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Revealing the Effect of Swift Carbon Ion Beam on Radiation Sensitivity of Radio-frequency Magnetron Sputtered Grown Alumina Thin Films

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Abstract

Regarding targeted treatment, hadron therapy is beneficial than conventional radiotherapy based on X rays/gamma rays due to characteristic energy loss profile of energetic charged particle during ion matter interaction. In this regard, dose monitoring and its precise measurement with spatial accuracy are very crucial. Thermoluminescence (TL) based thin film radiation dosimeter has the potential to serve this purpose. In hadron therapy, use of swift carbon ion beam is found to be best due to both physical and biological benefits. Regarding swift ion irradiation, although electronic energy loss (S_e) *dominates over nuclear energy loss* (S_n)*, still the possibility of* S_n *induced lattice defects formation cannot be completely ruled out. Therefore, efficiency/sensitivity of dosimeter could be changed. Aluminium oxide* (Al_2O_3) *is one of the promising materials in the field of dosimetry. For the use of* Al_2O_3 *based radiation dosimeter for ion beam dosimetry, it is necessary to understand the effect of swift carbon ion irradiation on* Al_2O_3 *thin films. In this article, the effect of irradiation of swift carbon ion beam on radiation efficiency/sensitivity of radio frequency magnetron grown alumina* (Al_2O_3) *is presented and the feasibility of* Al_2O_3 *based radiation dosimeter is discussed.*

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INTRODUCTION

Radiation dosimetry is an active field of research owing to its large-scale application in environmental radiation monitoring, food irradiation, personnel dosimetry, radiation therapy, etc. [1, 2]. Most of the studies are based on electromagnetic (*viz.* gamma, high energy X-ray, etc.) radiation as their chances of unintentional exposure are high in daily life. The unintentional exposure of charged particles is severe in outer space only [3] thus data on the sensitivity variation of dosimeter (used in space dosimetry) for charged particle is scarce. However, recent development of hadron therapy (dealing with treatment of cancer by charged particles) [4–7] demands special attention to ion beam dosimetry for precise measurement of energy deposition per unit mass (absorbed dose) of the targeted region of a biological object. In general, single crystal, pellets, or powders are used as passive detector materials/phosphors for X-ray/gamma radiation dosimetry, where the dose is estimated by thermoluminescence (TL) technique (involving the trapping of electrons and/or holes in defect states under radiation exposure, followed by the emission of light owing to radiative recombination of thermally stimulated detrapped charge particles at the luminescent centers) [1]. However, the use of thin phosphor film is crucial for ion beam dosimetry, especially for online dose monitoring. The absorbed dose monitoring during cancer treatment is a challenging task. One of the ways is to monitor absorbed dose to the targeted tissue by estimating the entrance skin/exit skin dose using an online dosimetry system. The development of highly sensitive thin phosphor films having properties of optically stimulated luminescence (OSL) could be a potential candidate for such purpose [5].

Among various phosphors, alumina (Al_2O_3C) has attracted a considerable attention due to its simple and desirable glow curve below 250°C [8]. This material is especially advantageous for being sensitive at low dose, high thermochemical stability, and also for showing OSL at room temperature (RT) [9, 10]. In case of alumina, the role of lattice defects and their control, particularly oxygen vacancies (V_0) like F centers (V_0 with two electrons), F^+ centers (V_0 with single electron), is found to be very crucial to influence TL and OSL sensitivity [8, 11–13].

When gamma rays pass through matter, it loses energy by three ways: i) photoelectric effect, ii) Compton scattering, and iii) pair production. Depending upon the energy of the incident gamma rays, any of the above mentioned process dominates, however, none of the process affects the lattice structure of the matter. On the other hand, during the passage of ion beam through matter, it loses energy by two process: i) electronic energy loss (S_e), and ii) nuclear energy loss (S_n). While ionization occurs during the electronic energy loss component, nuclear energy loss component is responsible for damaging the lattice structure, resulting intrinsic defect formation in lattice. It is true that during the passage of swift ion (highly energetic ion; energy of the order of MeV or GeV) through a thin film, S_e dominates over S_n, thus the chances of creation lattice defects is much less. However, there is still a chance of stabilizing lattice defects due to the passage of swift ion beam. As mentioned earlier, intrinsic defects (F-center and/or F^+ center) plays a crucial role in TL and OSL response. If such kind of defects get stabilized due to ion beam irradiation, then only thin film radiation sensitivity can be affected. Since this kind of study is scarce in literature to the best of our knowledge, the present study was taken up.

