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# Surface smoothing of metallic glasses by swift heavy ion irradiation

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#### Abstract

Self supporting samples of Metglass 2204 ( $Ti_{50}Be_{40}Zr_{10}$ ) and substrate bound samples of Metglass 2705M ( $Co_{69}B_{12}Si_{12}Fe_4Mo_2Ni_1$ ) have been irradiated with 260 MeV <sup>107</sup>Ag and 130 MeV <sup>28</sup>Si ions, respectively at liquid nitrogen temperature. The surfaces have been examined by Scanning Electron Microscopy prior to and after irradiation. It is found that the surfaces have been smoothened after irradiation in both cases. The heights of the 'hills' have been decreased and the 'valleys' have been filled, without any detectable mass loss. The observed surface smoothing has been explained on the basis of shear flow mechanism within the framework of the viscoelastic model. © 1999 Elsevier Science B.V. All rights reserved.

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#### 1. Introduction

In a swift heavy ion irradiation experiment, most of the slowing down of the ions takes place due to electronic energy loss  $S_e$ . This process dominates for most of the projected range  $(R_p)$  of the ions in the target. A large number of near surface and bulk atoms of the target are set into motion by this electronic excitation, even at very low irradiation temperatures. Subsequently, the mobility set in the bulk results in atomic rearrangements. For thin amorphous targets, the occurrence of atomic rearrangements leads to some well known macroscopically observable effects, such as the increase in electrical resistivity and the ion beam induced plastic deformation [1–4].

Rearrangement of the near surface atoms of amorphous targets, as a result of fast heavy ion bombardment, leads to modifications of the irradiated surface without detectable mass loss. For a given value of  $S_e$ , these surface modifications depend upon two parameters: (i) the fluence of the ion beam  $\Phi t$  ( $\Phi$  being the flux of the beam and t is the total time of irradiation) and (ii) the angle of incidence  $\theta$  (also called the tilt angle) of the beam with respect to the z-axis (the axis perpendicular to the sample surface) [5,6]. These tilt angle and fluence dependent surface modifications have been accounted for by the ion beam induced shear flow mechanism [5,6] according to the viscoelastic

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model originally proposed [7,8] to explain the plastic flow phenomenon. However, Cliché et al. [9] have considered the momentum transfer driven mass transport process responsible for this effect.

The value of  $S_e$  in the works of Guntzmann et al. [5,6] was about 35 keV/nm in <sup>129</sup>Xe ion irradiation of metallic glass samples. So far, no attempt has been made to investigate the surface modifications of metallic glasses at smaller values of  $S_e$ . Therefore, the validity of the shear flow mechanism for surface modifications at lower  $S_e$  ( $\approx 10$ keV/nm) requires experimental verification. The present work is motivated to study such surface modifications. Since the experiment was designed to study plastic flow and on-line electrical resistivity also, ion-implantation was avoided. Therefore, the ion species and the energies used were selected so as to meet this requirement within the limitations imposed by the sample thickness and beam parameters of the 15 UD Pelletron accelerator at Nuclear Science Centre, New Delhi.

In this paper, we present swift heavy ion beam induced surface modification results of two metallic glass systems MG2705M ( $Co_{69}B_{12}Si_{12}Fe_4$  Mo<sub>2</sub>Ni<sub>1</sub>) and MG2204 (Ti<sub>50</sub>Be<sub>40</sub>Zr<sub>10</sub>) using 130 MeV <sup>28</sup>Si and 260 MeV <sup>107</sup>Ag ions, respectively. The value of electronic energy loss for <sup>28</sup>Si irradiation was 5.75 keV/nm and that for <sup>107</sup>Ag irradiation was 16.25 keV/nm [10]. The resulting surface modifications have been investigated by Scanning Electron Microscopy. The validity of the shear flow mechanism to the observed surface modification has also been discussed.

# 2. Experimental

# 2.1. Specimen preparation

Metglass MG2705M and MG2204 were procured from Goodfellow Cambridge England, in the form of foils of thickness about 25  $\mu$ m. The TRIM97 code gives projected range,  $R_p = 19.15$  $\mu$ m, for 130 MeV <sup>28</sup>Si ions in the MG2705M and  $R_p = 19.30 \mu$ m for 260 MeV <sup>107</sup>Ag ions in MG2204 [10]. Therefore, the foils were thinned down using the method of cold rolling, to thicknesses of about 17 and 14  $\mu$ m, respectively, to avoid the ion implantation. The detail about sample preparation and irradiation plans are given in Table 1.

