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ABSTRACT

The SmFe₅ intermetallic compound was synthesized using the melt-spinning technique. A TEM study revealed that the as-rapidly quenched SmFe₅ melt-spun ribbon consisted of fine SmFe₅ grains. Subsequent annealing at various temperatures resulted in crystal growth of the SmFe₅ grains. The SmFe₅ melt-spun ribbon annealed at 1073 K still consisted of SmFe₅ grains together with a small amount of SmFe₃ grains. The specimen annealed at 1073 K exhibited a coercivity of 1.2 kOe.

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INTRODUCTION

Due to the increase in production of high-energy-product Nd-Fe-B permanent magnets, Nd-Fe-B permanent magnets are still the predominant target of research in this field.¹ Other studies have been focused on the search for new rare-earth intermetallic compounds such as the RFe₁₂ (R: rare-earth),²–⁸ R₃Fe₂⁹,⁹–¹³ and R₅Fe₁₇ intermetallic compounds.¹⁴–¹⁷ These compounds are considered to be highly promising, but have not yet been developed into high-performance permanent magnets. We have therefore continued the search for new rare-earth intermetallic compounds.

One of the most promising candidates is the SmFe₅ phase, which has a hexagonal structure like that of the well-known SmCo₅ magnet. In studies on sputtered films, Sm(Fe,Cu)₅ film with the SmFe₅ phase has been produced.¹⁸,¹⁹ Although specimens the SmFe₅ metastable phase have not yet been produced, the SmFe₅ metastable phase has been detected in amorphous Sm-Fe melt-spun ribbons that have been subjected to annealing.²⁰–²³ In recent years, we have succeeded in producing another Sm-Fe metastable phase, the Sm₅Fe₁₇ phase, by rapid solidification processing using the melt-spinning technique.²⁴ The purpose of this study was to seek the possibility of producing binary Sm-Fe alloys with the SmFe₅ phase by optimizing the annealing conditions of rapidly quenched melt-spun ribbons. The relationship between the structures and magnetic properties of the Sm-Fe melt-spun ribbons is discussed.

EXPERIMENT

SmFe₅ alloy ingots were prepared by induction melting under an argon atmosphere. The molten alloy ingots were then ejected through an orifice with argon onto a copper wheel rotating at a surface velocity of 50 ms⁻¹. The resultant melt-spun specimens were obtained as continuous ribbons (thickness 20 μm; width 2 mm). The melt-spun ribbons were annealed under an argon atmosphere at 773–1073 K for 1 h. The phases in the specimens were investigated by X-ray diffraction (XRD) using Cu Kα radiation. The microstructures of the specimens were examined using a transmission electron microscope (TEM) after ion-beam thinning. The magnetic properties of the specimens were measured at room temperature using a vibrating sample magnetometer (VSM) with a maximum applied field of 25 kOe.

RESULTS AND DISCUSSION

Since the SmFe₅ phase is metastable, it is impossible to produce alloys with an SmFe₅-type phase using the conventional casting method. Thus, the specimens were prepared by rapid solidification processing using melt-spinning. Figure 1 shows the XRD pattern of the as-rapidly quenched SmFe₅ melt-spin ribbon. The XRD pattern has a fairly broad halo-like peak, but small and somewhat broad crystalline peaks can also be seen, which are not identified.
In order to examine the structure of the SmFe$_5$ melt-spun ribbon, a TEM study was carried out. Figure 2 shows a TEM micrograph of the as-rapidly quenched SmFe$_5$ melt-spun ribbon and the corresponding selected electron diffraction pattern. A representative one-dimensional electron diffraction pattern obtained from the selected area diffraction pattern of the SmFe$_5$ melt-spun ribbon is shown below the micrograph. The TEM micrograph shows fine grains with an average diameter of 4–5 nm. The electron diffraction pattern indicates that the fine grains are the SmFe$_5$ phase.

FIG. 1. XRD pattern of the as-rapidly quenched SmFe$_5$ melt-spun ribbon.