In this article, we investigated the effect of swift carbon ion beam on the radiation sensitivity of the radio frequency (RF) magnetron sputtered grown alumina films. The investigation was carried out in 1200°C annealed thin film due to its better radiation sensitivity than its pristine counterpart. Here, gamma radiation response of the said film was considered as main parameter to understand the radiation sensitivity changes due the passage of ion beam through the thin film, if any. The results shows that radiation sensitivity increases for initial ion beam doses but approaches the value of unirradiated alumina thin film. The possible reason of such changes and its limitation is discussed.

EXPERIMENTAL OUTLINE

The alumina thin film deposition was carried out on clean SiO_2 coated p-type Si(100) substrates [Silicon Quest International, Inc] by RF magnetron sputtering technique at room temperature. Alumina target (MTI corp., purity 99.99%) was used for deposition purpose and during the six hours of deposition, 100 W power was maintained. Further details of the deposition can be found elsewhere [14]. The as-deposited films were annealed at 1200°C for three hours in a muffle furnace (ambient air) and cooled down slowly there itself. Annealed thin films were cut into six small pieces with same dimension (0.5×0.5 cm^2), where five of them were irradiated with 80 MeV carbon ion beam to various fluence (3×10^{10}– 1×10^{13} ions/cm^2). The remaining one which was not ion irradiated has been considered as control sample. For simplicity, the control sample is named as A, whereas the irradiated samples with increasing ion fluence will be called B, C, D, E, and F in the following. The corresponding absorbed doses are calculated to be 9, 26, 130, 260, 2630 kGy, respectively. The irradiation of the samples work was carried out using 15UD Pelletron accelerator, Inter University Accelerator Centre (IUAC), New Delhi, India.

TL signals were examined in a Harshaw 3500 Reader with a heating rate of 5°C/sec up to a maximum temperature of 400°C. To check the gamma radiation response, all the samples were exposed to required doses with gamma radiation (source: Co-60; 3.4 kGy/h) before and after ion irradiation and then the TL signal was recorded.

RESULTS AND DISCUSSION

Figure 1 shows the energy loss profile of 80 MeV carbon ion beam in bulk alumina as simulated using SRIM software [15]. Here, the density of alumina is considered to be 3.95 g/cm^3. It can be seen from the plot that S_e dominates over S_n except last few nm (shown in the inset) of the traversed distance. As the deposited thin film is ~500 nm thick, thus deposited energy in alumina thin film will be mostly dominated by S_e, causing ionization and electronic excitations. In this case, the magnitude of absorbed dose (D') can be estimated using the relation [16]

$$D' = 1.602 \times 10^{-10} \times \frac{dE/dx}{\rho} \times \varphi \qquad \text{(i)}$$

where, D' is in Gy, $\frac{dE/dx}{\rho}$ is in MeV-cm^2/g denoting the mean energy loss (mass stopping power) in the target with density ρ, and the ion fluence φ is in ions/cm^2. In the Figure 1, a schematic of the alumina thin layer (~500 nm thickness) is shown, revealing that 80 MeV C ion loses 7210 MeV/cm energy by means of S_e whereas only 4 MeV/cm energy is deposited for S_n. Therefore, the value of *dE/dx* can be taken as ~7210 MeV/cm for the deposited thin films. As mentioned earlier, the different samples were irradiated to fluence in the range of 3×10^{10}–1×10^{13} ions/cm^2, which is equivalent to be ~9-2630 kGy absorbed dose (D').

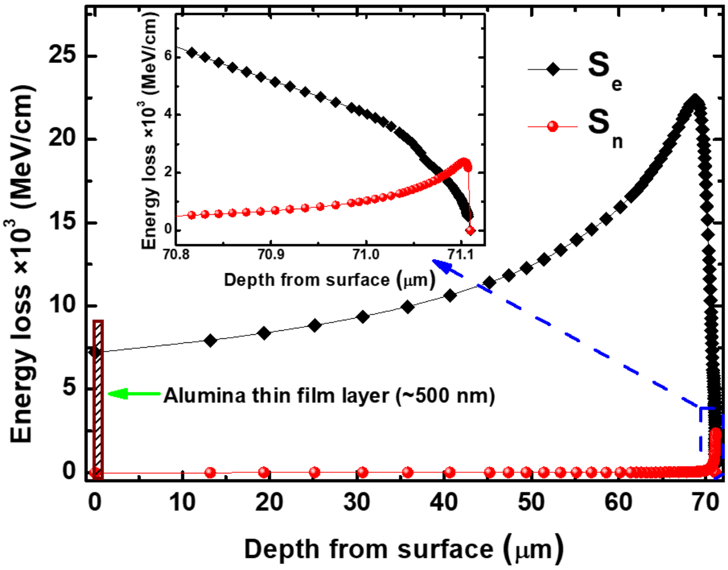

Figure 1. Energy Loss Profile of 80 MeV Carbon Ions in Bulk Alumina and its Thin Film of 500 nm Thickness.

