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POSITRON ANNIHILATION STUDY OF STRUCTURAL RELAXATION AND CRYSTALLIZATION IN METALLIC GLASSES $Fe_{78-x}Co_xSi_9B_{13}$

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Results of measurements of positron lifetimes and X-ray diffraction in the annealed metallic glasses $Fe_{78-x}Co_xSi_9B_{13}$ ($x = 0, 20, 40, 60, 78$) before and after the process of crystallization are presented. From the results it follows that the annealing process below the crystallization temperature causes changes in concentrations of positron-trapping areas and can be consistently described as the result of two contributions: topological and chemical short range ordering. The process of crystallization causes precipitation of α-Fe and β-Co crystalline phases and also formation of microvoids in the investigated samples.

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1. Introduction

The isothermal annealing of metallic glasses, even at temperatures below the crystallization temperature, gives rise to changes in many physical properties. The process is caused by atomic rearrangements in the amorphous structure, and is generally called structural relaxation. In a series of previous investigations [1, 2, 3] it was shown that the property changes can be separated into contributions of two processes:

1. Chemical short range ordering (CSRO) describing reversible changes in the local surroundings of a given atom [4].

2. Topological short range ordering (TSRO) describing an irreversible decrease in the free volume [5].

The crystallization process is caused by the structural changes which result from annealing at sufficiently high temperatures. These changes cause the appearance of the long range ordering after crystallization. Hence, from the positron lifetime and X-ray diffraction investigations we can obtain information concerning isothermal annealing before and after crystallization.

2. Experimental

Measurements of positron lifetimes were carried out on an ORTEC spectrometer of resolution FWHM = 270 ps. A ^{22}Na isotope of 3.7×10^9 Bq activity was used as a positron source. The metallic glasses were made by the Institute of Materials Engineering of Warsaw Technical University. Each specimen consisted of ribbons 15 mm wide and 0.04 mm thick and was made up of a system layers of total size 15 mm × 15 mm × 0.25 mm. The source was situated between two identical investigated samples, forming a "sandwich" system. The samples of metallic glasses $Fe_{78-x}Co_xSi_9B_{13}$ (x = 0, 20, 40, 60, 78) were first preannealed for 10^5 s at 530 K and next the same specimens were further annealed at 578 K and 778 K for 10^5 s. The process of isothermal annealing was carried out in the atmosphere of gaseous argon. The X-ray diffraction measurements show that after preannealing at 530 K and 578 K the amorphous structure of investigated specimens does not change (Fig. 1a). However, 10^5 s annealing at 778 K of samples preannealed at 530 K and 578 K caused a distinct crystallization of all the investigated samples (Fig. 1b).

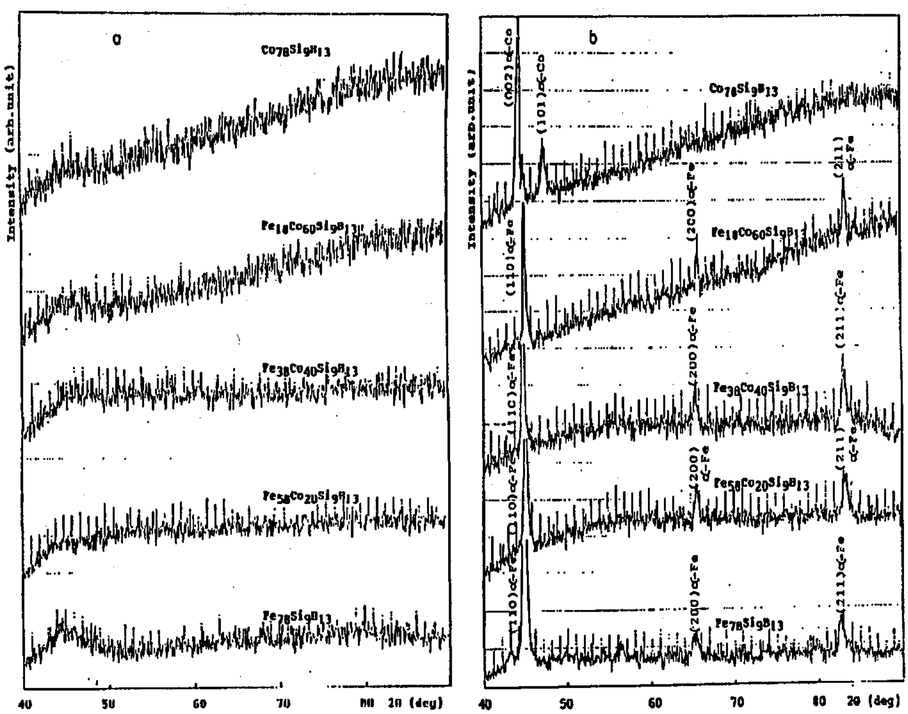

Fig. 1. X-ray patterns of investigated samples of metallic glasses $Fe_{78-x}Co_xSi_9B_{13}$ (x = 0, 20, 40, 60, 78); (a) — before crystallization; (b) — after crystallization.

