A detailed study on Gd$_2$SiO$_5$ scintillators: recovery from increased photon yield following irradiation

T. Sako,$^{a,b,1}$ Y. Sugiura,$^a$ Q. Zhou,$^a$ Y. Makino,$^a$ E. Matsubayashi,$^a$ K. Kawade,$^{a,c}$ H. Menjo$^c$ and K. Masuda$^a$

$^a$Solar-Terrestrial Environment Laboratory, Nagoya University, Nagoya, Japan
$^b$Kobayashi-Maskawa Institute for the Origin of Particles and the Universe, Nagoya University, Nagoya, Japan
$^c$Graduate School of Science, Nagoya University, Nagoya, Japan

E-mail: sako@stelab.nagoya-u.ac.jp

ABSTRACT: Increased light intensity and recovery of GSO scintillators following irradiation were studied by using accelerator beams of 290 MeV/n carbon nuclei. This is the first measurement of recovery phase by using accelerator beams. The obtained results were consistent with previous results obtained by using $^{60}$Co gamma rays and UV excitation. We confirmed a 23% increase in light intensity following irradiation with 7.4 kGy as well as exponential recovery on a time scale of $10^4$ s. The temporal profile of recovery was fitted by a single exponential with an asymptotic increase of $13 \pm 1\%$ relative to the light intensity before irradiation. Active recovery was attempted by exposing an irradiated GSO sample to infrared light with intensity up to 8.3 kW/cm$^2$, but no indication of accelerated recovery was observed.

KEYWORDS: Scintillators, scintillation and light emission processes (solid, gas and liquid scintillators); Radiation-hard detectors; Radiation damage to detector materials (solid state)

---

$^1$Corresponding author.
1 Introduction

Gd$_2$SiO$_5$ (GSO) scintillators are among the most radiation-resistant scintillators. In general, both organic and inorganic scintillators lose their transparency after irradiation, owing to the formation of color centers. In the case of GSO scintillators, this process occurs following irradiation by gamma rays and hadrons with 10 MGy and 0.1 MGy, respectively [1, 2]. The radiation-damaged scintillators can be recovered in several ways, including natural recovery, heating, exposure to UV, optical and infrared (IR) light [2–5]. The recovery by IR exposure is interesting because the emission wavelengths of the scintillators are usually far from the IR band. By choosing the sensitivity of photo-sensors or by applying filters, active recovery can be achieved even while operating the photo-sensors. This is not the case for the recovery by heating or by optical UV exposure. Some contributions of the IR (860 nm and 740 nm) light to the recovery of transparency in PWO scintillators were reported in [4] and [5].

Apart from the transparency loss, GSOs exhibit an increase in the light yield. This was first reported in [6] for the irradiation by gamma rays from $^{60}$Co source and was confirmed in [7] for the irradiation by accelerator carbon beams. Both studies reported a 20 to 30% increases following irradiation with 10 kGy and saturation at that level only for the GSO with 0.4–0.5% cerium dope. It was also reported that the light yield increase relaxes naturally or by heating. Regarding the natural recovery at room temperature, in [6] it was reported that for a sample exhibiting a 30–35% increase in its light yield, the light yield increase recovered to 18% and 15% after 60h and two weeks, respectively. In [7] it was reported that a sample with 21% increase in light yield recovered to the asymptotic level of 12% increase. Though UV pulses from a Xenon lamp were used for exciting the scintillator in this setting, the recovery level is similar to the result reported in [6].
The mechanism of the increase and recovery of light yield was explained in [6] as follows. Unavoidable crystal impurities lead to stable energy levels that trap some of the excited electrons before these are transferred to the cerium dopant’s emission band. This occurs even in the absence of irradiation, and a certain level of ‘inefficiency’ is usually maintained [8]. When a GSO is irradiated, these trapping levels are occupied by electrons and the inefficiency is reduced. This explains the increase and saturation of the light yield. The recovery is explained by thermal fluctuations owing to which the trapped electrons are gradually released.

