Phase Stability and Site Occupancy for the L21-type phase in Co2YGa (Y = Ti, V, Cr, Mn and Fe) Alloys

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Many Co2YZ L21 (full-Heusler)-type half-metallic ferromagnets (HMFs) have been extensively studied from both theoretical and experimental points of view due to their potential applications in spin-dependent devices such as magnetic random access memories (MRAMs). From the recent studies [1-3] on Co-, Ni-, and Cu-based X2YZ L21-type alloys, it has been confirmed that the phase stability of L21-type alloys strongly depends on the kind of elements occupying the Y (4a) site and that the order-disorder transition temperature from the L21 to B2-type phase Tt(B2/L21) tends to decrease with increasing electron number of the Y element in the periodic table, except for in the case of Cr. The Tt(B2/L21) of Co2CrGa is the lowest among the Co2YGa (Y = Ti, V, Cr, Mn and Fe) alloys. In the present study, the site occupancy in the L21-type phase of Co2YGa alloys was investigated to clarify the correlation between the phase stability and the site occupancy.

The specimens were prepared by induction melting under an argon atmosphere. After alloying, each ingot was annealed at 1373 K for 72 hours and quenched in ice water. Powdered specimens were additionally annealed at 773 K for 2 hours. Neutron powder diffraction experiments were carried out using the Kinken powder diffractometer, HERMES [4], of the Institute for Materials Research (IMR), Tohoku University, installed in the JRR-3M reactor at the Japan Atomic Energy Agency (JAEA), where the wavelength was 0.182646 nm. The neutron diffraction data were analyzed with the Rietveld method using the RIETAN2000 program [5].

Figure 1 shows the observed and calculated neutron diffraction patterns and their differences between them at 300 K in the paramagnetic state for the Co2TiGa alloy which has a Curie temperature at TC=137 K. It was confirmed from the Rietveld analysis that the lattice constant a = 0.5858 nm, that the 8c site is entirely occupied by Co atoms, and that the 4a site is occupied by the Ti atoms with the site occupancy g = 0.96. In order to simplify this analysis, we introduced an assumption that the Co (8c) site is not occupied by Ga atoms [6], which is supported by the thermodynamic calculation based on the Bragg-Williams-Gorsky (BWG) approximation [7]. The high site occupancy obtained for the L21-type phase in the Co2TiGa alloy is consistent with the high phase stability that is suggested by the fact that the order-disorder transition temperaturefrom the L21 to B2-type phase Tt(B2/L21) is very high at 2390 K [3].

The observed and calculated neutron diffraction patterns and the difference betwen them at 573 K in the Co2CrGa alloy are shown in Fig. 2; the TC of this alloy is known to be 495 K [3]. Structural parameters including the site occupancy were refined under the same assumption as that for the Co2TiGa alloy. We have obtained the following results; the lattice constant a = 0.5805 nm, the 4a site is occupied by Cr atoms with g = 0.86, the 4b site is occupied by Ga atoms with g = 0.88, and the 8c site is occupied by the Co atoms with g = 0.99. It was confirmed that while the site occupancy of the 8c site by the Co atoms in the Co2CrGa alloy as well as that in the Co2TiGa alloy is almost perfect, the site occupancies of the 4a and 4b sites are lower than those in the Co2TiGa alloy.

According to the BWG approximation [6],

the Tt(B2/L21) of the stoichiometric X2YZ alloy is simply indicated by the following relation under the assumption that the occupancy of the 8c site by the X atoms is perfect at the ordering temperature Tt(B2/L21) [8]:

$$Tt(B2/L21) = 3W(YZ)/2k,$$

where W(YZ) is the interchange energy of the Y-Z bond between the second nearest neighbors and k is the Boltzmann constant. The above equation means that there is a linear relation between Tt(B2/L21) and W(YZ). Therefore, it can be concluded that the difference of the site occupancy between Co2TiGa and Co2CrGa can be attributed to the difference of the W(YZ), which can be evaluated from the Tt(B2/L12) of Co2CrGa (1056K) and Co2TiGa alloys (2390K) by Eq 1. Further systematic quantitative analysis of the site occupancy in Co2YGa (Y = V, Cr, Mn and Fe) alloys is needed to clarify the relation between the phase stability and the site occupancy.

In conclusion, the site occupancies of the Co2TiGa and Co2CrGa alloys were investigated and it was found that for the Co2TiGa alloy, the 4a, 4b and 8c sites are almost entirely occupied by Ti, Ga and Co atoms, respectively. On the contrary, for the Co2CrGa alloy, the 4a and 4b sites are occupied by the Cr and Ga atoms with g = 0.85 and 0.88, respectively. These results can be explained by the difference of the order-disorder transition temperature Tt(B2/L21) from L21 to the B2-type phase for these two alloys.

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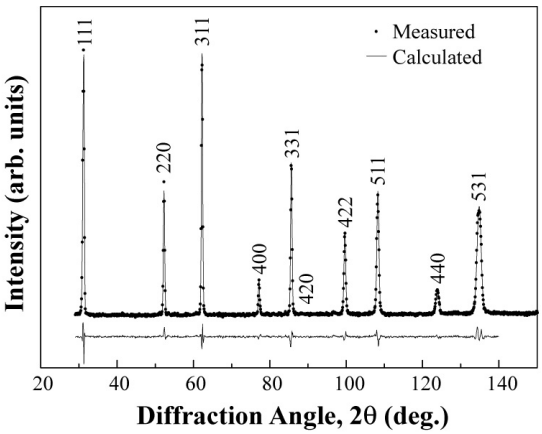

Fig. 1 The observed and calculated neutron powder diffraction patterns at 300 K for the Co_2TiGa alloy and the difference between the two.

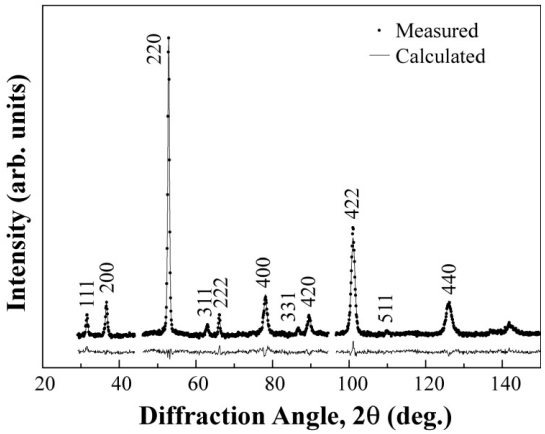

Fig. 2 The observed and calculated neutron powder diffraction patterns at 573 K for the Co_2CrGa alloy and the difference between the two.

Fig. 1.