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Phase equilibria and stability of B2 and $L2_1$ ordered phases in the Co–Fe–Ga Heusler alloy system

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Abstract

The phase equilibria, A2/B2 and B2/$L2_1$ order–disorder transitions and ferromagnetic/paramagnetic transition on the Co–Fe side of the Co–Fe–Ga system were examined by electron probe microanalysis (EPMA), differential scanning calorimetric (DSC) measurement and vibrating sample magnetometer (VSM). The equilibrium compositions of interrelations mainly among γ (A1: disordered fcc-Co), α (A2: disordered bcc-Fe), β (B2: ordered bcc-CoGa or FeGa) and β′ ($L2_1$ or $D0_3$: ordered bcc-Co_2FeGa or Fe_3Ga) phases were determined using diffusion couples and two-phase bulk specimens. It was confirmed that a bcc single-phase region composed of α, β and β′ at 700–1000° C exists in a wide composition range and that the ferromagnetic region appearing on the Co–Fe side extends to the center of the Gibbs triangle with decreasing temperature. On the other hand, the β′ phase appears along the Fe_3Ga–Co_3Ga section and the maximal critical temperature of the B2/$L2_1$ order–disorder transformation was determined to be 825 °C at a stoichiometric composition expressed as Co_2FeGa. The obtained phase diagram was examined in comparison with that of the Co–Fe–Al system.

Keywords: Intermetallics; Data storage materials; Magnetic measurements

1. Introduction

Many Co-based $L2_1$ (full-Heusler)-type alloys, in which half-metallic magnetic (HMM) behavior useful in the field of spintronics is expected, have been extensively studied from both theoretical and experimental viewpoints [1–15]. Recently, it has been demonstrated in the Co_2CrAl alloy, theoretically expected to be one of the most excellent HMM materials, that an inevitable A2–B2 spinodal decomposition [12] strongly affects the HMM properties. On the other hand, Umetsu and Kobayashi et al. pointed out from their investigations of magnetic properties and electronic structures that Co_2CrGa and $Co_2Cr_{1-x}Fe_xAl_{1-y}Ga_y$ Heusler alloys showing no spinodal decomposition may be promising candidates for applications to spintronic devices due to their HMM properties such as high spin polarization [13,14]. Since the HMM properties, including spin polarization, strongly depend on the crystal structure [15] and the degree of order of the $L2_1$ phase [9,16], detailed information on the phase stability is useful for realizing HMM characteristics. In the case of the $Co_2Cr_{1-x}Fe_xAl_{1-y}Ga_y$ Heusler alloy system, basic information on the phase equilibria among the related phases, including some ordered phases in the ternary systems Co–(Cr or Fe)–(Al or Ga), are very important for material design. In these basic ternary systems, although the Co–(Cr or Fe)–Al systems have already been established [17–19] and the Co–Cr–Ga system was recently investigated by the present authors' group [20], there has been no report on the phase equilibria for the Co–Fe–Ga system.

Fig. 1 shows the phase diagram of the Fe–Co [21], Fe–Ga [22] and Co–Ga [23] binary systems. It can be seen that single-phase regions of the bcc phase, including the β (A2: disordered bcc-Fe), β (B2: ordered bcc-CoFe, CoGa or FeGa) and β′ ($D0_3$: ordered bcc-Fe_3Ga) phases, exist in the every binary system and that the order–disorder transformations from A2 to B2 and/or from B2 to $D0_3$ appear in the Co–Fe and Fe–Ga systems. Another interesting point is the magnetic transformation which appears on the Co–Fe side. Recently, Kamiya et al. [19] reported that there is a significant interaction in phase stability between