3. Results and discussion

The mean values of positron lifetimes were calculated using a computer program POSITRONFIT which takes into account the positron source correction and the resolution function [6]. The calculated mean values of the positron lifetime in amorphous samples correspond to the annihilation of positrons trapped at the monovacancy-sized free volumes. The mean positron lifetime values τ for crystallized samples indicate the occurrence of microvoids (agglomerates of several vacancies) [7, 8, 9]. From such measurements we can obtain direct information concerning electron phenomena which proceed in interatomic structures of investigated specimens during isothermal annealing.

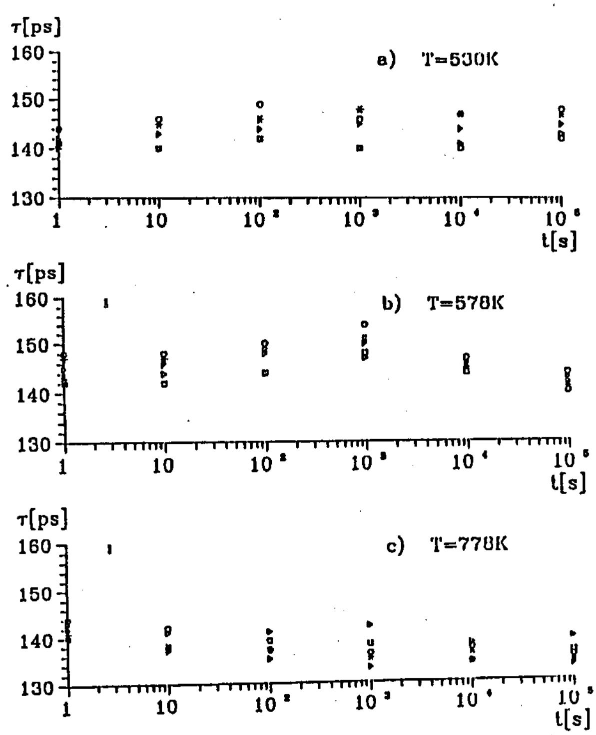

Fig. 2. The calculated values of positron lifetimes τ in metallic glasses $Fe_{78-x}Co_xSi_9B_{13}$ ($x = 0$, 20, 40, 60, 78); $Fe_{18}Co_{60}Si_9B_{13}$ — o o o; $Fe_{78}Si_9B_{13}$ — * * *; $Fe_{38}Co_{40}Si_9B_{13}$ — □ □ □; $Co_{78}Si_9B_{13}$ — full triangles; $Fe_{58}Co_{20}Si_9B_{13}$ — △ △ △, at different temperatures.

Figure 2a shows the calculated values (530 K) of positron lifetimes τ versus time for the 5 as-quenched investigated metallic glasses $Fe_{78-x}Co_xSi_9B_{13}$. In all cases the changes of positron lifetimes are insignificant and irregular, hence, it is difficult, at present stage of study,to explain these changes distinctly. However, it may be assumed that these changes are connected with positron trapping in microdefects formed due to the changes of the local surroundings of a given atom (CSRO).

The same specimens were further annealed at 578 K. The calculated lifetimes τ are plotted vs. time in Fig. 2b. In all cases the maximum was observed (at 10^3 s of annealing). It seems that the positron lifetime in positron trapping areas initially increases and next after annealing above 10^3 s decreases. This fact suggests that these phenomena are caused during the atomic rearrangements as a result of annealing at 578 K after preannealing at 530 K and they are connected with the changes of topological ordering of the amorphous structures (TSRO). Thus, it can be stated that in the investigated metallic glasses after annealing at 578 K one observes the increase in the positron trapping in microdefects concerned with escalation of the monovacancy-sized free volumes in the amorphous structures.

The calculated values of positron lifetimes after the process of crystallization vs. time are plotted in Fig. 2c. In all cases the distinct decrease in τ was observed. From this fact it is possible to conclude that the decrease of positron lifetimes in positron trapping areas after crystallization is connected with formation of microvoids (several vacancies sizes) in the investigated specimens. Simultaneously, from the X-ray diffraction measurements (Fig. 1b) it follows that the process of crystallization causes the appearance of α-Fe and β-Co crystalline phases.

Recapitulating, from the results of the present measurements of positron lifetimes and earlier reported changes of Young's modulus [1, 10] during structural relaxation in metallic glasses $Fe_{78-x}Co_xSi_9B_{13}$ ($x = 0$, 20, 40, 60, 78) it follows that the preannealing process at 530 K is connected with the chemical short range ordering CSRO and the next annealing process at 578 K is connected with topological short range ordering TSRO (positron trapping in the monovacancy-sized free volumes).

Simultaneously, the process of crystallization of metallic glasses $Fe_{78-x}Co_xSi_9B_{13}$ causes formation of microvoids in dense random packing structures and causes the precipitation of the α-Fe and β-Co crystalline phases.

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