Figure 2 shows the TL glow curves of 1200°C annealed thin films after exposing to 100 Gy – 5 kGy gamma radiation. The glow curve corresponding to 5 kGy gamma radiation response of unannealed thin film is also shown (with ten times magnification) by dashed line for comparison. It clearly reveals the improvement in TL response after annealing the thin film at 1200°C. Further, TL response curve of the annealed thin films is shown in the inset which gives a better understanding about its radiation sensitivity. It is found that the dose response of thin films is linear in this range.

The TL intensity improvement in annealed thin films can be attributed to temperature driven increase in F centers, though a spatial separation between electron traps and the F centers are also found to be a factor. The detailed analysis regarding TL intensity variation and possible reason behind the variation in the shape of TL glow curve can be found in our earlier report [14].

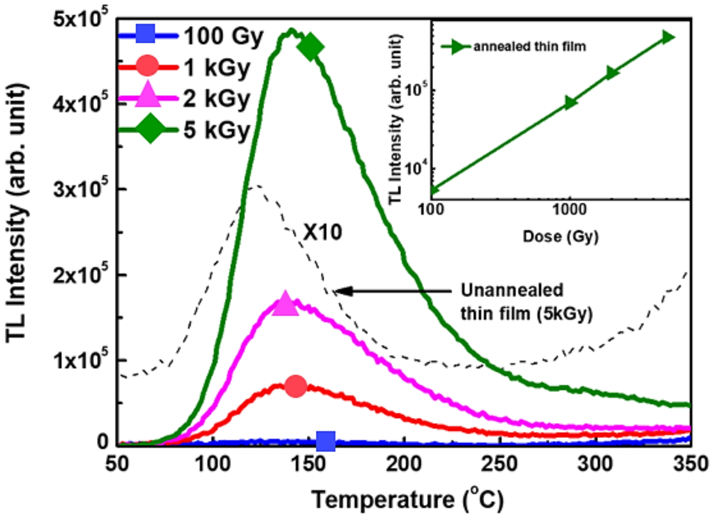

Figure 2. Thermoluminescence Glow Curves of Unannealed and Annealed Alumina Thin Films. TL Response Curve of Annealed Alumina Thin Film is Shown in The Inset.

Before ion irradiation, all the samples (A-F) have been irradiated with 2 kGy gamma ray and its TL glow curve are recorded as exhibited in Figure 3. The purpose of these kind of study is to check the homogeneity of all the samples which are to be used for further studies.

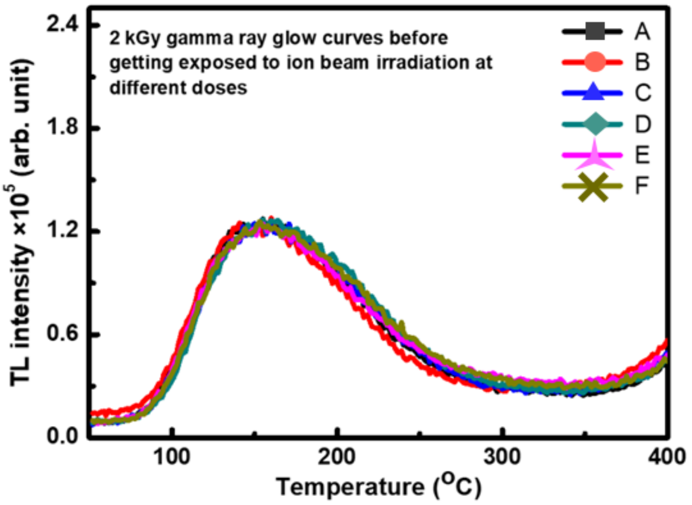

Figure 3. TL Glow Curves of Thin Films Irradiated to 2 kGy of Gamma Ray Exposure.

The glow curve clearly reveals that all six thin film samples (A-F) glow curve is similar and their peak intensities are also almost same (less than 5% variation is recorded). Thus, the thin film samples grown in this study could be considered as homogeneous in terms of their structure and TL properties. Subsequently, the samples (B-F) were irradiated with 80 MeV carbon ion beam to different fluences and its ion beam dosimetric response is recorded (not shown). Further, to confirm the release of all trapped electrons/holes, the TL glow curves of all the samples are again recorded and no signal above background is observed.

