

Microstructure of peritectic Tb-Dy-Fe alloys after electromagnetic levitation melting^①

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Abstract: The electromagnetic levitation melting of the rare earth giant magnetostrictive materials Tb_{0.27}Dy_{0.73}Fe_{1.90} alloys is realized. The compound is difficult to realize levitation melting at terrestrial environment because of its high density and low electric conductivity. The microstructure of the sample near peritectic composition obtained by this process contains REFe₂ matrix phase, a large amount of rod-like REFe₃ phase embedded in the matrix phase, rare earth rich phase enriching around the REFe₃ phase and a small amount of rare earth rich phase in the REFe₂ matrix phase far from the REFe₃ phase, which is significantly different from that of the sample obtained by general melting-casting process with the same composition, which contains almost complete REFe₂ phase and a small amount of RE rich phase in the REFe₂ matrix phase. The formation of the microstructure can be attributed to the coupled growth of the peritectic phase—REFe₂ and the primary phase—REFe₃, and the subsequent eutectic reaction under this experimental condition.

Key words: electromagnetic levitation melting; Tb-Dy-Fe alloys; peritectic; coupled growth

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1 INTRODUCTION

Tb-Dy-Fe alloy, as a rare earth (RE) magnetic functional material, is of significant technological interests, because of its giant magnetostriction at room temperature as well as high energy density and fast response velocity at low frequency^[1]. Wide application fields have been identified since the material had been exploited^[2]. The competition growth between the peritectic phase—(TbDy)Fe₂(REFe₂) and the primary phase—(TbDy)Fe₃(REFe₃) exists in the solidification processing of the peritectic Tb-Dy-Fe alloys, which has great effect on the microstructure and magnetostriction property of the materials, so that it is necessary to investigate the solidification mechanism of the materials.

Electromagnetic levitation melting is a containerless processing method. In such a way, the metallic sample is melted in a suspended state by high frequency electromagnetic field without contacting the crucible. This technique cancels the limitation of crucible materials, avoids the contamination of crucible wall, and inhibits the inhomogeneous nucleation leading by inner wall of crucible, so that the electromagnetic levitation melting is used to process high melting point or high reactive metallic materials and research the

undercooling, solidification and phase selection of metallic samples^[3-5].

As Tb-Dy-Fe alloy contains a large amount of RE element, the contamination and oxidation of the alloy, which may affect the solidification mechanism, are imperative in the process of alloys melting and solidification. For the advantage of electromagnetic levitation melting, considering the contamination and oxidation question, the scheme of vacuum electromagnetic levitation melting is adopted to process the alloy. For the first time, the vacuum electromagnetic levitation melting of the peritectic Tb-Dy-Fe alloy is realized, and the levitation melting process and solidification characteristics of the alloys are researched.

2 EXPERIMENTAL

Generally, Tb-Dy-Fe has composition approaching Tb_{0.27}Dy_{0.73}Fe₂. Concerning the melting loss, the Tb_{0.27}Dy_{0.73}Fe_{1.90} alloy for the levitation melting investigation was prepared from raw materials of the following purities: rare earth elements Tb 99.53%, Dy 99.53% and transition metal Fe 99.869%. The process of master alloy melting was carried out in a vacuum induction furnace under a high-purity argon atmosphere^[6]. The master alloy was cut into small

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cylinders which were used as levitation melting samples.

Static induction transistor (SIT) solid state high frequency power supplies with 200 kHz, 100 kW and specially designed levitation melting coil is selected to perform the electromagnetic levitation melting of the alloys in vacuum chamber. SONY video camera recorder records the levitation melting photographs of the materials. Optical microscope and scanning electron microscope (SEM) fitted with backscattered electron (BSE) detector are used to investigate the microstructure. Electric probe X-ray microanalysis (EPMA) are used to analyze the composition.

