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Properties of Fe–Ga based powders prepared by mechanical alloying

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Abstract

Alloys of Fe–Ga with starting compositions of 17, 19, 21, 23, and 25 at% Ga and $Fe_{81}Ga_{17}Z_2$ (Z = Si, Sn) have been prepared by mechanical alloying. Samples were milled in a SPEX Model 8000 mill with a ball to sample weight ratio of about 4:1. Phase formation as a function of milling time has been investigated for the 19 at% Ga sample and suggests that milling times of 12 h produce fully alloyed samples. Alloys have been studied by electron microprobe, X-ray diffraction, vibrating sample magnetometery and ^{57}Fe Mössbauer effect spectroscopy. Fully milled powders have measured compositions of $Fe_{100-x}Ga_x$ with $x = 15.7$, 17.0, 19.0, 22.4, and 24.0 and $Fe_{83.1}Ga_{15.2}Z_{1.7}$ (for both Z = Si and Sn). X-ray diffraction showed the presence of a disordered bcc phase with no indication of an ordered $D0_3$ phase. However, the latter is difficult to observe with X-ray diffraction because of the low intensity of the fcc superlattice peaks. A bimodal Fe hyperfine field distribution as obtained from Mössbauer effect spectra indicated the presence of two discrete Fe environments. The results suggested a lower degree of Ga clustering than has been previously observed in Fe–Ga alloys, of similar composition, prepared by melt spinning. The microstructure is similar to that of Fe–Ga thin films prepared by combinatorial sputtering. Some samples have also been studied after annealing at 800 °C for 8 h. No changes were observed in X-ray diffraction patterns after annealing. However, Mössbauer effect studies show the formation of $D0_3$ and $L1_2$ order in annealed samples analogous to the phases observed in melt spun ribbons of similar composition.

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1. Introduction

The addition of Ga to Fe has been shown to substantially increase its magnetostriction [1–6]. In particular, alloys of the form $Fe_{100-x}Ga_x$ with x around 19 have attracted interest because of their large magnetostriction and have been considered as possible candidates in place of the more traditional rare-earth-based magnetostrictive alloys for applications as sensor or actuator materials. It is known that there is a close correlation between the structure of these alloys and their resulting magnetoelastic properties. In particular, it is commonly believed that Ga–Ga pairing in alloys near $x = 19$ is responsible for an increase in magnetostriction and that the formation of an ordered $D0_3$ phase as the alloy composition approaches $x = 25$ is detrimental to this property [7]. Recent reports have shown that the microstructure of these alloys may be influenced by processing conditions and that rapid quenching from the melt [8–10] and combinatorial sputtering [11] methods may provide a means of controlling the microstructure and hence the magnetostriction.

Mechanical alloying is a non-equilibrium processing technique that is known to influence microstructure and is a common method of preparing metastable phases and alloys with extended solubility limits [12]. This technique is

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cost-effective and yields powders that may be suitable for a number of possible applications, such as preparation of magnetostrictive alloy–polymer matrix composites. We have recently reported a preliminary study of mechanically alloyed Fe–Ga alloys [13], which to our knowledge represents the first investigation of Fe–Ga alloys prepared by this method. In the current work we present a considerably more detailed investigation of the preparation and properties of mechanically alloyed Fe–Ga. The present study includes detailed X-ray diffraction, Mössbauer effect spectroscopy and magnetization investigations of both as milled and annealed samples. Results for Fe–Ga with Si and Sn additions are also reported. These studies allow for a careful comparison of the properties of mechanically alloyed Fe–Ga with those for alloys prepared by rapid solidification from the melt [10] and sputtering [11].

2. Experimental methods

Alloys of Fe–Ga with starting compositions of 17, 19, 21, 23, and 25 at% Ga and $Fe_{81}Ga_{17}Z_2$ (Z = Si, Sn) have been prepared by mechanical alloying high-purity iron powder and gallium metal under an argon atmosphere. Milling was performed in a hardened steel vial using a SPEX Model 8000 Mixer/Mill. Two 7/16 in diameter hardened steel balls were used to mill approximately 3 g of sample. This gave a ball to sample weight ratio of about 4:1 which has been proven to be effective in other alloy systems [12]. To determine the effectiveness of the milling process, a sample of $Fe_{81}Ga_{19}$ (starting composition) has been investigated at various stages of milling. Subsequent samples have been prepared by milling for a period of 12 h. Some samples have also been investigated after annealing at 800 °C for 8 h under an argon atmosphere.

