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Crystallization of FeSiB Amorphous Ribbons Induced by Laser Interference Irradiation

J. Kusinski [a], *, O. Czyż [a], A. Radziszewska [a], J. Morgiel [b], R. Ostrowski [c], M. Strzelec [c], K. Czyż [c], A. Rycyk [c]

[a] Faculty of Metal Engineering and Industrial Computer Science, AGH University of Science and Technology,
30 Mickiewicza Ave., 30-059 Cracow, Poland,
[b] Institute of Metallurgy and Materials Science, Polish Academy of Sciences, 25 Reymont Str., 30-059 Cracow, Poland
[c] Institut of Optoelectronics, Military University of Technology in Warsaw,
2 Gen. S. Kaliskiego Str., 00-908 Warsaw, Poland
* Corresponding author. E-mail address: kusinski@agh.edu.pl

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Abstract

Detailed studies on the effects of pulsed laser interference heating on surface characteristics and subsurface microstructure of amorphous $Fe_{80}Si_{11}B_9$ alloy are reported. Laser interference heating, with relatively low pulsed laser energy (90 and 120 mJ), but with a variable number (from 50-500) of consecutive laser pulses permitted to get energy accumulation in heated areas. Such treatment allowed to form two-Dimensional micro-islands of laser-affected material periodically distributed in amorphous matrix. The crystallization process of amorphous FeSiB ribbons was studied by means of scanning and transmission electron microscopy. Detailed microstructural examination showed that the use of laser beam, resulted in development of nanostructure in the heated areas of the amorphous ribbon. The generation of nanocrystalline seed islands created by pulsed laser interference was observed. This key result may evidently give new knowledge concerning the differences in microstructure formed during the conventional and lased induced crystallization the amorphous alloys. Further experiments are needed to clarify the effect of pulsed laser interference crystallization on magnetic properties of these alloys.

Keywords: Laser interference heating, FeSiB, Amorphous alloy, SEM, TEM

1. Introduction

In recent years different approaches have demonstrated the use of pulsed laser sources for the periodic modification of material surface layers, such as melting, phase transformations, hardening, oxidation, thin film deposition, micro-structuring, etc. [1-4]. In many cases, a suitable heat treatment, which leads to partially nanocrystalline microstructure, may provide an improvement in specific properties. One of the advantages of employing short pulsed lasers is that they allow the modification of the surface morphology and/or structure without significant damages or alterations to the underlying material. Metallic glasses are not thermodynamically stable, and they tend to structurally relax and finally crystallize upon appropriate heat treatment, which leads to the transition from the metastable state to the equilibrium one, resulting in the formation of crystalline phases. In the past 3 decades, there has been increasing interest and corresponding significant research activity in the area of soft magnetic materials. The amorphous FeSiB(X) alloys, have been in the past extensively studied [5, 6], due to their excellent soft magnetic properties. Metallic glasses based on FeSiB alloys are soft magnetic materials characterized by high permeability and low energy loss. It has been established that proper annealing treatment improves the permeability and coercivity of the rapidly quenched amorphous ribbons [5-7]. The crystallization of amorphous alloys based on the laser interference heating is among the most practical and elegant approaches to the formation of extended periodic micro- and nano-structures [8]. The laser interference heating of amorphous alloys may produce complex composites consisting of nanocrystals embedded in an amorphous interacting matrix. Thus, understanding the crystallization (nanocrystallization) process during the laser interference heating is of both scientific and technological importance.

In this paper, studies of the crystallization process fabricated by the laser interference heating of FeSiB ribbons are reported.

2. Experimental procedure

In the present study, we have used FeSiB amorphous alloy in the form of 30 μm thick, 25 mm wide ribbons. The $Fe_{80}Si_{11}B_9$ alloy was applied by Allied Signal Corp. of Morristown, New Jersey. The pulsed laser interference heating (PLIH) was carried out using a Q- switched Nd: YAG (operating in the TEM_{00} mode), with laser wavelength λ of 1064 nm, a frequency of 10 Hz, a pulse time τ of 10 ns and laser energy, F, 90 and 120 mJ and variable number (from 50-500) of consecutive laser pulses. Two-dimensional, interference illumination field was realized using quartz tetrahedral prism with an apex angle of 172^0. The surface morphology of etched (in Cr_2O_3 water solution at 8V) ribbons was examined using scanning electron microscopy (SEM). Finally, TEM lamellas were prepared: perpendicular to the heated surface with the aid of a focused ion beam (FIB) Tecnai G2 F20 and parallel to the heated surface using dual beam (PIPS) Gatan system for milling of 3 mm discs cut from laser interference heated areas. TEM examinations of the structured samples were carried out using a JEOL JEM 200 CX and Tecnai G2 F20.

