huashan Road, Shanghai 200030, PR China

Received in revised form 14 November 2005; accepted 29 November 2005

Abstract

The melt-textured technique in a magnetic field for giant magnetostrictive materials $TbFe_x$ and $Tb_{0.3}Dy_{0.7}Fe_x$ ($1.9 \leqslant x \leqslant 2$) alloy was investigated. The cooling rate, melt temperature, and the chemical composition were found to influence the texture formation along the easy magnetic axis during crystallization in a field. The experiments showed that the ⟨1 1 1⟩ orientation of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ and $TbFe_{1.9}$ alloy was dominant in the samples when those alloys were solidified at a cooling rate of less than 0.8 °C/min from a temperature slightly above their melting point, respectively, while in a magnetic field of above 100 mT. However, with the Fe concentration increasing from $x = 1.93$, excessive RFe_3 phases precipitate in the melt and degrade the orientation degree of the sample during the slow solidification. Therefore the chemical composition of $Tb_{0.3}Dy_{0.7}Fe_x$ and $TbFe_x$ alloy should be controlled within $1.90 \leqslant x \leqslant 1.93$ for better orientation and comprehensive performance.

Keywords: Giant magnetostrictive materials; Magnetocrystalline anisotropy; Magnetic field; Slow cooling; Chemical composition

1. Introduction

The rare-earth giant magnetostrictive materials, $TbFe_2$ and $Tb_{0.3}Dy_{0.7}Fe_x$ ($x = 1.9$–2.0), are of significant technological interest [1–6] for their excellent magnetostrictive properties at room temperature. In particular, the alloy composition with $Tb_{0.3}Dy_{0.7}Fe_x$ ($x = 1.9$–2.0), known as Terfenol-D, possesses low magnetocrystalline anisotropy energy, and can exhibit high sensitivity to an external magnetic field as well as large magnetostrictive strains in a considerably lower magnetic fields [3]. Because of the strong anisotropy of the magnetostrictive strain, $\lambda_{111} \gg \lambda_{100}$ [7], the preparation of defect-free crystals with ⟨1 1 1⟩ orientation is desirable for those compounds. The directional solidification methods, such as the Czochralski method, the modified Bridgman method, the vertical float zoning method and the zone melting liquid metal cooling method, have been used to manufacture rods of Terfenol-D alloy [8–11]. However, the ⟨1 1 0⟩ and ⟨1 1 2⟩ orientation were obtained by using such directional solidification methods. The ⟨1 1 1⟩-oriented crystal can only be achieved by seeding technique [12,13], but application of the technique requires a high quality seed crystal. The process has proved to be too complex to suit commercial production.

For $TbFe_x$ and $Tb_{0.3}Dy_{0.7}Fe_x$ ($x = 1.9$–2.0) alloy, the ⟨1 1 1⟩ direction with the greatest magnetostrictive constants for $TbFe_y$ and $Tb_xDy_{1-x}Fe_y$ ($x = 0.27$–0.3, $y = 1.9$–2.0) alloy is also their crystallographic orientation of easy-magnetization axis. So during the process of preparing Terfenol-D alloy sintered compacts, a static magnetic field can be used to induce pre-alignment of the powders [14]. The pre-alignment of the powders by powder metallurgy technique is performed at temperatures below the material's Curie temperature. Mei et al. [14] have investigated those works. The intensity of (1 1 1) peak in the final products was slightly stronger, and many other peaks can be observed in the X-ray pattern and some pores defects existing in matrix were inevitable.

The recent investigations indicate that due to the persistence of residual magnetocrystalline anisotropy at high temperature far above their Curie point, texturing of magnetic materials along their easy magnetic axes can be achieved directly by solidification in a magnetic field [15,16]. The value of the anisotropy of the paramagnetic susceptibility $\Delta\chi = 3 \times 10^{-8}$ m^3/kg for Sm_2Co_{17} was measured at temperature of 1175 °C [16]. Textured bulk samples of the high temperature superconductor $YBa_2Cu_3O_7$ had been produced successfully by slow cooling in a magnetic field of 5 T [15]. Legrand et al. prepared SmCo compounds by

* Corresponding author. Tel.: +86 21 6293 3074; fax: +86 21 6293 2026.
E-mail address: lijg@sjtu.edu.cn (J. Li).

0921-5093/$ – see front matter © 2006 Published by Elsevier B.V.
doi:10.1016/j.msea.2005.11.058

solidification in the copper crucible in a magnetic field of 2.5 T, solidification occurred within only a few seconds and a high degree of orientation in sample was also obtained though in unfavorable conditions of solidification [16].

At present, Minggawa et al. have produced samples with ⟨1 1 1⟩ crystallographic alignment of the crystal grains for $TbFe_2$ and $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ alloy by a magnetic field 4.5×10^{-2} and 3.7×10^{-2} T respectively in microgravity [17,18], it provided good orientation, but cracks easily formed in samples during rapid cooling. The convection and heat disturbance in melt were totally suppressed in microgravity. When the solidification experiments were performed in normal gravity by the same field and cooling rate, no texture formed in samples. The process in microgravity also lacks industrial value presently.

