CRYSTALLIZATION OF AMORPHOUS (Fe1-xCox)76Mo8Cu1B15 ALLOY

T. Hatala, M. Miglierini

Slovak University of Technology, Ilkovičova 3, 812 19 Bratislava, Slovakia E-mail: tomas.hatala@stuba.sk

Received 30 April 2011; accepted 29 May 2011.

1. Introduction

NANOPERM-type nanocrystalline alloys based on Fe-M-B-Cu where M= Zr, Nb, Hf have been proposed by Suzuki [1]. That alloys show small magnetostrictive coefficients and concomitant large permeabilities. Substitution of Co atoms at the expense of Fe in the (Fe,Co)-M-B-Cu-type alloys called HITPERM [2], show high permeabilities and high Curie temperatures. The aim of this work is to study the effect of Co substitution on crystallization behaviour of amorphous (Fe_{1-x}Co_x)₇₆Mo₈Cu₁B₁₅ alloy for x = 0, 0.25, 0.5. Changes introduced by modification of composition are investigated by ⁵⁷Fe Mössbauer spectra taken at room temperature in transmission geometry. In addition, diffraction of synchrotron radiation (DSR) was used during *in situ* heat treatment to follow the onset and subsequent progress of the crystallization.

2. Experimental details

As-quenched alloys (Fe_{1-x}Co_x)₇₆Mo₈Cu₁B₁₅ with various amounts of Co (x = 0, 0.25, 0.5) were prepared at the Institute of Physics SAS by the method of planar-flow casting in a form of ribbons about 10 mm wide and 20 μ m thick. Diffraction patterns were acquired by 7 keV monochromatized synchrotron radiation. They were recorded every 10 s from both sides of the ribbon shaped samples, during *in situ* heat treatment using a 2D detector. Originally amorphous precursors were exposed to continuous temperature change from room temperature up to 800 °C with a ramp of 10 K/min. ⁵⁷Fe Mössbauer spectra were obtained at room temperature in transmission geometry using ⁵⁷Co(Rh) source.

3. Results

Contour plot of diffractograms obtained from the wheel side of the $(Fe_{0.5}Co_{0.5})_{76}Mo_8Cu_1B_{15}$ alloy in Fig. 1(a) demonstrate how the crystallization temperatures were determined. The first T_{x1} and the second T_{x2} crystallization points are characterized by the occurrence of bcc-Fe(Co) phase and other crystalline phases, respectively. An influence

of Co-content x on T_{x1} and T_{x2} is plotted in Fig. 1(b). A strong effect of Co substitution is seen by a dramatic decrease of both crystallization temperatures. At the same time, differences between the air and the wheel side of ribbon shaped samples are revealed. Fig. 2(a) shows the evolution of nanocrystalline fraction of the bcc Fe(Co) phase. The values of relative crystalline content were obtained diffraction patterns of synchrotron radiation by their deconvolution into Lorentz and Gaussian line shapes. They represented relative fraction of nanocrystallites and residual amorphous matrix, respectively. From Fig. 1(b) and 2(a) we observe an earlier onset of on the wheel side of the ribbon (note: wheel side of the ribbon was in contact with quenching wheel). Lattice parameters were obtained from a position of the maximum of the Lorentzian line shape. Fig. 2(b) shows the evolution of lattice parameters on temperature and Co content. Substitution of Co causes a decrease of the lattice parameter with a gain of Co content x. For x=0.5, the smallest value of lattice parameter of 0.2876 nm is observed for the bcc Fe(Co) lattice with a optimum configuration. The lattice parameter values for x = 0 alloy, i.e. without Co, are quite high as compared with 0.2866 nm of pure bcc-Fe at room temperature. This is most probably by incorporation of some Mo atoms into the bcc-Fe lattice.



Fig. 1: Contour plot of the (110) reflection acquired from the wheel side of the x = 0.5 alloy with indicated temperatures of the first $T_{x1} = 375$ °C and the second $T_{x2} = 575$ °C crystallization (a) and Co-content dependence of T_{x1} and T_{x2} (b).