The importance of radiation-resistive scintillators for future high-intensity and high-luminosity accelerator experiments has been steadily increasing. To make the experimental conditions as stable as possible one must understand the characteristics of post-irradiation recovery. In this paper, we studied the early timescale recovery of GSO scintillators after the irradiation with accelerator carbon beams. The recovery was monitored by using a low-intensity beam of carbons for clarifying the response to particles. Active recovery was also tested by exposing an irradiated GSO sample to IR light. The setup and procedure of the irradiation with carbon beams are explained in section 2. The experimental results are summarized in section 3. In section 4 these results are compared to the previously obtained results. Conclusions are presented in section 5.

2 Beam irradiation at HIMAC

The experiment was performed at the Heavy Ion Medical Accelerator in Chiba (HIMAC) in the National Institute of Radiological Sciences, Japan, from November 12 to 14, 2013. The facility was available only at night time. The period from the first night until the beginning of the second night is labeled as DAY1, and the corresponding period for the second night is labeled as DAY2. In this test, a beam of carbon with 290 MeV/n was used. The beam was extracted every 3.3 s and the spill cycle within the beam existed for 1.0 s. The de-bunched beam had reasonably flat temporal and spatial structure with a diameter of about 7 mm. A low-intensity (5,000 particles per spill before collimator) beam, called “the probe beam” was used for monitoring the response of GSO scintillators, while high-intensity beams (within up to $5 \times 10^9$ particles per spill) were used for irradiation.

2.1 Setup

The experimental setup is illustrated in figure 1. The beam was collimated to the diameter of 3 mm by using an aluminum collimator, and irradiated on the GSO samples located inside a shielded box. The beam intensity was monitored by using an ion chamber, a plastic scintillator plate and a scintillating fiber. The charge from the ion chamber was integrated by using a current integrator (ORTEC 439) which generated a logic pulse every $10^{-10}$ Coulombs of charge, and the number of pulses was recorded. The lights from the scintillation detectors were monitored by using photomultiplier tubes (PMTs) and the number of discriminated signals was recorded. After irradiation with 10 kGy, the signal amplitudes of the plastic scintillators decreased to half of their values before irradiation. The counting thresholds were sufficiently low for avoiding post-irradiation inefficiency. The scintillators were replaced before the start of DAY2. Because the sensitivities of the ion chamber and the scintillation detectors are complementary for beam intensities above and below $10^6$/ spill, two
different types of beam monitors were used. The absolute number of particles passing through the ion chamber was calibrated in the overlapping intensity range by using the scintillation detectors.

Figure 1 also illustrates the experimental setup in the shielded box. Three GSO bars ([9], 0.4% Ce doped, HITACHI Chemical) with dimensions 1 mm × 1 mm × 40 mm were aligned perpendicular to the beam. A 290 MeV/n carbon deposits on average 50 MeV in a 1-mm-thick GSO bar, resulting in the radiation dose of $1.7 \times 10^{-7}$ Gy/particle in the aperture of the 3 mmφ collimator (the density of GSO is 6.71 g/cm$^3$). All GSO bars were optically glued to 0.7-mm-diameter quartz fibers. All GSO-fiber pairs were held in thin aluminum tubes with inner and outer diameters of 1.6 mm and 2.0 mm, respectively. Two bars, labeled GSO-A and GSO-B, were irradiated with an intense beam, while the third bar (GSO-C) was spared and served as a reference sample. The GSOs responses to the probe beam were monitored by using two PMTs HAMAMATSU R7400U (PMT1 and PMT2) for redundancy. To eliminate a possible gain instability of the PMTs, the two PMTs were used for monitoring all GSO bars by moving the GSO bars along the beam direction as detailed below in figure 2. A plastic scintillator (BC-404) with dimensions of 25 mm × 25 mm × 1 mm was placed behind the GSO-C and was monitored by using a PMT HAMAMATSU R7400U (PMT3). This scintillator and PMT3 were moved together with GSO-C for avoiding the intense beam, and the discriminated signal of PMT3 was used for triggering data acquisition during the probe beam operation. PMT1 and PMT2 were operated with HV of −1000V. Signals from both PMTs were amplified by a factor of 10 and the integrated charge of the amplified PMT signals was recorded by using a charge ADC.
Figure 2. Alignment of the components in the shielded box. POSITION-0 was used for high-intensity irradiation while the other positions were used for measuring the GSOs responses to the probe beam.

