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Annealing effects in amorphous Fe₇₇Gd₃B₂₀ ribbons

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Abstract

The crystallization kinetics of the amorphous alloy Fe₇₇Gd₃B₂₀ were examined under isothermal flash annealing conditions both in air and in a vacuum. Magnetic measurements were used to determine the reaction order (n) and activation energy (E_a), and Mössbauer spectroscopy (MS) was used to identify some resulting crystalline phases.

Keywords: RE–TM amorphous alloy; Crystallization kinetics; Mössbauer spectroscopy

1. Introduction

Ferromagnetic amorphous alloys of transition metals (TM) with rare earth (RE) contents are of interest due to their thermal stability and because their magnetic properties are controlled by the fluctuating local anisotropy induced by the RE ions [1]. The magnetic behaviour of TM–RE–B amorphous alloys is controlled by both the RE–TM exchange interaction and the local crystalline field acting on the RE ion sites [2,3].

Buschow et al. [4] have examined the thermal stability of the RE_{1-x}Co_x (RE–La, Gd, Tb, ...) ($0.3 \leq x \leq 0.6$) amorphous alloys, showing that the crystallization temperature T_x depends on both the RE atomic number and the Co concentration.

Differential thermal analysis (DTA) confirmed that the Fe_{80-x}Sm_xB₂₀ ($1 \leq x \leq 9$) amorphous alloys exhibit up to three crystallization stages, depending on the RE content [5].

The crystallization kinetics of several TM–RE amorphous alloys were examined in Refs. [6,7] and their magnetic properties were reported in Refs. [8,9]. MS data of Fe_{80-x}Gd_xB₂₀ ($0 \leq x \leq 20$) amorphous thin films revealed the dependence of the spin orientation and the hyperfine field on both the Gd content and the preparation conditions [10]. In the present paper we examine some aspects of the crystallization kinetics of Fe₇₇Gd₃B₂₀ amorphous alloy.

2. Experimental

Amorphous Fe₇₇Gd₃B₂₀ ribbons were prepared by the single-roller technique and their amorphous state was confirmed by both X-ray diffraction (XRD) and MS.

Isothermal ‘flash’-type annealing both in air and in vacuum were performed; the annealing temperatures ($T_a = 828$ and 838 K) were held within ± 1 K. The technical saturation magnetic moments (\mathcal{M}_s) at room temperature of the amorphous phase in the samples were measured using a hysteresigraph described in Ref. [11]. The cycling time was 36 s and the error in determining \mathcal{M}_s did not exceed 2% of the value $\mathcal{M}_s(0)$, corresponding to the as-quenched state.

The annealing time (t_a)-dependent volumic fraction $X(t_a)$ of the crystalline phase was expressed as

$$X(t_a) = 1 - \frac{\mathcal{M}_s(t_a)}{\mathcal{M}_s(0)}. \quad (1)$$

The amorphous and crystalline phases were identified by fitting the data given by a standard-constant acceleration Mössbauer spectrometer using a ⁵⁷Co in Rh matrix source.

3. Results and discussion

The evolution of $X(t_a)$ for three Fe₇₇Gd₃B₂₀ samples is shown in Fig. 1. The ribbons were partially crystallized both in air ($T_a = 828$ K, $X < 0.7$) and in vacuum ($T_a = 828$ K, $T_a = 838$ K, $X < 0.4$).

Assuming that the volumic fraction of the crystallized phase obeys a Johnson–Mehl–Avrami law,

$$X(t_a) = \sum_i a_i \left\{ 1 - \exp \left[- (k_i (t_a - t_{0i}))^{n_i} \right] \right\}, \quad (2)$$

with $a_i < 1$ the amplitude, k_i the reaction rate, n_i the order, t_{0i} the incubation time of the i th reaction, fitting procedures of the experimental data were performed. In the case of the air-annealed sample two diffusion-controlled

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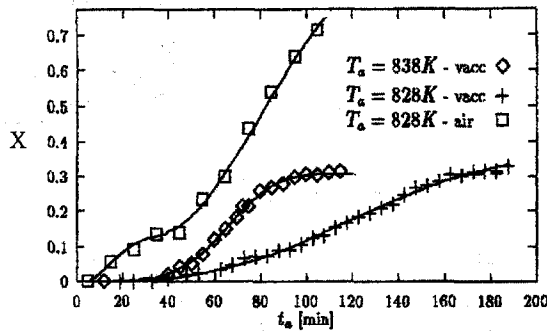


Fig. 1. Annealing time dependence of crystallized fraction for $\text{Fe}_{77}\text{Gd}_3\text{B}_{20}$ ribbons.

microcrystal growth processes ($n_1 = 1.5$, $n_2 = 1.8$) were identified.

The vacuum-annealed partially crystallized ($X < 0.4$) samples exhibit single-process kinetic reactions with $n = 2.9$ ($T_a = 828$ K) and $n = 4.8$ ($T_a = 838$ K), respectively; the responsible mechanisms are nucleation and grain growth and the mean activation energy of the process is $E_a = 3.96$ eV.