Compositional analysis was performed with a JEOL 8200 microprobe. X-ray diffraction patterns were obtained at room temperature for all samples using Cu Kα radiation with a Siemens D-500 scanning diffractometer. Room temperature ^{57}Fe Mössbauer effect spectra were obtained for all samples using a Wissel System I constant acceleration spectrometer and a Rh^{57}Co source. The velocity scale for all spectra was referenced to room temperature α-Fe. The intrinsic spectrometer linewidth was about 0.13 mm/s (half-width at half-maximum, HWHM). Spectra were analyzed using Voigt distributed Lorentzian sextets [14,15] which has been shown to be appropriate for disordered bcc Fe alloys [10,11]. Magnetization measurements were obtained at room temperature using a PAR 155 vibrating sample magnetometer (VSM), calibrated to room temperature Ni (54.43 emu/g).

3. Results and discussion

3.1. Properties of Fe–Ga as a function of milling time

X-ray diffraction data for the samples with a starting composition of $Fe_{81}Ga_{19}$ are shown in Fig. 1. All X-ray patterns showed peaks that correspond to the reflections associated with bcc Fe. In addition, the pattern for the sample milled for the shortest period also showed the presence of several small peaks that could correspond to elemental gallium near 34° and 41°. This is shown in the inset of Fig. 1, which is an expanded view of the section of the scan from 25° to 43°. For milling times of 6 h or greater, the X-ray patterns indicated the presence of a single-phase bcc structure. This suggests that a minimum of 6 h of milling time is required to homogenize the powder.

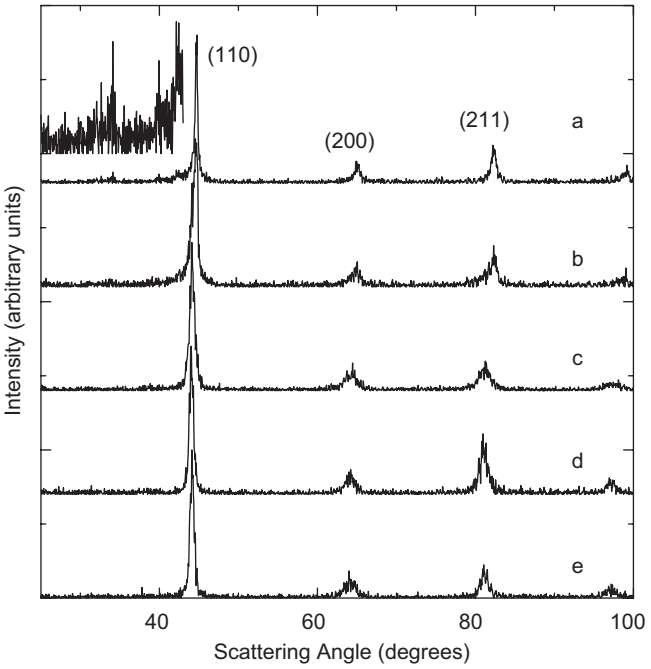

Fig. 1. Room temperature Cu Kα X-ray diffraction patterns of a sample with a starting composition of $Fe_{81}Ga_{19}$ milled for (a) 1.5 h, (b) 3.0 h, (c) 6.0 h, (d) 12.0 h and (e) 24.0 h. Miller indices correspond to the bcc phase. The inset is an expanded view of (a) from 25° to 43° in scattering angle.

Room temperature ^{57}Fe Mössbauer effect spectra for the $Fe_{81}Ga_{19}$ sample (starting composition) as a function of milling time are illustrated in Fig. 2. In the spectrum for the sample milled for the shortest period of time (1.5 h), a hyperfine split magnetic component is seen as well as a central unsplit component. The spectrum was, therefore, fit to a combination of distributions of sextets for the magnetic sites and a singlet for the central component. The outer peaks of the sextet are seen to be slightly asymmetric indicating the presence of at least two reasonably distinct Fe environments. As a result, the magnetically split component of the spectrum was fit to a Voigt-based distribution consisting of two Gaussians. For samples milled for 3 or 6 h there is evidence for the central singlet in the Mössbauer spectra and these were fit as for the 1.5 h sample. For the 12 and 24 h milled samples there is no evidence for the central component and its inclusion did not improve the validity of the fits. Fitted parameters for the spectra shown in Fig. 2 are given in Table 1.