Magnetostriction and magnetomechanical properties of grain-oriented \( \text{Tb}_{0.33}\text{Dy}_{0.67}\text{Fe}_y \)/Epoxy composite

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ABSTRACT The magnetostriction and dynamic magnetomechanical properties of the directionally solidified \( \text{Tb}_{0.33}\text{Dy}_{0.67}\text{Fe}_y \)/Epoxy composite were investigated as a function of volume fraction of RFe\(_2\) under a magnetic field. This composite was fabricated by infiltrating epoxy after extracting a eutectic phase of the directionally solidified \( \text{Tb}_{0.33}\text{Dy}_{0.67}\text{Fe}_y \). It was found that the magnetostriction of the composite is higher than that of the as-grown specimens, and the maximum magnetostrictive strain of the sample, which has a volume fraction of 0.72, showed about 1000 ppm at \( H = 80 \text{ kA/m} \) and an optimum preload of 8 MPa. Furthermore, the dynamic magnetomechanical properties of the composite \( \text{Tb}_{0.33}\text{Dy}_{0.67}\text{Fe}_y \), which is the sensitivity of the strain to the applied field, increased by about two times that of as-grown samples.

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1 Introduction

Giant magnetostrictive composites have attracted a great deal of attention by supplementing shortcomings of monolithic Terfenol-D such as brittleness, eddy current loss and formability. Since the 1970’s, a powder metallurgy technique using Terfenol-D particulates has been explored for a magnetostrictive composites [1]. \( \text{Tb}_x\text{Dy}_{1-x}\text{Fe}_9 \) (where \( 0.27 \leq x \leq 0.30 \)) and \( 1.90 \leq y \leq 2.0 \) [2–4] particles, using particulate composite for enhancement of the energy efficiency, are produced by using a ball mill or other processes. In general, since the magnetocrystalline anisotropy in monolithic Terfenol-D is controlled by the Tb/Dy ratio, it is desirable to maintain the \( x \)-value above 0.27 in order for the magnetic moment to orient preferably along \( \langle 111 \rangle \) directions [5]. Verhoeven et al. [6] have demonstrated that the reduction in \( x \) from 0.316 to 0.285 has produced a significant drop in magnetostriction, and this result is due to the microsegregation of the Tb in the dendritic region of the directionally solidified crystal; the initial solidified regions of the Laves phase have lower content Dy and the final one has a higher one of the Tb.

According to the anisotropic rotation model [7], a magnetic field applied along the \( [112] \) axis induces a jump of magnetic moments from the \( [1 1 1], [1 1 1] \) directions to the \( [1 1 1] \) direction. This moment reorientation causes the “burst effect”, which shows a rapid magnetostrictive strain. According to Galloway et al. [8], the magnetostrictive burst region was seen in \( \text{Tb}_{0.316}\text{Dy}_{0.684}\text{Fe}_{1.982} \). Thus, the initial Tb/Dy ratio is desirable to be established at 0.30/0.70 in case of the particulate composite.

Recently, we introduced a new process [9] to fabricate a grain-aligned composite called PIGC (polymer infiltrated grain-aligned composite). This process summarized: the first step is to grow a crystal with a low value of Fe in order to form the eutectic phase having a low melting point in between the Laves phase. The second step is to extract the eutectic phase composed of the rare earth RE and RFe\(_2\) from the contact area between quartz ampoule and as-grown specimen through high temperature annealing, 1000 °C. This extraction is caused by the reaction between the quartz ampoule (SiO\(_2\)) and the liquid melt above eutectic point, 847 °C–896 °C. The final step is to infiltrate the liquid epoxy into the porous precursor using pressure difference between them. In this process, however, the eutectic phase is extracted in the second step in which both phases (RFe\(_2\) and liquid) are in equilibrium at 1000 °C, and the RFe\(_2\) is partly melted at the interface of RFe\(_2\)/liquid, and hence the Tb/Dy ratio becomes lower than that of as-grown specimen. Thus, it is preferred to select an alloy composition with higher Tb/Dy ratio (\( \geq 0.30 \)) than that of Terfenol-D used in particulate composite, accordingly we selected the ratio to 0.33/0.67.

The purpose of the present work is to identify the effect of \( \text{V}_{\text{RFe}_2} \) (the volume fraction of the Laves phase) on magnetostriction and dynamic magnetomechanical properties of PIGC with the Tb/Dy ratio of 0.33/0.67.