3. Results and discussion

The effect of pulsed laser interference heating is demonstrated clearly in a two sets of SEM images, Fig. 1a-d, where we have processed the amorphous FeSiB alloy ribbon with energy 120 mJ, by using (a, b) - 50 and (c, d) - 500 consecutive laser pulses. The evolution of the laser heated dotes in amorphous FeSiB ribbon, with increasing number of laser shots is well visible. The SEM analysis of the locally heated ribbon showed a uniform periodic distribution of irradiated dotes (Fig.1a, c). It should be pointed out that the laser beam has a Gaussian distribution of intensity which results in inhomogeneous heating across the laser beam diameter.

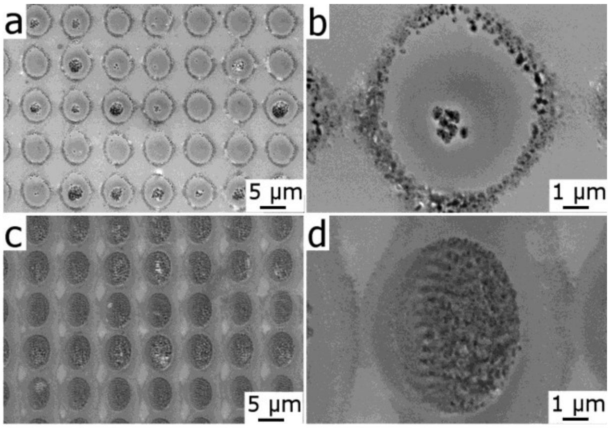

Fig. 1. SEM images of etched samples show periodic dot-like patterns fabricated on FeSiB ribbon surface by 120 mJ laser interference irradiation with 50 (a, b) and 300 (c, d) consecutive laser pulses; b, d are magnified images of a, c

The laser energy density decreases exponentially in the radial direction and has a maximum value at the center. As the center of the irradiated dotes is at the highest temperature, indeed is much more deeply melted than the other regions (Figs 1b, d). This probably arises from local differences in absorption of the laser radiation. When the number of laser shots increases effect of absorption is not so important, and the ripple pattern is evidently observed after irradiation with 500 subsequent laser shots (Figs 1c, d). The ripples originates from the interference between the incident laser light and the scattered tangential wave [9]. It is clearly seen that with 50 pulses, with a pulse energy of 120 mJ, exceeding the ablation threshold were found to leave smooth craters on the surfaces, exhibiting a characteristic rim marking the ablated area (Fig. 1a, b). With higher number (over 100) of consecutive laser pulses periodic surface structure was observed (Fig. 1c, d), with spatial periods close to the free space wavelength of the laser excitation pulse (~1μm). The large temperature gradients achieved with localized laser heating can lead to rapid self-quenching of the material, trapping in highly non-equilibrium structures. Also, the rapid generation of large temperature gradients can induce thermal stresses and thermo-elastic excitation of acoustic waves [10]. Un-melted regions, laying about 5 μm from center of melted zone, experienced a gradual decrease in the temperature value what finally resulted in partial crystallization of amorphous substrate. Non-processed material between the irradiated spots (white-etched zones in Fig. 1c) remained in amorphous state. The effect of the laser interference heating on the microstructure evolution have been investigated by means of TEM analysis. In Fig. 2a TEM image of laser-irradiated area is presented indicating the evolution structure, from nano-crystalline in the center (zone 1), through large-grains structure in the rim surrounding melted zone (zone 2) and again nano-crystalline in the peripheral of laser processed area. In the central area of laser-irradiated spot grain size varies form ~ 60 nm to 500 nm, while in the rim (in zone 2) one may observe large grains of ~ 1,5 μm. Observed abnormal grain growth in this area is due to intensive material heating caused by heat accumulation during consecutive 300 laser pulses. Microstructure in the peripheral of laser processed area is presented in Fig. 2c. It is apparent from Fig. 2c that amorphous material crystallizes in the form of columns composed of nanocrystals.