3 RESULTS AND DISCUSSION

3.1 Process analysis

It is well known that for the electromagnetic levitation melting of the metal materials, whether the electromagnetic levitation melting of the materials is easy or not decided by the electric conductivity and density of the materials. The density and electric conductivity of the $Tb_{0.27}Dy_{0.73}Fe_{1.90}$ alloy is $9.42 \times 10^3 \text{ kg/m}^3$ and $1.67 \times 10^6 \Omega^{-1}m^{-1}$ respectively. According to its high density and lower electric conductivity, the $Tb_{0.27}Dy_{0.73}Fe_{1.90}$ alloy is difficult to be electromagnetically levitated. From traditional theoretical calculation, the levitation forces increase with increasing frequency, so the power supply for levitation melting has rather high frequency. But concerning the levitation stability, there exists an upper limit frequency for a specific metallic samples. Exceeding the frequency, the rotating and oscillating destabilization can occur^[7]. From comprehensive consideration, the 200 kHz, 100 kW SIT power supply was selected to perform the electromagnetic levitation melting of the $Tb_{0.27}Dy_{0.73}Fe_{1.90}$ alloy. The element Dy of the $Tb_{0.27}Dy_{0.73}Fe_{1.90}$ alloy has large saturation vapor pressure, so it is necessary to backfill argon to reduce the loss of the element and preserve the nominal composition of the alloy. The purity of argon is 99.999%.

3.2 Electromagnetic levitation melting process

The process of the $Tb_{0.27}Dy_{0.73}Fe_{1.90}$ electromagnetic levitation melting is as follows: sample preparation (derusting, acetone cleaning, air dry) \rightarrow vacuum pumping \rightarrow filling with argon gas \rightarrow preheating levitation sample (low current and low voltage) \rightarrow levitation of solid sample (rapid rise of power) \rightarrow levitation of liquid sample (slight decrease of power). When the power decreases, the levitation sample falls down, which leads to the increase of absorption power of the sample, so the sample melts. With increasing temperature, the levitation of liquid metals is often unstable, partly because of the decrease in melt

surface force and electrical conductivity. For the $Tb_{0.27}Dy_{0.73}Fe_{1.90}$ sample, it can be stably levitated for a long time from solid to moderate melt overheat. Fig. 1 shows the transient levitation state of a liquid $Tb_{0.27}Dy_{0.73}Fe_{1.90}$ sample. If the overheating degree is too large, the time of stable levitation is very short, which can be contributed to the change of sample sizes for the severe loss of rare earth components especially for the high saturation vapor pressure of Dy and the deposition of rare earth element in the levitation melting coil.



Fig. 1 Levitation state of liquid $Tb_{0.27}Dy_{0.73}Fe_{1.90}$ sample in coil

3.3 Microstructure analysis

The microstructures of $Tb_{0.27}Dy_{0.73}Fe_{1.90}$ before and after levitation is shown in Fig. 2. The back-scattered SEM micrographs and micro-area compositions analyses of the sample after levitation melting are shown in Fig. 3 and Table 1. It should be pointed out that the white microstructure, the black microstructure, and the gray microstructure in the optical micrograph, correspond to the dark gray microstructure, the white microstructure, and the light gray microstructure in the back-scattered SEM micrograph, respectively.

Table 1 Component analyses for microstructure of $Tb_{0.27}Dy_{0.73}Fe_{1.90}$ after levitation melting (corresponding to Fig. 3)

	Light Grey microstruc. element	Dark gray microstruc. element	White microstruc. element	Average
Tb	9.52	6.52	19.87	9.27
Dy	23.64	18.54	58.82	24.17
Fe	66.84	74.94	21.3	66.56

According to Table 1, the average composition of the sample after electromagnetic levitation melting is $Tb_{0.28}Dy_{0.72}Fe_{1.99}$ around peritectic composition point; the light gray microstructure, the dark gray microstructure and the white microstructure, as shown in Fig. 3, correspond to $(TbDy)Fe_2$ -($REFe_2$)