FIG. 2. TEM micrograph of the as-rapidly quenched SmFe$_5$ melt-spun ribbon and the corresponding selected electron diffraction pattern. A representative one-dimensional electron diffraction pattern obtained from the selected area diffraction pattern of the SmFe$_5$ melt-spun ribbon is shown below the micrograph. The thicker red bars below the electron diffraction pattern indicate the peak positions calculated from the unit cell parameters for the SmFe$_5$ phase.

FIG. 3. XRD patterns of the SmFe$_5$ melt-spun ribbon annealed at (a) 773 K, (b) 873 K, (c) 973 K, and (d) 1073 K.

FIG. 4. Hysteresis loops of (a) the as-rapidly quenched SmFe$_5$ melt-spun ribbon and (b) the SmFe$_5$ melt-spun ribbon annealed at 1073 K.
This confirms that the as-rapidly quenched SmFe$_3$ melt-spun ribbon consisted of SmFe$_3$ grains.

Annealing of the as-rapidly quenched SmFe$_3$ melt-spun ribbon was carried out to investigate the resultant changes in structure and magnetic properties. Figure 3 shows the XRD patterns of the SmFe$_3$ melt-spun ribbon annealed at 773–1073 K. The diffraction peaks of the SmFe$_3$ phase become clearer and larger as the annealing temperature increases. This suggest that annealing of the as-rapidly quenched SmFe$_3$ melt-spun ribbon resulted in crystal growth of the SmFe$_3$ grains.

Figure 4 shows the hysteresis loops of the as-rapidly quenched and annealed SmFe$_3$ melt-spun ribbons. The as-rapidly quenched SmFe$_3$ melt-spun ribbon exhibited a coercivity of 0.45 kOe. The coercivity of the annealed SmFe$_3$ melt-spun ribbon gradually become higher as the annealing temperature increased. The SmFe$_3$ melt-spun ribbon annealed at 1073 K exhibited a coercivity of 1.2 kOe. However, although this specimen exhibited a coercivity exceeding 1 kOe, the level of coercivity is not sufficiently high for the permanent magnet applications. It has been reported that the addition of other elements such as Ti or V stabilizes the metastable SmFe$_{12}$ phase and that the Sm(Fe$_x$M)$_{2}$ (M = Ti, V) phase exhibited high coercivity.$^{1,2}$ The addition of another element to the SmFe$_3$ phase would therefore be expected to stabilize the SmFe$_3$ phase and lead to a further increase in coercivity.

The structure of the SmFe$_3$ melt-spun ribbon annealed at 1073 K was examined by TEM. The result is shown in Figure 5. The annealed specimen consisted of fine grains of around 100 nm in diameter. The electron diffraction pattern indicates that the fine grains are not only the SmFe$_3$ phase but also the SmFe$_{17}$ phase. Further optimization of the annealing conditions together with compositional modification may lead to the production of specimens containing only the SmFe$_3$ phase.

CONCLUSION

An Sm-Fe binary alloy with the metastable SmFe$_3$ phase was produced by melt-spinning. It was found that the as-rapidly quenched SmFe$_3$ melt-spun ribbon consisted of SmFe$_3$ grains and exhibited a coercivity of 0.45 kOe. Annealing of the as-rapidly quenched SmFe$_3$ melt-spun ribbon resulted in an increase in coercivity. The SmFe$_3$ melt-spun ribbon annealed at 1073 K exhibited a coercivity of 1.2 kOe, although a TEM study revealed that the annealed specimens consisted of SmFe$_3$ grains together with a small amount of SmFe$_5$ grains. This suggests that annealing at a high temperature not only resulted in crystal growth of the metastable SmFe$_3$ grains but also in nucleation of the stable SmFe$_3$ phase. Since no clear diffraction peaks of other phases such as the Fe and Sm$_2$Fe$_7$ phases were detected in the electron diffraction pattern, the observed SmFe$_3$ phase may be an off-stoichiometric SmFe$_3$ phase; in other words, an Fe-rich SmFe$_3$ phase. Further optimization of the annealing conditions together with compositional modification may lead to the production of specimens containing only the SmFe$_3$ phase.