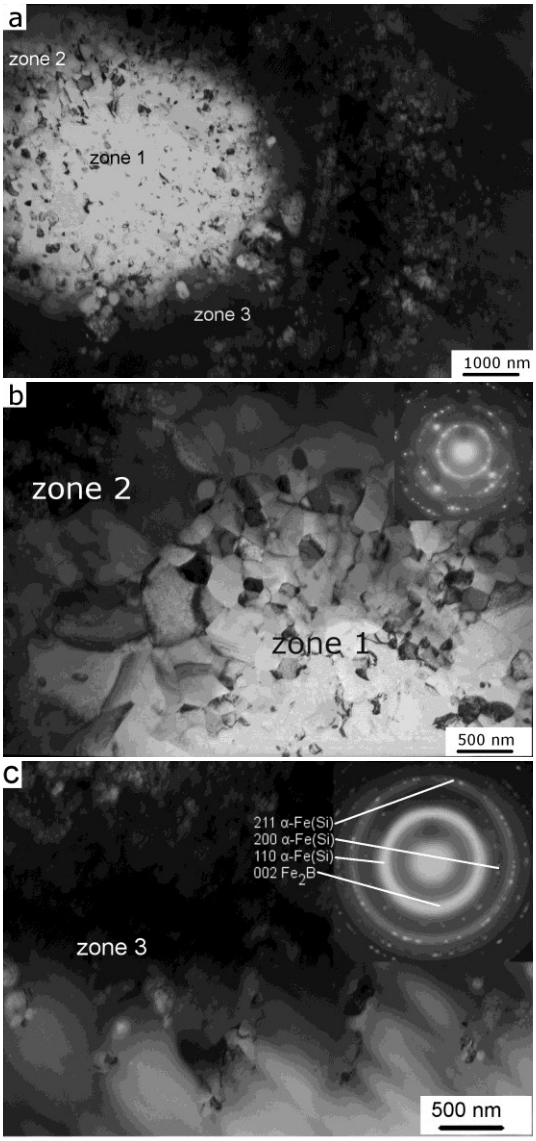

Fig. 2. TEM bright-field images of laser-processed sample (120 mJ and 300 pulses); (a) image of laser-irradiated spot with marked three zones of different grain size, (b) magnified image of zones 1 and 2, (c) magnified image of the peripheral zone 3

Distance between such columns is ~ 1 μm, which is the same as a distance between the ripples observed around the strong ablated zone (in Fig. 1d). It can be stated that the laser heated zone spreads probably more into amorphous material along the ripples originated from the interference between the incident laser light and the scattered tangential wave. An electron diffraction pattern from the nano-crystalline phase is shown in Fig. 2c and this ring pattern can be consistently indexed as containing rings representing: amorphous matrix, α-Fe(Si) and Fe_2B phases.

Cross-sectional investigations were carried out by milling the sample surface with the focused ion beam after Pt deposition to protect the sample. Representative TEM images, shown in Fig. 3a and b, of sample cut perpendicular to the heated surface with the aid of a focused ion beam (FIB) confirmed presence of the nano-size grains in the laser irradiated spot.

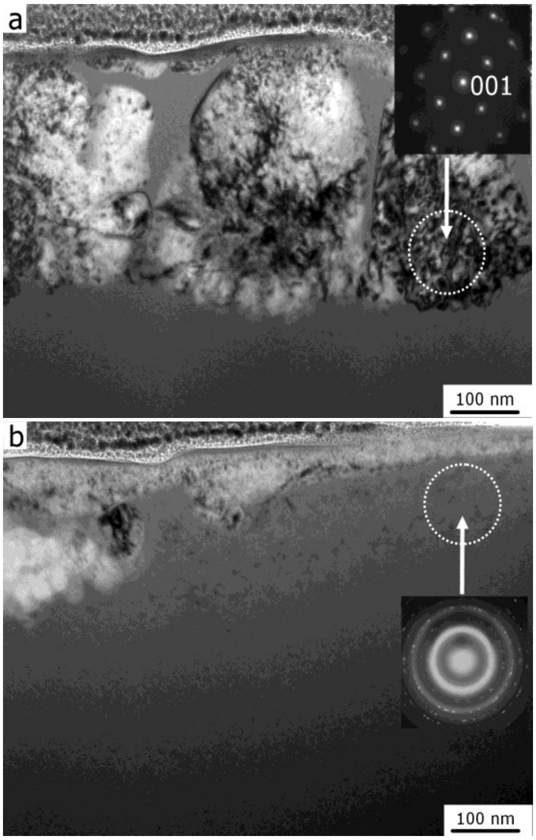

Fig. 3. TEM bright field images showing microstructure of the laser irradiated area in the plane perpendicular to the heated surface; a- in the irradiated-spot center, b - in the peripheral