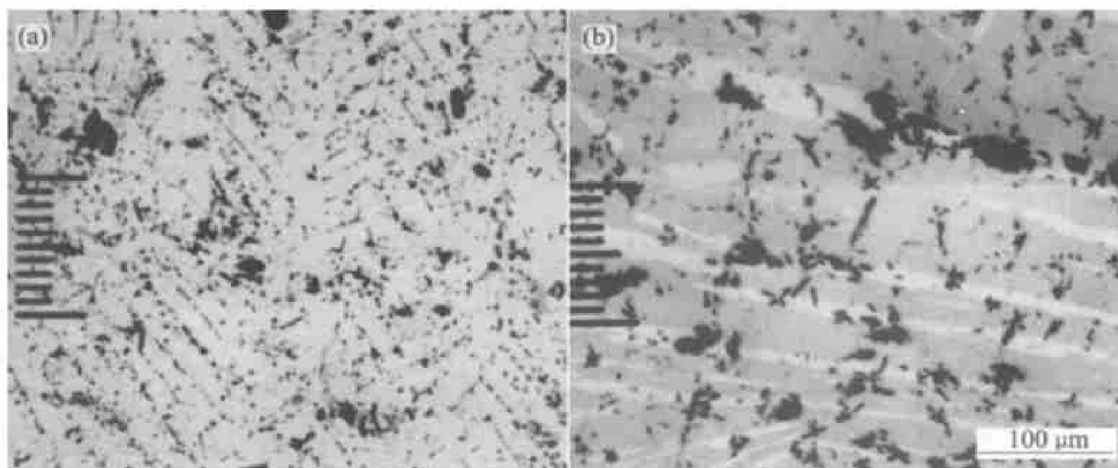


Fig. 2 Optical micrograph of Tb_{0.27}Dy_{0.73}Fe_{1.95} alloy
(a) —Before levitation; (b) —After levitation

phase, (TbDy)Fe₃(REFe₃) phase and RE rich phase, respectively. In other words, the gray microstructure, the white microstructure and the dark microstructure, as shown in Fig. 2(b) correspond to REFe₂ phase, REFe₃ phase and RE rich phase respectively. Before levitation melting, a small amount of rare earth phase as well as REFe₂ matrix phase are presented in the micrograph of this alloy, as shown in Fig. 2(a). The pseudo-binary equilibrium phase diagram of Tb_{0.27}Dy_{0.73}Fe_x is shown in Fig. 4^[8]. Based on the phase diagram, the microstructure of Tb_{0.28}Dy_{0.72}Fe_{1.99} should contain almost complete REFe₂ phase and a small amount of RE phase in the REFe₂ matrix phase under the conditions of equilibrium solidification^[9]. Contrary to the predication, a large amount of rod-like iron-rich phase REFe₃ phase, rare earth rich phase enriching around REFe₃ phase and a small amount of rare earth rich phase in the matrix phase, as well as REFe₂ Laves matrix phase are presented in the micrograph of this alloy after levitation melting, as shown in Fig. 2(b). The mechanism of this phenomenon is not very clear. In general condition, the REFe₂ matrix phase is formed by a peritectic reaction of the rare earth-rich liquid phase and primary REFe₃ phase. When the peritectic phase REFe₂ forms, it surrounds the primary REFe₃ phase and prevents further peritectic reaction. Through solute diffusion, the peritectic reaction is proceeding. If the solidification velocity is large, the peritectic reaction may proceed incompletely. Rare earth phase enriches in the liquid phase. Subsequent eutectic reaction results in a large amount of rare earth-rich phase enriching in the matrix REFe₂ phase and separated with the REFe₃ phase. Such microstructure formed through incomplete peritectic reaction is incongruent with the microstructure obtained through levitation melting, thus the possibility of incomplete peritectic reaction

