INFLUENCE OF γ-IRRADIATION ON PROPERTIES OF AMORPHOUS $Fe_{78}B_{13}Si_9$ ALLOY STUDIED BY MÖSSBAUER SPECTROSCOPY AND POSITRON ANNIHILATION TECHNIQUE

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The influence of γ-irradiation on properties of METGLAS 2605S2 and FC3-1 $Fe_{78}B_{13}Si_9$ has been investigated by Mössbauer spectroscopy, positron annihilation technique as well as bending test. The ductility and orientation of magnetization were changed by γ-irradiation. They probably correlate with the change of microscopic free volume and release of stress in the ribbon, respectively.

I. INTRODUCTION

The magnetic metallic glasses produced by rapid quenching of a melt of iron-based transition-metal-metalloid systems have been studied extensively due to scientific and technological interest in recent years. The most studied metallic system is Fe_xB_{1-x}(0.72 x 0.86), but it has not satisfied the practical application requirements. After consistent efforts by numerous investigators it has been found that the inclusion of Si in Fe-B system improves the thermal stability as well as some magnetic properties /1/. For the nominal composition $Fe_{78}B_{13}Si_9$(METGLAS 2605S2) produced by Allied Corp.(USA) it has extremely low core loss as power transformer, and high ductility. The same composition $Fe_{78}B_{13}Si_9$ alloy (FC3-1) produced by Shanghai Iron and Steel Research Institute (SISRI) has slightly worse properties, in particular ductility. Therefore we tried to modify its properties using γ-irradiation, measuring them by Mössbauer spectroscopy, positron annihilation technique as well as macro-analysis such as brittlement and magnetic properties. A comparative study on 2605S2 has been carried out. There are a few papers describing the effect of nuetron irradiation on amorphous alloys /2,3/, but no one, as far as we know, has investigated γ-irradiation effects except on polymers, biomolecules and some glasses. Obviously the advatage of γ-irradiation is much lower damage than that caused by neutrons and even high energy electrons in the alloys, since the displacement and cascade collision events induced by γ-rays are quite small.

2. Experimental

Two sets of amorphous $Fe_{78}B_{13}Si_9$ ribbons in a continuous reel of 2.54 cm width and nominal 30 um thickness were taken from SISRI and the Allied Corp.(USA), respectively. These ribbons were cut into small pieces of suitable size for measurements of bending test, Mössbauer effect and positron annihilation. For studying the surface effect of the ribbons, specimens were subsequently prepared by chemical thinning in steps of about 2 μm. Wound ribbon reels were used for the measurements of magnetic properties. Irradiation was performed in the gamma facility of the Shanghai Institute of Nuclear Research using ^{60}Co gamma source. The as-quenched and annealed samples were irradiated simultaneously at a dose rate of of 10^7 R h^{-1} to a total dose from 4.5 x 10^7 to 7.5 x 10^8 R at 313 K. The isothermal annealing was performed in argon atmosphere at 650 K for 1.5 h after stress annealing in which the ribbons were quenched to room temperature after annealing at 693 K for 0.5 h. The annealing field of 60 Oe was applied in the plane of the ribbon, parallel to the long axis (x) of the ribbon. Annealing at the above field strength was sufficient to produce specimens having net anisotropy of less than one gauss /4/.

Mössbauer spectra were taken at room temperature using a constant acceleration spectrometer with a source of ^{57}Co in Pd. The spectrum was fitted to six Lorentz curves without any constraint. In addition, in order to understand the distribution of magnetization, several samples were measured in three different geometrical arrangements as shown in Fig.1. Mössbauer spectra were first taken with the ribbon plane perpendicular to the incident γ-ray ($\theta = 0$, $\psi = 0$), then other two sets of spectra were recorded, after rotation by $\theta = 30^{\circ}$ about the y axis and after subsequent rotation by $\psi = 90^{\circ}$ about the z axis.

Doppler broadening parameters S from positron annihilation lines were obtained at room temperature using a high purity Ge detector and a source of $^{22}NaCl$ between two identical thin mylar films. The measurements were only performed on as-quenched ribbons (FC3-1).

3. Results

Fig.2 shows the temperature dependence of the relative strain at fracture, ε_f for as-quenched ribbons of METGLAS 2605S2 alloy (1#, 3#) and FC3-1 (2#, 4#) with a dose of 5×10^8 R and without, respectively.

The isochronal annealing was performed in argon for 0.5 h. The curves indicate that the amorphous ribbons of FC3-1 become completely ductile, $\varepsilon_f = 1$ below 523 K after irradiation; only slight improvement is observed for the ribbons of 2605S2.

As is well known, the hyperfine field in the magnetic metallic glasses has a distribution due to random atomic arrangement. The Mössbauer spectra of annealed FC3-1 alloy are shown in Fig.3. after different doses. The spectrum appears a little asymmetric. The ratios of the intensities of the second (fifth) line to the third (fourth) line decrease with increasing doses. They correspond to the average magnetization lying at an angle of 31° out of the plane of the ribbon before irradiation, and 38° after an irradiation dose of 7.5×10^8 R. No significant effects were observed on the as-quenched FC3-1 alloy and both the annealed and as-quenched specimens of 2605S2 alloy.

The ratios between the intensities of the second and third lines for the 2605S2 alloy, as-quenched and annealed, are listed in table 1.