It is essential for determining the orientation formation to suppress the turbulence in melt during solidification and crystallization. In normal gravity, a strong magnetic field of several tesla was usually used to suppress the thermal convection, mass transfer, and temperature fluctuation. In research on Y–Ba–Cu–O superconductor, it has been shown that the turbulence in melt also can be reduced by slowing down the solidification rate [19]. Such results suggest that the heat disturbance should be further controlled during slow cooling by the use of a relatively weak magnetic field. Utech and Flemings [20] and Chedzey and Hurle [21] showed that fluctuation of temperature during zone melting and Bridgeman crystal growth can be suppressed mainly by application of a weak magnetic field of several hundreds mT, which are also enough to satisfy with industrial production to manufacture semiconductor nowadays. In this article, the experiments relating to the solidification of $TbFe_2$ and Terfenol-D alloy in a magnetic field are reported.

2. Experimental procedure

The melting and solidification experiments were performed in vacuum container of the super high temperature gradient directional solidification installation. Master alloys of $TbFe_x$ ($x = 1.86$–1.88, and 1.96–1.98) and $Tb_{0.3}Dy_{0.7}Fe_x$ ($x = 1.85$–1.87, and 1.96–1.98) with size of Ø 16 × 20 mm were firstly prepared from high quality start materials with 99.9 wt% purity by induction melting under purified argon at a pressure of 0.08 MPa. The alloy compositions were analyzed by using an inductively coupled plasma (ICP) method.

A magnetic field apparatus made by us was introduced into the container of the solidification installation. Ingot bar of master alloy was contained in an Al_2O_3 crucible of 16 mm i.d., then fixed in the center of magnetic poles and the axis of cylinder sample was parallel to the direction of magnetic field and gravity. The following re-melting and solidification experiments were also carried out in argon atmosphere at a pressure of 0.08 MPa after the container was evacuated below 5×10^{-2} Pa. Those samples were preheated rapidly to about 1050 °C for a short time, and then heated slowly to the temperature slightly above their melting point for $TbFe_x$ ($x = 1.86$–1.88, and 1.96–1.98) and $Tb_{0.3}Dy_{0.7}Fe_x$ ($x = 1.85$–1.87, and 1.96–1.98) alloy respectively. After the samples were melt fully, they were kept for 10 min in a static magnetic field of 100–140 mT in order to even the temperature field in the melt. Then they were solidified at a different cooling rate. Solidification in a magnetic field from the temperature below the melting point was also studied.

The specimens from the middle section of samples were examined, X-ray diffraction with Cu Kα radiation was used to determine the crystal orientation of those specimens along the external magnetic field, and the scanning electron microscopy and the optic microscope were used to observe the microstructure and macrostructure in the specimens, respectively. The specimens were etched by the nital solution (HNO_3, 5.0 vol%/C_2H_5OH).

3. Results

3.1. The effects of cooling rate on orientation

The master alloy of $Tb_{0.3}Dy_{0.7}Fe_{1.86}$ was heated to the temperature of 1260 °C. In the presence of the magnetic field 140 mT, the melt solidified at four cooling rates, namely, a rapid cooling rate, 3.5, 1.5, 0.8 °C/min in the field. The rapid cooling rate is about 0.7 °C/s.

Fig. 1 shows that slower cooling rates yields a higher degree of ⟨1 1 1⟩ growth. Fig. 1a demonstrates the randomly oriented state for crystal grains while solidification occurred at a rapid

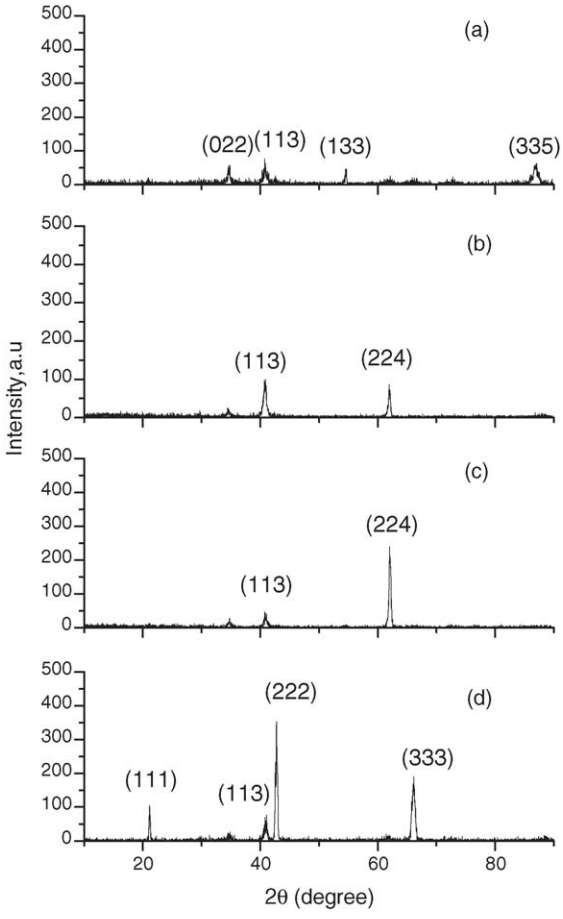

Fig. 1. X-ray diffraction patterns of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ from the samples solidified in a magnetic field of 140 mT at difference cooling rate (a: 7 °C/s, b: 3.5 °C/min, c: 1.5 °C/min and d: 0.8 °C/min).