Improved magnetostriction and mechanical properties in dual-phase FeGa single crystal

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ABSTRACT
Single-phase Fe$_{81}$Ga$_{19}$ and dual-phase (Fe$_{0.81}$Ga$_{0.19}$)$_{99.95}$Tb$_{0.05}$ single crystals with perfect ⟨001⟩ orientation were prepared by directional solidification technology. The performance of the dual-phase single crystal (SC) can achieve 399 ppm in magnetostriction and 10.2% in tensile fracture strain, which are, respectively, 28% and 5 times larger than those in single-phase Fe$_{81}$Ga$_{19}$ SC. The slight solid solution of Tb and dispersive distributed Tb-rich particles in dual-phase SC, respectively, lead to the improvement in magnetostriction and ductility. This dual-phase SC can become the candidate for new-generation magnetostrictive materials combining significant advantages in both structural and functional properties.

IMPACT STATEMENT
This dual-phase (Fe$_{0.81}$Ga$_{0.19}$)$_{99.95}$Tb$_{0.05}$ single crystal combining large magnetostriction and excellent mechanical properties can be new-generation magnetostrictive materials which can satisfy the application requirement in structural and functional properties.

1. Introduction
Magnetostrictive materials have attracted numerous attention in the last decades for the indispensable role in energy harvesting, high-precision displacement sensor and transducers [1,2]. Presently, the Terfenol-D (Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.95}$) alloys are the dominant commercial magnetostrictive material due to the superior room-temperature magnetostriction [3,4]. However, the mechanical brittleness limits the machinability of them, and the high cost of rare earth (RE) severely restricts widespread applications [1,5]. Therefore, new-generation FeGa magnetostrictive alloys are developed due to the unique characteristics [6,7]. Compared with Terfenol-D materials, FeGa alloys possess decent magnetostriction, low saturation magnetic field and fair ductility [8–11]. These advantages make FeGa alloys a perfect candidate in numerous application fields. Even so, further improvements in both magnetostriction and ductility are still in strong requirement, and numerous attempts through doping the third elements or the carbides have been employed. Researchers have studied the effect of doping some main group element atoms (B [12], C [13,14], Al [15], In [16], Ge [16], Si [17], Sn [17]), transition elements (Cr [13,18], Mn [14], V [13], Nb [14,19], Mo [14]) and carbides (NbC [20,21], TaC [22]) on the performance of FeGa-based magnetostrictive alloys. However,
for most of them mentioned above, the dopant of them will degrade the magnetostriction or ductility. Only B and carbides are confirmed to improve the two properties simultaneously. The slight addition of B or carbides (less than 1%) can improve the room-temperature tensile fracture strain to \( \sim 4\% \) and 1%, respectively [12,20–23]. This allows the possibility for further processing such as rolling for FeGa-based magnetostrictive materials. However, the improvement in magnetostriction of the alloys is quite limited, the largest magnetostriction of the doped alloys is only a bit larger than that of the FeGa binary SC. Recently, it is observed that the doping of RE elements possesses the potential to improve the structural and functional properties simultaneously [24–28]. The trace solid solution of RE (<0.2 at.%) can result in a significant improvement in magnetostriction [25–27]. Dual-phase microstructure containing the matrix and the dispersively distributed RE-rich precipitates is able to transform the fracture mechanism from brittle to ductile, and this significantly enhances the ductility [28–30]. The investigations suggest that doping tiny RE elements to form a dual-phase structure may be able to realize large magnetostriction and excellent ductility simultaneously.

The magnetostriction generally exhibits a feature of anisotropy, which is largest along (001) direction in FeGa alloys. The alloys generally require to be initially processed to rods for applications [31,32]. The (001) oriented FeGa crystals containing grain boundaries parallel to the growth direction are easily to crack along the grain boundaries during rolling process because of the formed stress perpendicular to the boundaries, and the SC can effectively avoid the problem because of the absence of grain boundaries. Therefore, (001) preferred orientated SC can give full play to the properties and are more favored for applications [33,34]. The comparison of un-doped and RE-doped FeGa (001) SCs can definitely prove the effect of RE on magnetostriction and ductility. In this work, single-phase \( \text{Fe}_{81}\text{Ga}_{19} \) and dual-phase \( \left(\text{Fe}_{0.81}\text{Ga}_{0.19}\right)_{99.95}\text{Tb}_{0.05} \) (named as Tb0 and Tb0.05) (001) SCs are prepared by Bridgman directional solidification technology. Although the particulate precipitates are formed in Tb-doped alloys, still no grain boundaries appear in the alloy, so it is still named as SC which is similar to some high-entropy alloys [35]. The performance of the dual-phase SC can achieve 399 ppm in magnetostriction and 10.2% in tensile fracture strain, which are, respectively, 28% and 5 times larger than those of single-phase \( \text{Fe}_{81}\text{Ga}_{19} \) SC. The slight solid solution of Tb leads to a larger magnetocrystalline anisotropy which contributes to the magnetostriction. The dispersively distributed Tb-rich particles in dual-phase SC can transform the fracture path and result in an ultrahigh tensile plastic deformation. This SC can become the candidate for new-generation magnetostrictive materials combining the excellent properties in both structural and functional properties.

