acta physica slovaca vol. 49 No. 3, 429 – 432 June 1999

STRUCTURE AND MAGNETIC PROPERTIES OF MECHANICALLY ALLOYED Fe-Si SYSTEM*

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Received 9 October 1998, accepted 6 November 1998

Mechanical alloying of elemental powder was used to prepare Fe_3Si alloys. Changes in structure, phase transitions and magnetic properties induced by mechanical alloying and subsequent heat treatment of Fe-Si powdered system using XRD, Mössbauer spectroscopy, DSC and magnetic measurements were studied. The development of the atomic ordering at heating is presented.

PACS: 61.10, 75.30, 76.80

1 Introduction

Mechanical alloying (MA) by high energy ball milling is a solid state reaction method for synthesizing a wide range of materials with novel compositions, crystal structures and microstructures, leading to excellent properties. MA technique has been also successfully applied to the Fe-Si system, for which the formation of various types of structures, depending on composition and milling conditions, has been reported [1-4].

In previous papers [5] we have showen that mechanical alloying results in a transition of the Fe-Si mixture into a metastable state characterized by large changes in bcc-Fe lattice accompanied by partial formation of disordered bcc solid solutions.The present work is devoted to the investigation of changes taking place during the evolution of the mechanically alloyed structure on heating.

2 Experimental

For the structural evolution study at heating there were selected unmilled starting Fe-Si mixture (3:1 in atomic proportion) and two samples with various structural and compositional metastability degree created during deformation by mechanical alloying in a planetary high-energy ball-mill

*Presented at 10th Czech and Slovak Conference on Magnetism, CSMAG'98, Košice, Slovakia, August 24-27, 1998

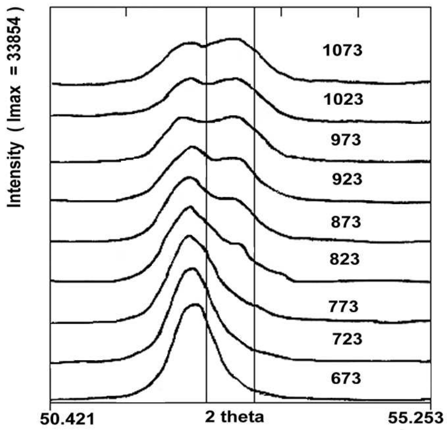

Fig.1 - Changes in the XRD line profile of the Fe(110) peak of the ball milled sample for 2 min at heating.

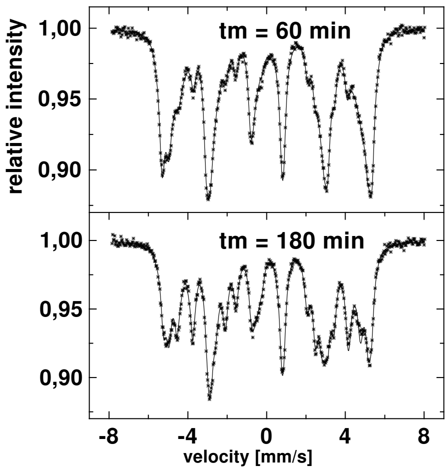

Fig.2 - The Mössbauer spectra of samples alloyed at different time of milling (t_m) and consequently annealed for 1 hour at 873 K

in a stationary argon atmosphere using steel vial and balls. The selected samples were annealed at different temperatures (673, 873 and 1073 K) in an inert atmosphere for 1 hour.

The room temperature Mössbauer spectra of annealed samples were taken in a transmission geometry using a ^{57}Co γ-source in an Rh matrix on a constant acceleration spectrometer calibrated by α-Fe. In situ X-ray diffraction analysis (CoK_α radiation) was performed at a heating rate of 2 K/min over the temperature range 300-1223 K in a vacuum of 10^{-2} Pa.

Thermomagnetic curves in the temperature range from 293 to 1073 K were measured with the vibrating sample magnetometer in a vacuum furnace with heating rate of 4 K/s in the magnetic field of 50 Oe. Hysteresis loops were measured before and after heating. The DSC curves were measured in argon in the temperature range 323 - 893 K at a rate of 0.67 K/s.

3 Results and discussion

The structural response of the starting and mechanically alloyed Fe-Si mixtures to the changes at heating is accompanied by two phase splitting of diffraction lines corres- ponding to the α-Fe and by the appearance of the new crystalline line of Fe_3Si on the diffraction patterns (Fig.1). The nature of changes in mechanically alloyed samples taking place during the heating depends on the disordering degree and phase composition. High temperature XRD analysis revealed that subsequent thermal treatment of disordered nanocrystalline samples leads to crystallization at temperatures, which are about 100-200 K lower than those at which the crystalline Fe_3Si is formed by annealing of powder prepared by spark erosion or sputtering. Annealing of the mechanically alloyed powders at 1073 K leads to the thermodynamically stable ordered DO_3 structures.