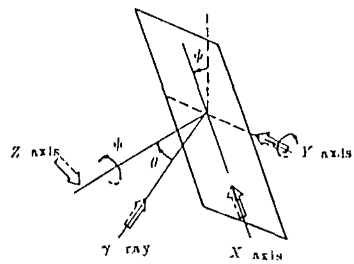

Fig.1. This figure shows the orientation of the specimen with respect to the incident γ-ray beam. θ is the angle through which the specimen is rotated about Y axis while ψ is the angle through which the plane is rotated about its perpendicular axis.

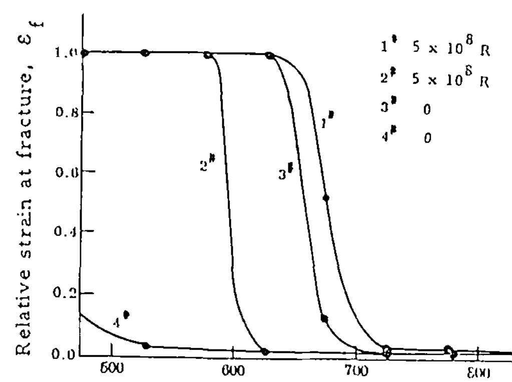

Fig.2. The temperature dependence of the relative strain at fracture, ε_f, for as-quenched METGLAS 2605 S2 (1#,3#) and FC-3-1 (2#, 4#) $Fe_{78}B_{13}Si_9$ ribbons without and with irradiation dose of 5×10^8 Rad. Annealing time 0.5 h.

The Doppler broadening parameter S for annealed speciments of FC3-1 decreases with increasing doses, and finally approaches the values obtained on ribbons of the 2605S2 alloy, which are the same with and without irradiation.

In addition, the macro--properties such as permeability and coercive force did not show big changes after irradiation for all samples.

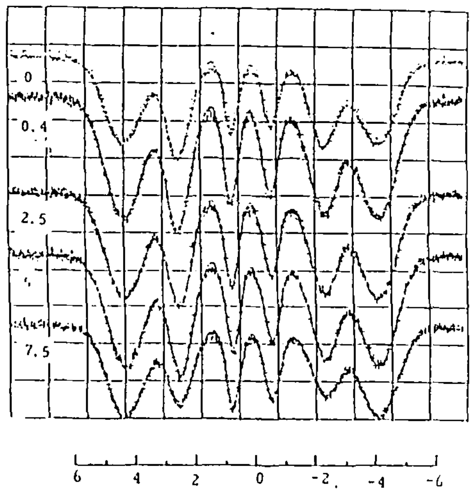

Fig. 3. Transmission Mossbauer spectra of FC3-1 $Fe_{78}B_{13}Si_9$ at various doses (10^8 R) after annealing at 650 K for 1.5 h.

4. Discussion

Gamma rays should be similar in behaviour to electrons, since the displacement damage produce by γ-rays from a ^{60}Co source with the energy of 1.17 and 1.33 MeV is caused predominantly by Compton electrons. But the probability of production of Compton electrons with an energy of 1 MeV is very small. Assuming the displacement energy 25 eV for (crystalline) iron metal, we estimated a displacement level of abour 10^{-7} dpa for a dose of 5 x 10^8 R. Therefore our results may not be explained by the production of vacancies and dislocations.

The improvement of ductility by γ-irradiation was probably caused by relaxation of atom positions leading to an increase of the microscopic free volume. The argument is that although the energy of most of the Compton electrons is not high enough to collide an atom out of its lattice site, it is sufficient to move an atom a little out from its initial position in the amorphous alloy. As a result of this process the state of atomic arrangement becomes more stable. This point of view confirms that the quenching process of the FC3-1 ribbon is not as good as that of 2605S2.

According to the model of Egami et al. /5/, there are positive and negative density fluctuations (p-and n-type defects) in the pre-quenched state, and these defects can annihilate mutually due to relaxation. Hence loss of microscopic free volume might occur both after thermal and irradiation relaxation. Probably it can be explained by this assumption that the Doppler-broadening parameter S decreases with increasing irradiation doses for annealed samples.

The rotation of the magnetization towards the normal of the ribbon plane during irradiation may be correlated with the release of stress in the ribbon plane. This effect can only play an important role when the stress is relatively weak. As is well known, strong stress remains in the as-quenched ribbon. That is why we did not see any significant change of the magnetization vector in as--quenched ribbons of the FC3-1 alloy. The results of etching experiments show that the out-of-plane declination of the magnetization changes from 26.4° to 16° after five etching steps. It means that the compressive stress from the surface is stronger than the tensile stress in the ribbon. This also helps us to understand the phenomenon of the orientation of magnetization. If the sample structure like 2605S2 alloy has good isotropy after annealing, it should show no effect.

From the ratios between intensities shown in table 1 the fractional volumes of domain magnetization in the direction of three main axes (Px, Py, Pz) can be obtained assuming that there exist only these three directions in the ribbon. We did find that there are three types of domains in the annealed 2605S2 ribbon,

Table 1. Ratios between the intensities of the second (fifth) and third (fourth) lines and fractional volumes of the domains for 2605S2 ribbons

State	θ(°)	ψ(°)	Ratio	P_z	P_x	P_y
Annealed	0	0	2.29	0.254		
	30	0	1.95		0.544	0.202
	30	90	2.43		0.547	0.199
As-quenched	0	0	3.17	0.073		
	30	0	2.59		0.543	0.384
	30	90	2.52		0.420	0.507

without any influence of irradiation, because the fraction were the same for different angles of measurement. In the as-quenched ribbon the symmetry of the magnetization distribution seems to be more complicated.

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