2. Experiment

The ingots with the nominal chemical compositions of \( \text{Fe}_{81}\text{Ga}_{19} \) and \( \left(\text{Fe}_{0.81}\text{Ga}_{0.19}\right)_{99.95}\text{Tb}_{0.05} \) were prepared in a vacuum arc furnace with high-purity iron, gallium and terbium. The ingots were cast into a copper mold to manufacture the rods with the diameter of 7 mm. The rods were put into an alundum tube with a \( \text{Fe}_{81}\text{Ga}_{19} \) (001) SC seed at the bottom of the master rods. The SCs were grown in a vertical Bridgman directional solidification furnace with the withdrawal rate of 800 μm/s. The microstructures were observed by a LEICA DM4000 optical microscopy and JEOL JXA-8100 electron probe microanalysis (EPMA). The SC structures and orientation were confirmed by the X-ray diffraction (XRD) with Cu K\( _\alpha \) radiation, X-ray Laue back-reflection and pole figures. The magnetostriction was measured by the standard strain gauge method along the growth direction. The magnetization behaviors were measured on the physical properties measurement system (PPMS) at 300 K. The tensile measurement was carried out in an Instron 8801 material testing system, and the fracture surface was observed by a JOEL JSM-6010LA scanning electron microscope (SEM).

3. Result and discussion

Figure 1(a,b), respectively, shows the optical morphologies of Tb0 and Tb0.05 specimens. No evident grain boundaries as well as other phases can be observed in the cross-section surfaces, as proved by the homogeneous color in optical images. This indicates the SC microstructure for both rods. In order to detect the phase compositions, the back-scattered electron (BSE) images are carried out for each SC as exhibited in Figure 1(c,d). Single-phase morphology can be monitored throughout the binary Tb0 SC sample. For the Tb0.05 SC, additional particulate Tb-rich precipitates dispersively distribute in the matrix, as marked by the arrows. The chemical compositions are analyzed by energy-disperse spectroscopy (EDS) equipped in EPMA. The atomic percentages in the matrix and the precipitates are detected to be Fe81.1at.-%–Ga18.9at.-% and Fe50.2at.-%–Ga34.4at.-%–Tb15.4at.-%, respectively. Since the sensitivity of EDS is over 0.1 at.%, so Tb element is unable to be detected by EDS in the matrix. The compositional line scanning is performed to exhibit the variation of chemical compositions, a line with the length of 10 μm which crosses a particle precipitate is selected,
as shown in the inset of Figure 1(e), the evolution of atomic percentage for Fe, Ga and Tb atoms are shown by the color curves in Figure 1(e). The line scanning results are in accordance with the EDS point analysis, and this further confirms that the compositions of the matrix almost keep at the atomic ratio of Fe81Ga19, and a vast of Tb atoms form the particulate precipitates. In summary, single-phase and dual-phase SCs are prepared by directional solidification, the particulate precipitates with relatively excess Tb dispersively distribute in the matrix.

The crystal structure and orientation are identified by XRD patterns and Laue back-reflection pattern, as shown in Figure 2(a). Both Tb0 and Tb0.05 SCs possess a perfect (001) orientation since only very weak (110) peak can be observed in the XRD patterns for each SC. Laue back-reflection patterns in the right inset of Figure 2(a), and the pole figures of each SC in Figure 2(b1) and (b2), both support the (001) orientation SC and suggest that only tiny deviation from the (001) direction (less than 2°) is confirmed for both SCs. Detailed information of the microstructures can be extracted after local enlargement of the main (002) diffraction peaks, as shown in the left inset in Figure 2(a). The locations of the (002) peak for Tb0 and Tb0.05 SCs are at 2θ = 63.74° and 63.64°, respectively. Based on this, the lattice parameter \( a \) of the A2 matrix is calculated to be 
\[
a_0 = 0.2920 \text{ nm for Tb0 SC} \\
a_{0.05} = 0.2924 \text{ nm for Tb0.05 SC}
\]

The full width at half maximum (FWHM) is determined through Gauss fitting method. The FWHM increases from 0.5011° to 0.5295° with Tb doping. Since grain size factor is neglected, the broadening effect of diffraction peak proves larger lattice distortion. The local tetragonal distortion may form larger magnetocrystalline anisotropy and this can contribute to a better magnetostrictive performance.
Figure 3. (a,b) Room-temperature magnetostrictive curves for (a) Tb0 SC and (b) Tb0.05 SC measured under the compressive stress of 0, 30, 60 and 120 MPa, respectively. (c) M–H curves of Tb0 and Tb0.05 SCs measured at 300 K, the inset in (c) is the calculated $M_s$ and $K_1$ for both Tb0 and Tb0.05 SCs.