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REFERENCES

1. K. Hono and H. Sepehri-Amin, Scripta Mater. 151, 6 (2018).
2. L. X. Liao, Z. Altounian, and D. H. Ryan, J. Appl. Phys. 70, 6006 (1991).
3. Y. Wang, G. C. Hadjipanayis, A. Kim, N. C. Liu, and D. J. Sellmyer, J. Appl. Phys. 67, 4954 (1990).
4. Y. Otani, H. S. Li, and J. M. D. Coey, IEEE Trans. Magn. 26, 2658 (1990).
5. Y. Yang, P. Oleinek, and K.-H. Muller, J. Appl. Phys. 88, 988 (2000).
6. E. Tomey, M. Bacmann, D. Fruchart, J. L. Soubeyroux, and D. Gignoux, J. Alloys Compd. 231, 195 (1995).
7. K. Kobayashi, S. Suzuki, T. Kuno, K. Urushibata, N. Sakuma, M. Yano, T. Shouji, A. Kato, and A. Manabe, J. Alloys Compd. 694, 914 (2017).
8. A. M. Gabay and G. C. Hadjipanayis, Scripta Mater. 154, 284 (2018).
9. M. Cadogan, H. S. Li, R. L. Davis, A. Margarian, S. J. Collocott, J. B. Dunlop, and P. B. Gwan, J. Appl. Phys. 75, 7114 (1994).
Z. Hu and W. B. Yelon, Solid State Commun. 91, 223 (1994).
11 O. Kalogirou, V. Psycharis, L. Saettas, and D. Niarchos, J. Magn. Magn. Mater. 146, 335 (1995).
12 H. S. Li, J. M. Cadogan, R. L. Davis, A. Margarian, and J. B. Dunlop, Solid State Commun. 90, 487 (1994).
13 V. Psycharis, O. Kalogirou, D. Niarchos, and M. Gjoka, J. Alloys Compd. 234, 62 (1996).
14 F. J. Cadieu, H. Hegde, R. Rani, A. Navarathna, and K. Chen, Mater. Lett. 11, 284 (1991).
15 H. S. Li, J. M. Cadogan, R. L. Davis, A. Margarian, and J. B. Dunlop, Solid State Commun. 90, 487 (1994).
16 F. J. G. Landgraf, F. P. Misell, H. R. Rechenberg, T. T. Schneider, P. Villas-Boas, J. M. Moreau, L. Paccard, and J. P. Nozières, J. Appl. Phys. 70, 6125 (1991).
17 T. Saito, J. Magn. Magn. Mater. 440, 315 (2007).
18 O. Yabuhara, M. Ohtake, Y. Nukaga, F. Kirino, and M. Futamoto, J. Phys.: Conf. Ser. 200, 082026 (2010).
19 T. Yanagawa, M. Ohtake, F. Kirino, and M. Futamoto, EPJ Web of Conferences 40, 06007 (2013).
20 Y. Xingbo, T. Miyazaki, K. Takakura, K. Hisatake, T. Hattori, and M. Takahashi, J. Magn. Magn. Mater. 62, 293 (1986).
21 Y. Xingbo, T. Miyazaki, T. Izumi, H. Saito, M. Takahashi, and M. Takahashi, IEEE Trans. Magn. MAG-23, 3104 (1987).
22 Q. F. Xiao, Z. D. Zhang, T. Zhao, W. Liu, Y. C. Sui, X. G. Zhao, and D. Y. Geng, J. Appl. Phys. 82, 6170 (1997).
23 T. Miyazaki, M. Takahashi, Y. Xingbo, H. Saito, and M. Takahashi, J. Magn. Magn. Mater. 75, 123 (1988).