#### 2.2. Sample mounting and irradiation

Samples were mounted on a specially designed target ladder [4]. The MG2204 samples were clamped between two copper plates so that part of the samples stood out for ion beam exposure (self supporting mounting). Samples of MG2705 were attached to the copper block using GE varnish (substrate bound mounting).

Irradiation of both types of the specimen was carried out at the Material Science Beam line of Nuclear Science Centre, New Delhi. To ensure uniform irradiation, the beam was made incident upon the samples in the scanning mode. The irradiation temperature was maintained at about 90 K throughout the experiments. This was constantly monitored using a platinum resistance thermometer (4W RTD type: PT385) attached to the target ladder. The variation in temperature was  $\pm 2$  K.

Metglass 2204 was irradiated with 260 MeV  $^{107}$ Ag ion (charge state + 19) beam with a flux of about  $10^{10}$  ions/cm<sup>2</sup>/s. The irradiation was performed at normal incidence and continued upto a fluence of about  $1.1 \times 10^{14}$  ions/cm<sup>2</sup>. Similarly, 130 MeV  $^{28}$ Si ion (charge state + 9) beam with a flux of about  $10^{10}$  ions/cm<sup>2</sup>/s was used to irradiate MG2705M at normal incidence, until a fluence of  $1.154 \times 10^{16}$  ions/cm<sup>2</sup> was reached.

### 2.3. Scanning electron microscopy

Surface morphology of the unirradiated and irradiated specimen was investigated ex situ by scanning electron microscopy at room temperature. For this purpose, the instrument model JSM-35CF, supplied by M/s Joel, Tokyo, was used. The electron microscopy was carried out at the National Physical Laboratory, New Delhi.

## 3. Results and discussion

Scanning Electron Micrographs (SEM) of the metallic glass samples were taken at various magnifications, prior to and after the irradiation. Figs.

Table 1 Irradiation plans

Sample	Composition	Energy/Ion beam	<i>R</i> <sub>p</sub> (μm)	Sample thickness (µm)	$(dE/dx)_e$ (keV/nm)	Mounting
MG2705M	Co <sub>69</sub> B <sub>12</sub> Si <sub>12</sub> Fe <sub>4</sub> Mo <sub>2</sub> Ni <sub>1</sub>	130 MeV <sup>28</sup> Si	19.15	17	5.75	Substrate bound
MG2204	$Ti_{50}Be_{40}Zr_{10} \\$	260 MeV <sup>107</sup> Ag	19.30	14	16.25	Self supporting

1 and 2 show the SEM of unirradiated and irradiated surfaces of metglass 2705M, respectively. Similarly, the SEM of unirradiated and irradiated surfaces of metglass 2204 are shown, respectively in Figs. 3 and 4. A comparison between Figs. 1 and 2 as well as between Figs. 3 and 4, shows that the sharp surface disorders have been smoothened out after irradiation for both types of metallic glasses. It may be clearly observed that the narrow and shallow channels are eliminated completely and the wider and deeper channels have become shallower and narrower. It is also evident that the heights of the 'hills' have decreased and the 'valleys' have been filled up during the smoothing process, as reported earlier by Guntzmann et al. [6].



Fig. 1. Scanning Electron Micrograph of unirradiated MG2705M (Co<sub>69</sub>B<sub>12</sub>Si<sub>12</sub>Fe<sub>4</sub>Mo<sub>2</sub>Ni<sub>1</sub>).



Fig. 2. Scanning Electron Micrograph of 130 MeV <sup>28</sup>Si ion irradiated MG2705M (Co<sub>69</sub>B<sub>12</sub>Si<sub>12</sub>Fe<sub>4</sub>Mo<sub>2</sub>Ni<sub>1</sub>).



Fig. 3. Scanning Electron Micrograph of unirradiated MG2204 (Ti<sub>50</sub>Be<sub>40</sub>Zr<sub>10</sub>).