(CAEN V965). Associated with each trigger, a delayed trigger was generated after 10µsec and charge was recorded for determining event-by-event pedestal value. In the analysis, we present the pedestal-subtracted ADC values.

At the POSITION-4 defined below in section 2.2 and figure 2, the GSO-A sample could remain under an IR lamp (KLV MAXIR 72-0100-08). The lamp has a peak wavelength of 1.3 µm, color temperature 2,000 K and intensity of 54.0 mW/cm² (at 20 cm from the lamp) when operated with a current of 1.56 A, as was the case in this experiment. A small hole was made in the aluminum tube to allow the irradiated part of GSO-A for IR exposure. To certify the exposure with IR only, we added a filter passing light with wavelength above 1 µm. Because the lamp was placed about 5 cm above the GSO-A, approximately 860 mW/cm² of IR light was exposed assuming the lamp was point like.

2.2 Experimental procedure

During DAY1 and DAY2, the same GSO samples were used for determining the response to repeated irradiation. Due to a mechanical problem, the GSO-C signal was not obtained after high-intensity irradiation on DAY2.

As explained in section 2.1, some components were moved inside the shielded box. Figure 2
shows the components positions. These positions were remotely controlled. POSITION-0 defined the position used for high-intensity irradiation. The fibers from the GSO bars were offset from the PMTs for keeping high voltages for the PMTs. POSITION 1-4 defined the positions for the probe beam operation, with POSITION-4 used for exposing the GSO-A to the IR lamp. During the two nights, before high-intensity irradiation, the responses of all GSO bars were measured by using the probe beam at the positions POSITION 1-3. At each position, 3,000 and 10,000 events were obtained on DAY1 and DAY2, respectively. Owing to the difference between the shapes of the collimator aperture and the GSO-bars, about half of events contained particle-related signals. On DAY1, the beam intensity was increased in 7 steps. After about 10 min of irradiation during each step, the accumulated doses became 2.2, 10, 37, 79, 2,300 and 7,400 Gy. During each step, the probe beam was operated for about 5 min at POSITION 1-3. On DAY2, the highest-intensity beam was radiated on the samples for 33 min for achieving 10.7 kGy dose. After high-intensity irradiation, the probe beam operations continued during both nights. On DAY1 (DAY2), the first 1 h (2 h) after high-intensity operation was devoted to monitoring the natural recovery without IR exposure at the POSITION 1-3. On DAY1 (DAY2), after 1 h (2 h), POSITION-4 was included in the scanning cycle, for exposing GSO-A to IR. Because the IR light exposures lasted 5 min each, the scanning cycle took longer for the sequence that included the IR exposure.

3 Results

Figure 3 shows the ADC spectra of GSO-A, GSO-B and GSO-C measured by using PMT1 and PMT2 before irradiation on DAY1. In figure 3 and figure 4, the combination of GSO-C and PMT1 is missing because POSITION-4 was not included in the scanning cycle before the recovery measurement. There was essentially no difference between the spectra measured by using PMT1 and PMT2. Because the event-by-event pedestals were already subtracted, events triggered without particle passage peaked at the 0 ADC count. In all spectra, clear peaks owing to the beam passage are seen at around 1,500 ADC counts. A flat component between the pedestal and carbon beam peaks is observed, especially in the GSO-B data. This can be explained as if the GSO bar was tilted around the bar axis relative to the beam, because the bar’s cross-section is square-shaped. By parameterizing the tilt angle for each GSO bar and energy resolution, the data were reasonably fitted as indicated by red dashed curves in the plots. The obtained tilt angles were 8 mrad, 47 mrad and 6 mrad for the GSO-A, GSO-B and GSO-C, respectively. In the analyses that followed, the peak positions were determined by fitting the spectra with the fixed tilt angles that were obtained as described above.