can be eliminated. In 1959, Chalmers predicted that, except for the peritectic point composition, it should be possible to form peritectic phase and primary phase simultaneously near the peritectic composition. Experimental attempts to achieve two phase coupled growth in peritectic alloys only succeeded in studying Ni-Al^[10], Ti-Al^[11], Fe-Ni^[12], Zn-Ag^[13] and Nd-Ba-Cu-O^[14] alloys, thus the coupled growth for the solidification of Tb_{0.28}Dy_{0.72}Fe_{1.99} is possible. The coupled growth of the peritectic phase REFe₂ and the primary phase REFe₃ can explain the phase distribution of the abnormal microstructure after levitation melting. With decreasing temperature, the reaction is L (liquid phase) \rightarrow REFe₂ + REFe₃. Because the composition of liquid phase is similar to REFe₂, the growing velocity of REFe₂ is larger than that of REFe₃, which results in rod-like REFe₃ phase distributed in the REFe₂ matrix phase. The rare earth content of REFe₃ is lower than that of the REFe₂, which leads to the enrichment degree of the rare earth in the liquid phase around the REFe₃ higher than that in the liquid phase around REFe₂. When the temperature lowers to the eutectic temperature, the eutectic reaction $L \rightarrow$ RE (rare earth rich phase) + REFe₃ results in a large amount of rare earth-rich phase enriching around rod-like REFe₃, a small amount of rare earth-rich phase distributed in the matrix phase far from the REFe₃. From the equilibrium phase diagram of Tb_{0.27}Dy_{0.73}Fe_x, it can be concluded that the difference between the nucleation temperature of the REFe₃ phase and the peritectic temperature is small for the Tb_{0.28}Dy_{0.72}Fe_{1.99} alloy. Through electromagnetic levitation melting, the large undercooling degree may be obtained for the alloy. While the nucleation temperature of the alloy is below its peritectic temperature, the REFe₃ phase and the peritectic REFe₂ phase may precipitate simultaneously. This