Figure 3(a,b) shows the measured magnetostriction under the compressive stress ($\sigma$) levels of 0, 30, 60 and 120 MPa, respectively. Both SCs exhibit significant stress effect, the magnetostriction increases from $\sim 100$ ppm when $\sigma = 0$ MPa to over 300 ppm when $\sigma = 30$ MPa. The magnetostriction is saturated when $\sigma \geq 30$ MPa for each SC, and the saturation magnetic field gradually raises with the increasing compressive stress level. The saturation magnetostriction of Tb0 SC is 311 ppm, which well matches the previous work [11,34]. The saturation magnetostriction of Tb0.05 SC achieves 399 ppm, which is almost 28% larger than that of Tb0 SC. Moreover, the saturation magnetic field (selected from the curve of $\sigma = 30$ MPa) almost remains unchanged at approximately 400 Oe. The significant improvement in magnetostriction is realized without the requirement of the higher magnetic field. $M$–$H$ curves for each SC are measured along the $\langle 001 \rangle$ direction at 300 K, as shown in Figure 3(c). The saturation magnetizations ($M_s$) are obtained using the law of approach to saturation, and the magnetocrystalline anisotropy constant ($K_1$) are calculated, $M_s$ and $K_1$ for both SCs are summarized in the inset in Figure 3(c). Although $M_s$ is slightly weakened, $K_1$ is faintly increased by traces of Tb addition. This is in consistent with the left motion and broadening of XRD peak for Tb0.05 SC (see Figure 2). The slight solid solution of Tb could lead to larger local tetragonal distortion in the matrix and result in larger $K_1$ [27,36], this is considered to contribute to magnetostriction in Tb-doped dual-phase SC.

Figure 4(a) shows the tensile stress–strain curves of Tb0 and Tb0.05 SCs at room temperature. The Young’s modulus ($E$) of both SCs is calculated to be $33.24 \pm 0.25$ GPa according to the elastic section, and this value is in well accordance with the theoretical value simulated by Wang et al. [37]. For the binary SC, the tensile curve exhibits a feature of brittle fracture and the tensile fracture strain is 1.8%, which is over five times larger than that of binary polycrystalline [13,28,29]. However, for the dual-phase SC, the shape of tensile curves is completely different, ultra-long plasticity section is obtained and the fracture strain achieves 10.2%, a further five times larger than that of the binary SC. Moreover, the tensile specimen of dual-phase specimen is significantly longer than that of the Tb0 SC specimen after tensile test, as shown in the inset of Figure 4(a). Compared with the as-cast alloy and oriented rod, the fracture strain of the SC
is significantly further improved [29,30]. One reason is the softening of the Young’s modulus because of the perfect (001) orientation. H. Wang et al. confirmed that the Young’s modulus is \(\sim 30\) GPa along (001) direction and \(\sim 70\) GPa along (110) direction, respectively. The softening can significantly benefit the ductility of the alloys [38]. Besides, the change in fracture behavior is also closely related to the superb ductility. The fracture surface morphologies for both SCs are observed by SEM for comparison, as shown in Figure 4(b,c). For Tb0 SC, homogeneous and parallel lines can be observed under low magnification, as shown in Figure 4(b1), only a small quantity of lines with a different direction can be seen in the single-phase SC, as marked by red ellipse in Figure 4(c). The direction of slip bands is more complex, and obvious step-like morphologies are observed, as, respectively, marked by dash lines and yellow ellipses in Figure 4(c1). Since the Tb-rich particles should play a key role in the evolution of fracture characteristics, a particle is observed as marked by the red arrow is observed in the enlarged figure as shown in Figure 4(c2). Different slip directions marked by yellow dash lines and obvious step morphologies marked by yellow circles are confirmed to be around the particle.

In the previous study, we have discovered that the Tb-rich particle is a faced crystal and generally possesses corner angles [29]. This easily produces the stress concentration during tensile deformation, so different slip systems can be activated in the stress concentration region. Since the density of particle is quite low, the connection of slip dislocations to take place the fracture behavior is more difficult. Thus, the special effect of Tb-rich particles results in further accumulation of dislocations and give rise to larger tensile fracture strain, significantly better ductility can be achieved in this dual-phase SC which contains dispersively distributed Tb-rich particles.

4. Conclusion

In summary, single-phase Fe\textsubscript{81}Ga\textsubscript{19} and dual-phase (Fe\textsubscript{0.81}Ga\textsubscript{0.19})\textsubscript{99.95}Tb\textsubscript{0.05} single crystals with excellent (001) orientation are prepared by directional solidification technology. The dual-phase SC possesses the performance of 399 ppm in magnetostriction and 10.2% in tensile fracture strain. The slight solid solution of Tb leads to a larger magnetocrystalline anisotropy and this contributes to the improvement in magnetostriction. The dispersive distributed Tb-rich particles can transfer the fracture path and results in an ultra-large tensile plastic deformation. This dual-phase SC can become the candidate for new-generation magnetostrictive materials combining the advantages in both structural and functional properties.

Acknowledgements

The authors thank Professor Sujun Wu of Beihang University for the meaningful discussions.

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding

This work was supported by National Natural Science Foundations of China (NSFC) [grant number 51331001], [grant number 51601007], [grant number 51674011]; China Postdoctoral Science Foundation Funded Project [grant number 2017M610738].

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