Fig. 4. Scanning Electron Micrograph of 260 MeV <sup>107</sup>Ag ion irradiated MG2204 (Ti<sub>50</sub>Be<sub>40</sub>Zr<sub>10</sub>).

These results could be qualitatively explained on the basis of viscoelastic model [7,8] originally proposed to explain the plastic flow phenomenon in metallic glasses subjected to fast heavy ion irradiation. Following Refs. [5,6], the ion beam induced shear flow  $v_x = 6R_pA_0\Phi\sin\theta\cos\theta$ , leads to a net shift  $\Delta x$  of the surface atoms after a time t, given by,

$$\Delta x = 6R_{\rm p}A_0 \Phi t \sin\theta \cos\theta,\tag{1}$$

where,  $R_p$  is the projected range of the ions in the target material and  $A_0$  is the value of deformation yield near the surface.

The deformation yield  $A_0$  as a function of  $S_e$  can be written as [4],

$$A_0 = \frac{1.16}{3e} \frac{1+v}{5-4v} \frac{\beta S_e}{\rho C},$$
 (2)

with  $\nu$ ,  $\beta$ ,  $\rho$  and *C* being the poisson number, thermal expansion coefficient, density and specific heat capacity of the material, respectively. From Eqs. (1) and (2) we get,

$$\Delta x = 6R_{\rm p} \ \frac{1.16}{3e} \frac{1+v}{5-4v} \frac{\beta}{\rho C} \ \{S_e(\Phi t)\}\sin\theta\cos\theta.$$
(3)

If the beam is incident normally upon the sample surface,  $\theta$  becomes the effective tilt angle provided by the roughness of the real surfaces. Metallic glasses prepared by the method of rapid quenching (at a rate of about 10<sup>6</sup> K/s) have rough surfaces (Figs. 1 and 3) because the disorders present in the liquid state are instantly frozen-in during the quenching process. Also, the value of  $\theta$  keeps on changing during the irradiation process. Nevertheless, the net contribution due to  $\theta$  is non-zero and finite at the end of certain irradiation

process. With this and the fact that v,  $\beta$ ,  $\rho$  and C are constants at a given temperature, it is clear from Eq. (3) that the shift  $\Delta x$  caused by the shear flow depends upon the product { $S_e(\Phi t)$ } [11].

For sufficiently high  $S_e$  the value of  $A_0$  is different from zero in all amorphous materials [5]. In the 260 MeV <sup>107</sup>Ag ion irradiation of MG2204  $(S_e = 16.25 \text{ keV/nm})$ , the plastic flow has already been observed [4] under the same irradiation conditions. The value of  $A_0$  has been determined to be  $\approx 10^{-16}$  cm<sup>2</sup>/ion [4], which gives a non-zero value to the shift  $\Delta x$  resulting from the local shear flow. It is argued that this non-zero value of  $\Delta x$  was responsible for the observed smoothing in the samples irradiated with  $1.1 \times 10^{14}$  ions/cm<sup>2</sup> as it decreased the heights of the 'hills' and filled up the 'valleys' [5]. No significant changes in surface morphology were observed in the samples irradiated with  $1.8 \times 10^{13}$  ions/cm<sup>2</sup>. Although, the value of  $S_e$  (5.75 keV/nm) for 130 MeV <sup>28</sup>Si irradiation of MG2705 was comparatively smaller, the associated fluence was high ( $\approx 10^{16}$  ions/cm<sup>2</sup>). Consequently,  $\Delta x$  was sufficient enough and therefore surface smoothing has been observed in the later case too.

More work in this direction is in progress. Experiments have been planned to observe the tilt angle dependence of surface modifications and to understand the role of  $\{S_e(\Phi t)\}$  factor in detail.

#### 4. Conclusion

The electronic excitation of near surface atoms caused by swift heavy ion irradiation of amorphous targets results in a transient mobility at the surface. The subsequent relaxation processes give rise to a shear flow even at the normal beam incidence due to local variations in angle of incidence because of roughness of the surface. For the values of electronic energy loss at which the deformation yield is non-zero, this shear flow produces modifications of the irradiated surfaces. These modifications of real surfaces of metallic glasses at normal beam incidence depend upon the factor  $\{S_e(\Phi t)\}$ .

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