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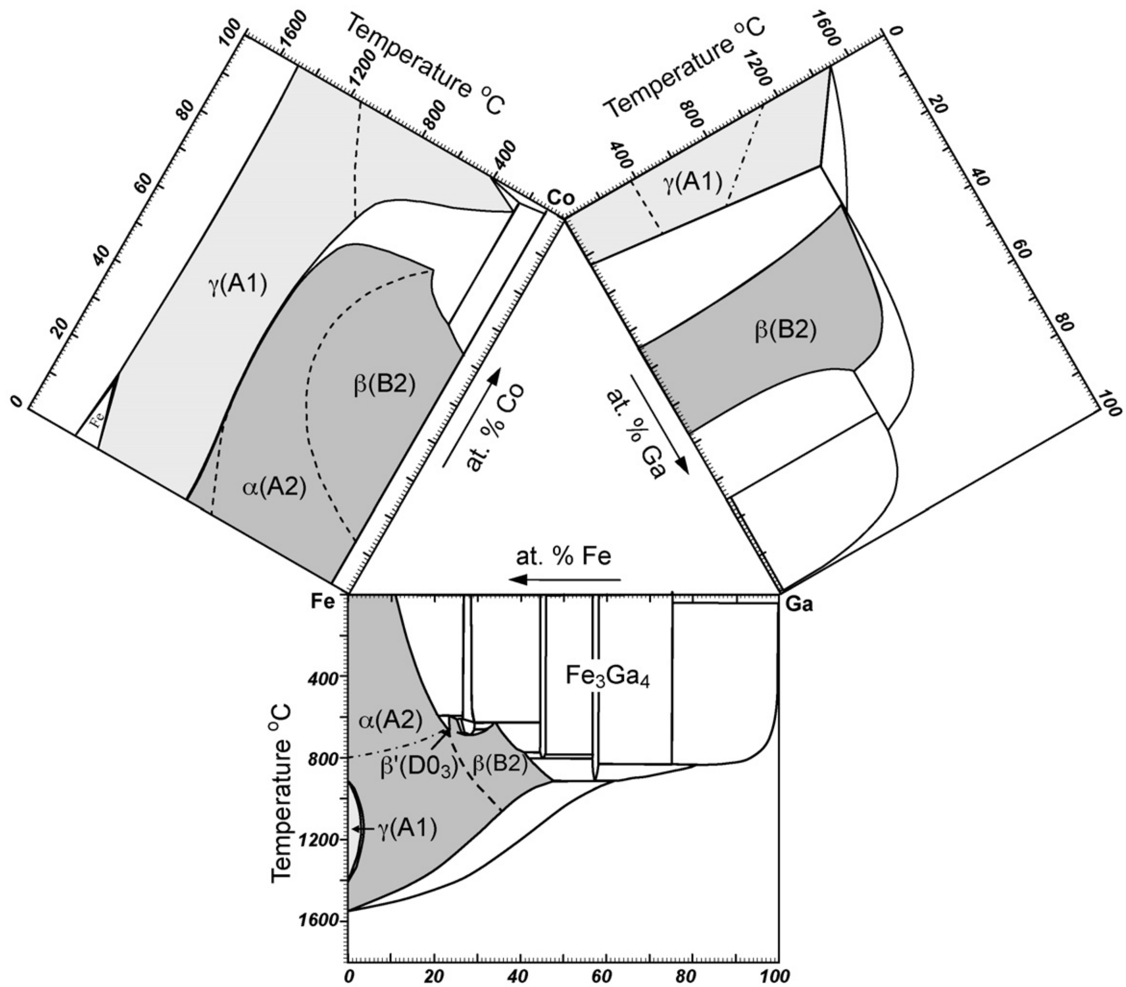

Fig. 1. Binary phase diagrams constituting the Co–Fe–Ga ternary system [21–23].

the magnetic and ordering transtions in the Co–Fe–Al system. In the present study, phase equilibria among the γ (A1: disordered fcc-Co), α, β, β′ and Fe_3Ga_4 (*C*2/*m*) phases at 700, 850 and 1000 °C in the Co–Fe–Ga system were determined, and the order–disorder and magnetic transition temperatures were also evaluated in a wide range of the bcc phase region.

2. Experimental procedures

Since there is no information on the Co–Fe–Ga ternary phase diagram, a special diffusion couple (DC) technique illustrated in Fig. 2 was applied to determine phase equilibria in a wide concentration region of some isothermal sections. The diffusion couples were prepared by the following three steps:

(1) Pure Fe (99.9%) and Co (99.9%) blocks were joined with a clamp consisting on two stainless steel disks and three molybdenum bolts at 1000 °C for 1 h in an evacuated quartz tube, and the DC specimens taken out of the clamp after the joining treatment were diffusion-annealed at 1000 °C for 4 h in an evacuated quartz tube to obtain a continuous diffusion zone between Co and Fe blocks. (Fig. 2(a)).
(2) A cylindrical hole with a diameter of 3 mm was formed by drilling near the diffusion zone of the Fe/Co DC, and some Ga (99.9999%) chips were inserted into the hole after removal of the surface layer with a nitric acid solution as shown in Fig. 2(b). Triple DCs were finally obtained by diffusion-annealing at 700 °C for 30 h, 850 °C for 10 h or 1000 °C for 4 h (Fig. 2(c)).
(3) The obtained DCs were cut from a section vertical to the Ga cylinder, and after polishing, were chemically analyzed using an electron microprobe analyzer (EPMA) along some lines parallel to the Fe/Co interface as shown in Fig. 2(d) and (e). Consequently, the information on phase equilibrium along the diffusion paths demonstrated in Fig. 2(f) was extracted.

In the present study, the obtained DCs covered most of the whole composition range in the ternary system except the Ga-rich corner in which a liquid phase appeared.

Two kinds of alloys, i.e., $Co_{70}Fe_{10}Ga_{20}$ and $Co_{70}Fe_{20}Ga_{10}$, were also prepared in order to determine the critical temperatures of the order–disorder and magnetic transitions. These alloys were induction-melted from pure Co, Fe and Ga, and annealed for an appropriate time depending on the annealing temperature in the quartz tube under an argon atmosphere. After the annealing, the specimens were quenched in ice water. Metallographic examination was performed by optical microscopy (OM) and scanning electron microscopy (SEM) with a composition imaging mode using back-scattering electrons (BSE).

The order–disorder and magnetic transformation temperatures were determined by differential scanning calorimetry (DSC) at a heating rate of 5 °C/min. When the DSC examination was not sufficient to determine the magnetic transition temperature (T_c) or when identification of the T_c in some DSC peaks was needed, the determination of T_c was performed with a vibrating sample magnetometer (VSM) at a heating rate of 2 °C/min.