Now, these samples along with sample A are exposed to 2 kGy gamma ray dose and their corresponding TL are plotted in Figure 4. The change in radiation sensitivity is observed (peak intensity is found to be different for samples) and it is found to be non-monotonic. Figure 4 shows that the radiation sensitivity is almost same for sample B (which were ion irradiated with equivalent 9 kGy dose), however an increasing trend is present and this trend continues up to sample D (which were ion irradiated with equivalent 130 kGy dose). After that, the radiation sensitivity decreases. Such change in radiation sensitivity can be due to change in stabilization of ion beam irradiation induced defects which participates in the TL process. The change in radiation sensitivity can be clearly visualized from the plot in Figure 5, where radiation sensitivity of an ideal dosimeter is also shown by black square legend.

This trend is quite consistent for several sets of samples, signifying a systematic evolution of defect centers in alumina films due to ion irradiation with increasing fluence within the given range. Note that, considering an impinging carbon ion covers 1 Å^2 area while hitting the film surface, a minimum of 10^{16} ions are required to cover 1 cm^2 area [17]. Since, in the present study, much lower fluence range is used, the alumina film is partially damaged. Therefore, it is not straightforward to analyze the observed trend and at higher fluence, this trend could be changed.

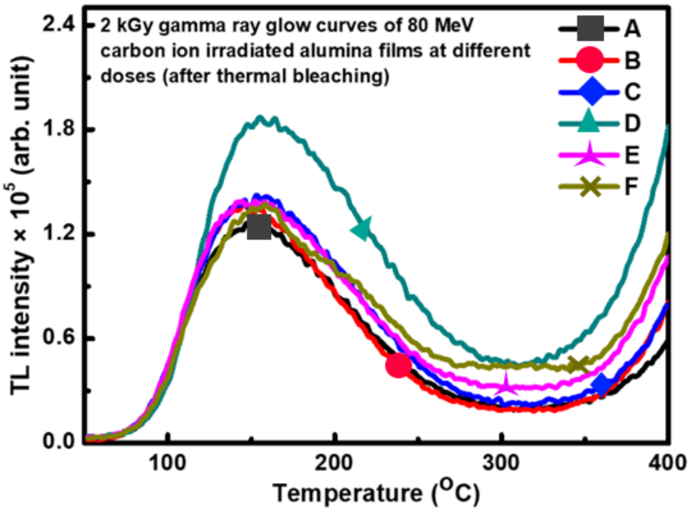

Figure 4. 2 kGy Gamma Ray Related TL Glow Curves of Post Ion Irradiated Thin Films.

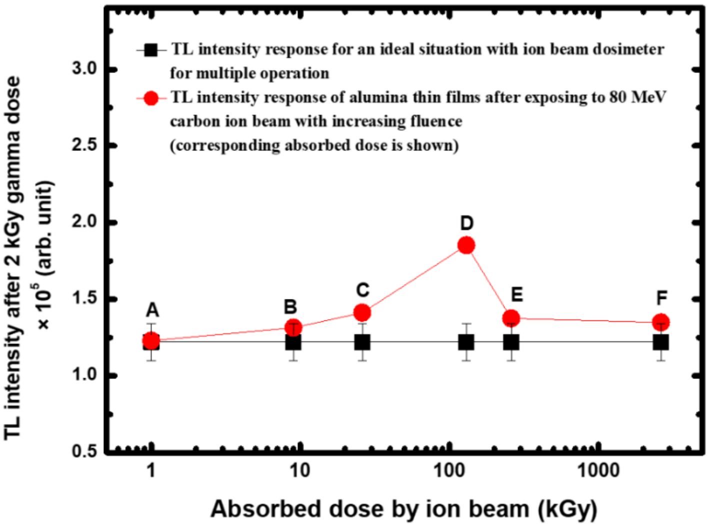

Figure 5. Variation of Radiation Sensitivity (in Terms of TL Peak Intensity) of Ion Irradiated Thin Films with Different Fluence.

CONCLUSION

Here, the variation of radiation sensitivity in ion irradiated alumina samples is checked and reported. It is observed that 80 MeV carbon ion irradiation can change the radiation sensitivity of alumina thin films non-monotonically. This results indicates that these films cannot be used multiple times for monitoring high fluence ion beam irradiation. However, the low fluence irradiated sample (sample B; absorbed dose 9 kGy) shows similar TL response like sample A (not ion irradiated). Since, for medical purpose, a dose much less than 9 kGy is often used, so these films has potential to be used multiple times in medical field. Nonetheless, further studies are required following this protocol, especially in low fluence regime (less than 10^{10} ions/cm^2) and irradiating a single sample multiple times.

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