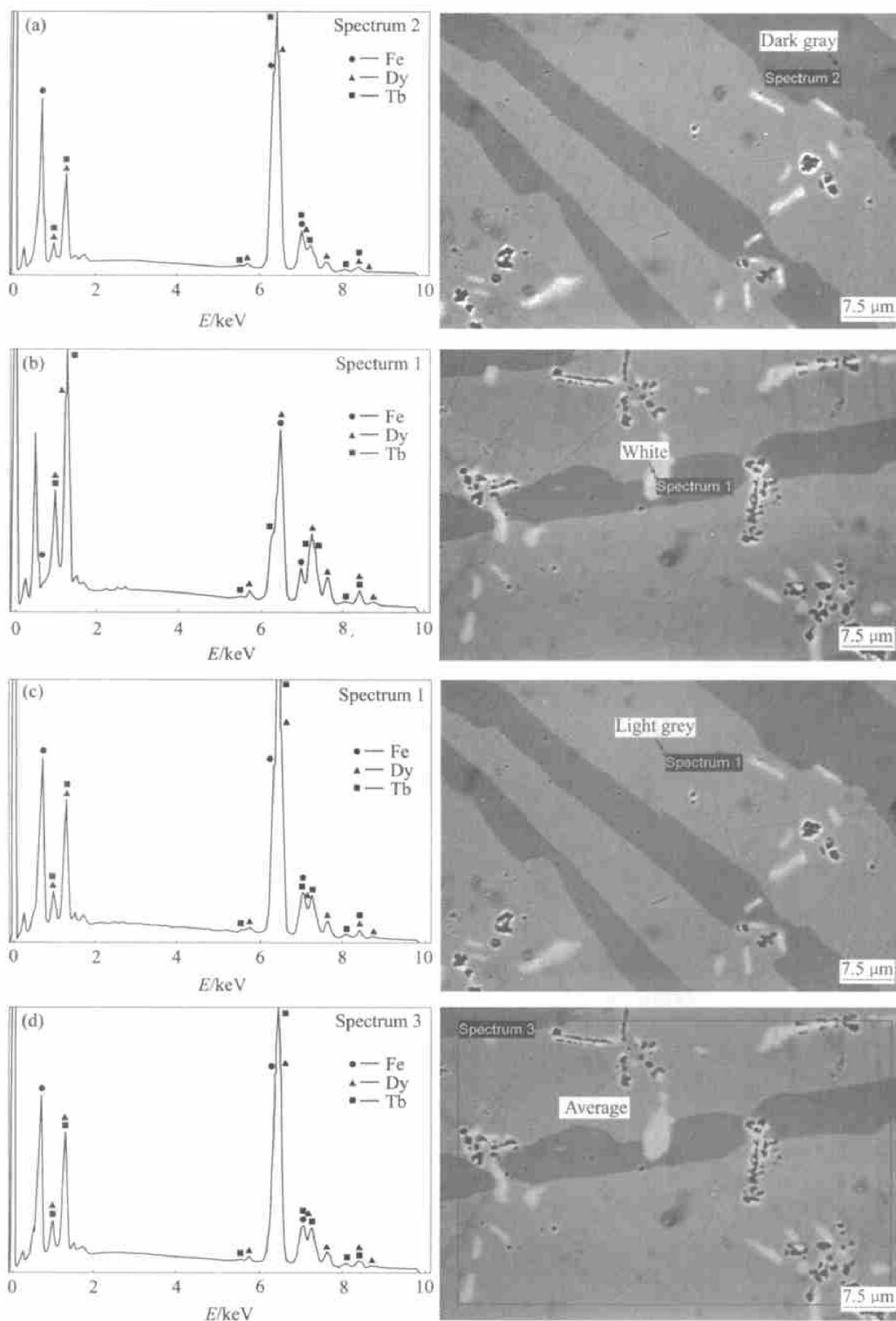


Fig. 3 Back-scattered SEM micrographs of sample after levitation melting and spectra of electric probe X-ray microanalysis (EPMA)

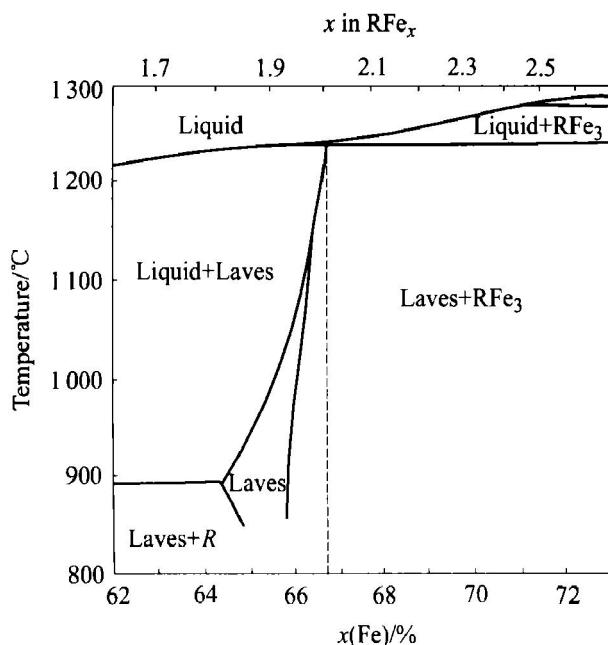


Fig. 4 Pseudobinary phase diagram of $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_x$

may explain the coupled growth of RFe_3 phase and peritectic RFe_2 phase.

4 CONCLUSIONS

1) The electromagnetic levitation melting of the rare earth giant magnetostriction material $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_{1.90}$ alloy, which has high density and lower electric conductivity, is realized. As the power supply decreases, the levitation-melting sample changes from solid to liquid. After levitation melting, the microstructure of the alloy near peritectic composition point comprises the matrix phase— RFe_2 , a large amount of rod-like iron-rich phase— RFe_3 embedding in the matrix phase, rare earth-rich phase enriching around RFe_3 phase and a small amount of rare earth rich phase in matrix phase. The formation of the microstructure can be attributed to the large undercooling degree of the alloy under this experimental condition, which leads to the coupled growth of the peritectic phase— RFe_2 and primary phase— RFe_3 . The coupled growth and subsequent eutectic reaction result in the microstructure after levitation melting.

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