The Mössbauer spectra of milled and annealed samples were fitted by a set of undistributed Lorentz sextets corresponding to various crystal phases in grains, and one distributed sextet correspoding to grain boundaries. Mössbauer phase analysis has shown that the relative amounts of intergranular phase and the crystal phases with various population of silicon atoms in the nearest

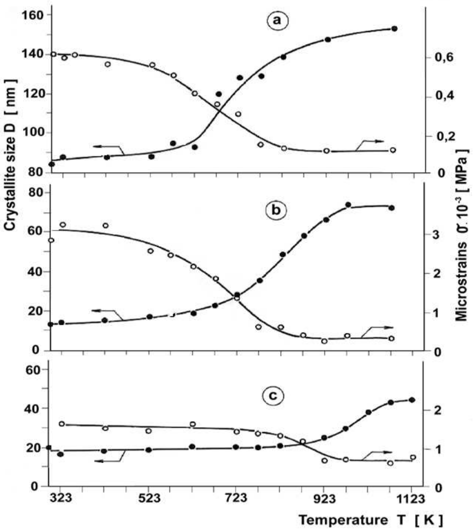

Fig.3 - Temperature dependence of crystallite size and microstrains for starting mixture (a), 60 minutes alloyed (b), and 180 minutes alloyed (c), samples

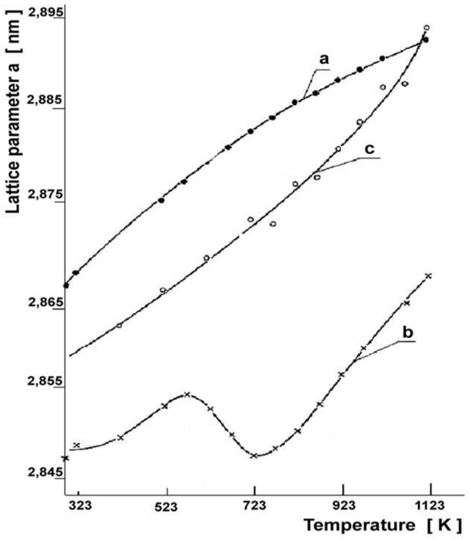

Fig.4 - Temperature dependence of lattice parameter of starting mixture (a), 60 minutes alloyed (b), and 180 minutes alloyed (c), samples

neighbourhood of iron atoms of annealed samples depend upon the conversion degree of disordered solid solution during mechanical alloying (Fig.2).

We can see the significant changes in crystallite size and microstrains in the temperature range 623 - 973 K in Fig.3. These results are in a good agreement with the results in [6]. The process of structural relaxation is accompanied by a microstrain decrease. Grain growth and ordering occur together because they both depend on diffusivity: ordering depends on bulk diffusion and crystallite growth should be dependent more on grain boundary diffusion [7]. It is evident that ordering results in a decrease in lattice microstrains in the investigated temperature region and can be attributed to lattice expansion represented by a sharp increase of the lattice parameters (see Fig.4). The temperature dependence of the lattice parameter for 60 min alloyed sample, however, has an extreme character which is in a good agreement with DSC effects corresponding to disorder-order phase transitions (defect annihilation, microstrain relaxation and crystallization processes). Structural changes occuring at heating lead to the decrease in coercivity (see Fig.5).

Thermomagnetic curves (Fig.6) reveal high homogeneity of alloyed powders. Curie temperatures of Fe_3Si at 823 K and α-Fe at 1040 K can be identified on the curves. In case of the sample alloyed for 60 min, the relative content of α-Fe is of the order of 10^{-2} only, therefore the critical temperature of α-Fe was not observed. Traces of other transformations were found in the temperature range between 573 - 723 K on branches for increasing temperature, which can be ascribed to the disorder-order phase transitions.

Summary of the results: Using MA by high-energy ball-milling the Fe_3Si alloys were prepared by disordered bcc solid solutions. During heating in the temperature range 323-1123 K crystallite size growth, lattice microstrains relaxing and disorder - order transformations were observed. The nature of structural changes in mechanically alloyed samples taking place during

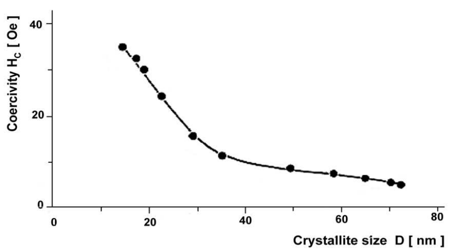

Fig.5 - Crystallite size dependence of coercivity for 60 min alloyed sample

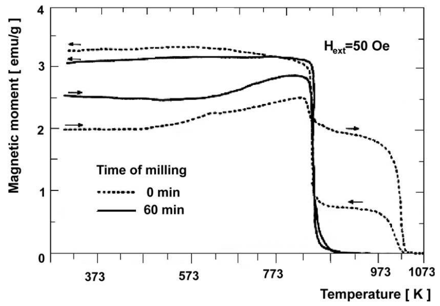

Fig.6 - Thermomagnetic curves of starting mixture and 60 min alloyed samples

heating depends on the disordering degree and the phase composition. Mössbauer spectroscopy and thermomagnetic measurements showed that annealing at 1073 K leads to the thermodynamically stable ordered structures corresponding to the nominal stoichiometry.

Acknowledgement This work was supported by the Slovak Grant Agency for Science - VEGA, Grants 2/4179/97 and 1/4252/97.

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