The significant differences in the crystallization behaviour between the air-annealed and vacuum-annealed samples can be attributed to surface oxidation.

The room-temperature Mössbauer spectra of some $\text{Fe}_{77}\text{Gd}_3\text{B}_{20}$ samples are given in Fig. 2. The as-quenched ribbons exhibit a typical amorphous spectrum (Fig. 2a) with a mean hyperfine field of 23.0 T and with a mean spin direction of 63° out of the ribbon plane. Annealing for short times ($t_a \approx 15$ min) and at moderate temperatures ($T_a \leq 670$ K) causes structural relaxation with spin rotation towards the ribbon plane.

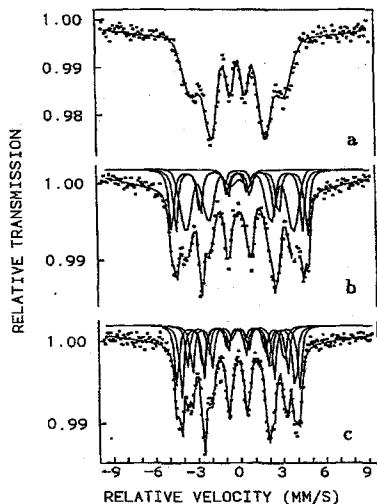


Fig. 2. Mössbauer spectra of $\text{Fe}_{77}\text{Gd}_3\text{B}_{20}$ ribbons: (a) as quenched; (b) partially crystallized at $T_a = 838$ K, $t_a = 100$ min; (c) fully crystallized at $T_a = 838$ K, $t_a = 150$ min.

Annealing at higher temperatures activates local crystallization ($X \approx 0.53$) and, as a consequence, various magnetic patterns superimpose on the Mössbauer spectrum, as can be seen in Fig. 2(b); they correspond to Fe_3B (with hyperfine fields 27.75 and 29.7 T) and to some nanocrystalline phase associated with a superparamagnetic-type doublet, respectively, while the Gd atoms remain in the amorphous phase (mean $H_{\text{hyp}} = 23.0$ T).

Vacuum annealing ($t_a = 150$ min, $T_a = 838$ K) produced full crystallization of the alloy and the corresponding Mössbauer spectrum (Fig. 2c) was fitted with four sextets: three of them ($H_{\text{hyp}} = 28.4$, 26.2 and 22.3 T) were attributed to the different iron sites corresponding to Fe_3B phase [12,13], and one ($H_{\text{hyp}} = 22.0$ T) to an iron site with 3 B and 1 Gd neighbours.

4. Conclusions

The crystallization of the vacuum-isothermally annealed $\text{Fe}_{77}\text{Gd}_3\text{B}_{20}$ amorphous ribbons are mainly based on Fe_3B grain nucleation and growth. The mean activation energy of the process is 3.96 eV, and the reaction order increases from 2.9 to 4.4 with increasing T_a .

The crystallization kinetics of the isothermally air-annealed $\text{Fe}_{77}\text{Gd}_3\text{B}_{20}$ ribbons are affected by surface oxidation which diminishes their thermal stability.

References

- [1] S. Tejada, B. Martinez, A. Labarta, R. Grössinger and A. Hernando, *J. Appl. Phys.* 67 (1990) 5984.
- [2] T.S. Zhao, *J. Appl. Phys.* 75 (1994) 3008.
- [3] S.H. Aly, G.N. Nicolaidis, Y.F. Tao and G.C. Hadjipanayis, *J. Phys. F: Metal Phys.* 16 (1986) L21.
- [4] K.H. Buschow and N.M. Beekmans, *Phys. Status Solidi (a)* 60 (1980) 193.
- [5] B. Idzikowski and A. Wrzeciono, *Phys. Status Solidi (a)* 108 (1988) 375.
- [6] F.E. Luborski, *J. Non-Cryst. Solids* 61–62 (1984) 829.
- [7] F. Leccabue, B.E. Wate, D. Carrillo, J.L. Sanchez, G. Bocelli, G. Galestani, L. Dimesso, A. Deriu and D. Negri, *J. Magn. Magn. Mater.* 115 (1992) 190.
- [8] J.P. Sinecher, M. Knobel, R. Grössinger and R.S. Turtelli, *J. Magn. Magn. Mater.* 133 (1994) 270.
- [9] V. Kuncser, G. Filoti, A. Avramescu, L. Cristea, A. Jianu, J.L. Dormann, M. Nogués and M. Seqqat, ICAME-95 Rimini (Italy), September 1995.
- [10] G. Filoti, V. Kuncser, M.D. Şerbănescu, A. Simu, M. Nogués, J.L. Dorman and J. Maknani, *IEEE Trans. Magn.* 29 (1993) 3123.
- [11] A. Ercuța and I. Mihalca, *Lucr. Semin. Mat. Fiz.*, IPTV Timișoara, November 1983, p. 126.
- [12] J.M. Dubois, M. Bastick, G.L. Caer and C. Tete, *Rev. Phys. Appl.* 15 (1980) 1103.
- [13] H. Franke and M. Rosenberg, *J. Magn. Magn. Mater.* 7 (1978) 168.