The initial decrease in the lattice parameter values with temperature for all samples is caused by evolution of the respective crystalline phase while the crystalline lattices evolve and start to acquire their optimum structural arrangement. At high enough temperatures, rather rapid increase in the lattice parameters is associated with thermal expansion of the lattices. Mössbauer spectrometry was performed at room temperature upon samples in the asquenched state and on samples annealed at selected temperatures, i.e. nanocrystalline samples. Transmission experiments provide information on the whole bulk of the samples.



Fig. 2: Evolution of the relative crystalline content (a) and lattice parameter (b) of the bcc *Fe*(Co) phase plotted against temperature of annealing.

The as-quenched alloys are fully amorphous and characterised by broad sextuplets with distributed hyperfine fields. After annealing at temperatures higher than T_{xl} , bcc-Fe(Co) crystalline phase was formed. As an example, Mössbauer spectrum of (Fe_{0.5}Co_{0.5})₇₆Mo₈Cu₁B₁₅ alloy heat treated at 435 °C for 1 hour is presented in Fig. 3(a). The spectrum consists of relatively narrow lines that are superimposed upon broadened feature. The former represent nanocrystalline phase whereas the latter is assigned to the residual amorphous matrix. Crystalline component was reconstructed by five narrow sextets with unique values of hyperfine fields. They are marked with thick vertical lines in Fig. 3(b).



Fig. 3: Mössbauer spectrum of the x = 0.5 alloy annealed at 435 °C (a) and corresponding distribution of hyperfine fields P(B) (b).

Broad spectral component of the amorphous rest was refined by distribution of hyperfine fields P(B) which is also plotted in Fig. 3(b). P(B) distribution exhibits notable asymmetry with peaks positioned at around 15 and 27 T. They indicate a presence of magnetically different regions of the resonant Fe atoms within the residual amorphous matrix. Relative fractions of the individual sextets which have been used to interpret the nanocrystalline bcc-

Fe(Co) phase, were first of all calculated. According to the theoretical predictions, a binomial distribution describing different number of Co atoms located at the nearest neighbour (nn) positions of the resonant Fe atoms was applied. Taking into consideration statistical significance (> 1%), the number of fitted spectral components was determined. In the case of the Mössbauer spectrum from Fig. 3(a), 5 independent Lorentz sextets were used. Their relative areas were not restricted during the fitting process. Comparison of theoretically calculated values and resulting fit for the Mössbauer spectrum from Fig. 3(a) is presented in Fig. 4. A satisfactory correlation between calculated (theory) and refined (fit) relative fractions was achieved which indicates appropriateness of the applied fitting model.



Fig. 4: Relative fraction of individual sextets used to reconstruct the bcc-Fe(Co) phase.

4. Discussion

Using *in situ* diffraction of synchrotron radiation we were able to determine the crystallization temperatures of the investigated alloys. It was shown that the crystallization starts earlier on the wheel side of the Co-containing ribbons. For x = 0, no appreciable differences were observed between the wheel and the air sides. The crystallization temperatures depend also on the Co contents, they decrease with increasing amount of Co. From the temperature evolution of lattice parameter, we can follow a tendency towards better structural arrangement of the crystalline lattice up to certain temperature where both the amount of nanocrystallites as well as their size are optimal. At higher temperatures of annealing, however a thermally induced lattice expansion is observed through an increase of the lattice parameter. Using Mössbauer spectrometry we were able to describe relative fractions of Co nearest neighbours of Fe resonant atoms in the bcc-Fe(Co) crystalline lattice. At the same time, their hyperfine fields were also determined.

Acknowledgement

This work was supported by the grants VEGA 1/0033/10, and SK-PL-0013-09.

References:

- [1] K. Suzuki et al.: Mat. Trans. JIM, 31, 743 (1990).
- [2] M. A. Willard et al.: J. Appl. Phys., 84, 6773 (1998).