2 Manufacture

The \( \text{Tb}_{0.33}\text{Dy}_{0.67}\text{Fe}_y \) alloys, with the \( y \)-values of 0.71, 0.90, 1.05, 1.36, and 1.65 composed of Tb, Dy and Fe of high purity (99.99%) were prepared by arc-melting under high purity argon and then were cast into rods through a suction method after induction melting under argon atmosphere. A well-aligned RFe\(_2\) phase was achieved by zone melting of 100 mm rod at a growth rate of 70 \( \mu \text{m/s} \), followed by sealing a cast rod into a quartz ampoule with an inner diameter of 9 mm. The grown samples were machined into cylindrical
shape of 6 mm diameter and 20 mm length. For PIGC, cylindrical samples were put into the quartz ampoule and sealed at 730 Torr with purified argon and then were heated at an annealing temperature of 1000°C. After annealing, the leaked phase attached to the sample was removed and the sample was immersed into liquid epoxy of 60°C and maintained for 30 minutes at 1 Torr. After curing the composite at 80°C for 3 h, its magnetostrictive strain was measured along longitudinal direction with LVDT at room temperature, up to 80 kA/m under compressive pre-stress in the range from 2 to 10 MPa. The magnetization was measured using a flux meter with a pick-up coil. The demagnetization was carried out prior to every measurement.

3 Results and discussion

The microstructure in the Tb0.33Dy0.67Fe3 alloy system is very sensitive to iron content. As shown in Fig. 1a, the as-grown microstructure reveals dendrite morphology of the RFe2 phase with the eutectic phase between the dendrites. The eutectic phase is extracted from the sample due to the reaction with oxygen constituent of SiO2 and the liquid phase at annealing temperatures. After the annealing, the leaked channel was replaced with the epoxy using polymer infiltration method as shown in Fig. 1b. Figure 1c shows the change for VRF2 in as-grown alloys and polymeric composites. The VRF2 of the polymeric composites obtained by polymer infiltration method is lower than that of as-grown alloys shown in Fig. 1c and Table 1. This result indicates that at annealing temperature of 1000°C, not only is the eutectic phase extracted but also the primary RFe2 phase is partly melted out to liquid phase.

Figure 2 shows the dependence of the magnetostrictive strains for several VRF2 of the composites as a function of bias field. As VRF2 is increased from 0.34 to 0.72, magnetostrictive strain of the composite is significantly increased as compared with as-grown samples. The composite with 0.72 volume fraction specimen yields the highest magnetostrictive response of 1000 ppm. The increased ratio of magnetostrictive strain in 0.34, 0.39, 0.43, and 0.52 VRF2 as compared with as-grown specimens shows about 90.3%, 201.5%, 125.1%, and 60.6%, respectively. Since the magnetostriction in polycrystalline grown by float zone melting was increased by 16% ~ 35% through heat treatment as suggested by Verhoeven et al. [11], this enlargement in strain is too large to be due to only the annealing effect that reduces the pinning of domain walls or enhances the rotation of domain moments through residual stress relief of as-grown crystal. Therefore, at the same y-value, the higher magnetostriction in the composite is due to the absence of the eutectic phase. In addition, as the volume fraction of the eutectic phase is increased, the magnetostriction decreases (see Table 1). This result suggests that the eutectic phase inhibits the large magnetization rotations required for giant magnetostrictive strain and accordingly RFe2 phase in the eutectic phase does not affect noticeably the magnetic properties of the primary RFe2 phase as suggested by Clark et al. [12].

Figure 3 shows the variations of the Tb and Dy content in the center and the edge of the Laves phase analyzed by the X-ray wavelength dispersive analysis. Since the initially solidified RFe2 phase will be richer in Dy and the final one will be richer in Tb, the Tb/Dy ratio within the RFe2 phase is definitely lower than the initial ratio [13]. Especially, the Tb/Dy ratio depends on the y-value in the Tb0.33Dy0.67Fe3, and affects the magnetocrystalline anisotropy, which is minimized at the critical ratio of 0.27/0.73 at which λ111 drops to near zero.

Since VRF2 in PIGC is much lower than that of the eutectic phase in as-grown specimens, the external interface of RFe2 dendrite with a high Tb ratio is partly melt to liquid phase.