The SAD pattern presented in Fig. 3a shows typical [001] oriented larger grain of the α-Fe(Si). Microstructure in the peripheral of laser irradiated micro-area is presented in Fig. 3b. The thermal shock that accompanied the pulsed laser heating caused the appearance of many crystalline nuclei in the amorphous matrix, beneath the heated surface. When the crystallization degree increases, the number of spots in the rings, which represent nanocrystalline phases, also increases. Characteristic ring diffraction pattern from partially nanocrystallized amorphous phase, is observed and shown in the inset, in Fig. 3b. Indeed, heating of the amorphous material with several consecutive laser pulses produced a fine nanocrystalline microstructure.

In term to show the advantage of using laser interference irradiation on crystallization of amorphous FeSiB alloy microstructure of conventionally heated samples was also examined. Fig. 4a shows the SEM microstructure of the FeSiB alloy obtained after heating 3 mm discs to 600^{o}C with the rate of 20^{o}C/min and cooling down rapidly in an argon atmosphere with flow rate 20 ml/min to the room temperature. After such treatment, instead of the equiaxed grain morphology, characteristic for the crystallization induced by pulsed laser-irradiation the dendritic morphology, with varied dendrite size, from 100 - over 1000 nm, was observed. This was confirmed also by TEM examinations. Fig. 4b shows the bright field image of the sample heated up to 600°C.

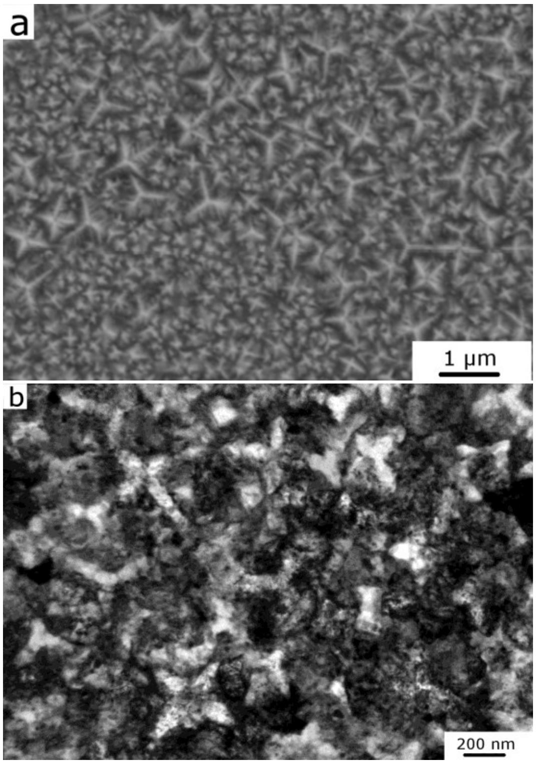

Fig. 4. SEM (a) and TEM (b) images show dendritic structure of FeSiB alloy after 30 min heating to 600°C

4. Summary

The effects of laser interference irradiation on surface characteristics and subsurface microstructure of an amorphous FeSiB alloy were experimentally studied. This result indicates that PLIH involves significant substrate heating and structural transformations in the subsurface layer up to ~300 µm. The advantages of laser interference heating for amorphous materials annealing consist in the heat up only the selected mico-area of the material in a very short time. The microstructural changes of the amorphous material after laser treatment depend on the temperature obtained during laser heating, in studied case of laser pulse energy 90 or 120 mJ and number of consecutive pulses (varied from 50 to 500). With increasing annealing temperature (starting from unheated amorphous and approaching the center of the laser heated micro-area) the following stages of crystallization of amorphous material were observed: ordering, nucleation of nanocrystalline phases, nuclei growth and disappearance of the metastable glassy structure and finally formation of grain boundaries between nano-crystallites. For the FeSiB alloy, TEM observations show the dendritic morphology after conventional heat treatment at 600°C. On the other hand, after interference laser heating instead of the dendritic morphology, an equiaxed morphology, was observed. Such morphology is characteristic only for the Cu or Cu and Nb doped crystallized FeCuSiB and FeCuNbSiB alloys. Indeed, the rapid heating and cooling rates involved in pulsed interference laser heating, permitted to saturate the amorphous FeSiB alloy with the randomly oriented, ultra-fine grains of FeSi dispersed in the remaining amorphous matrix.

Acknowledgments

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