Figure 4 shows the ADC spectra for different irradiation steps on DAY1. A gradual increase of the peak position with dose is observed for the GSO-A and GSO-B (not for the GSO-C as it was not irradiated). These probe beam results for the GSO bars are discussed by using the carbon peak positions obtained by fitting the spectra. When the peak positions were normalized with respect to the results before irradiation, the peaks positions were consistent between PMT-1 and PMT-2, with maximal discrepancy of 4%. Then, the PMT stability was normalized by using the GSO-C data. Because only PMT-2 was used for monitoring the GSO-C in this scanning sequence, the results for PMT2 (GSO-A and GSO-B), normalized with respect to the PMT2 (GSO-C) data, are summarized in table 1. The errors in the GSO-A and GSO-B results are the statistical errors, and
Figure 3. Responses of the GSOs to the probe beam before irradiation of DAY1. Blue histograms show the ADC distributions and red dashed curves are the best fit results.

Table 1. Relative light intensities of the GSO samples, at each step of irradiation on DAY1. Intensities are normalized relative to the measurements before the irradiation. They are also normalized relative to the GSO-C (PMT2) measurements for canceling a possible small variation of the PMT gain.

| Dose (Gy) | GSO-A   | GSO-B   |
|-----------|---------|---------|
| 2.2       | 1.00    | 1.00    |
| 10        | 1.017±0.008 | 1.012±0.012 |
| 37        | 1.018±0.006 | 1.006±0.011 |
| 79        | 1.060±0.008 | 1.045±0.011 |
| 750       | 1.102±0.008 | 1.074±0.008 |
| 2,300     | 1.213±0.009 | 1.205±0.013 |
| 7,400     | 1.218±0.006 | 1.220±0.010 |

The difference between the two samples is comparable to the statistical errors. We found a light intensity increase of about 23% after irradiation with 7.4 kGy; these results were consistent with results from previous studies [6, 7].

The same analysis was applied to all of the data on DAY1 and DAY2. The temporal variation of light intensity relative to the first data set is plotted in figure 5 for the GSO-A (red) and GSO-B (black), renormalized with respect to the GSO-C results. As explained in section 2.2, because the
Figure 4. Responses of the GSOs to the probe beam after irradiation steps on DAY1. Different colors indicate different doses in GSO-A and GSO-B as shown in the legend. Note that GSO-C was not irradiated with the high-intensity beam.

Figure 5. Relative light intensities of the GSO samples throughout the experimental period. Black and red points are the results for the GSO-A and GSO-B, respectively. All points are normalized relative to the measurements before the irradiation on DAY1 and to the GSO-C (PMT2) measurements.
GSO-C signal was not obtained after high-intensity operation on DAY2, normalization after that time was performed by using the last GSO-C data. From the DAY1 results, the GSO-C (PMT2) stability is expected to be at the level of ±2%. After a 23% increase that was discussed above, a gradual recovery was observed during DAY1. The intensity did not recover to unity at the end of DAY1 (around 13-22h). After the irradiation on DAY2, both samples re-exhibited the increase in light intensity. However, the increase did not reach the level of DAY1, although the dose was higher than that on DAY1. The responses for GSO-A and GSO-B were essentially identical.

4 Discussion

4.1 Timescale and asymptotic level of natural recovery

Recovery data are re-plotted in figure 6 separately for DAY1 and DAY2 as functions of the elapsed time after the high-intensity irradiation. For clarity, the data points were shifted along the vertical with constant offsets that are indicated in the figure. The data points were fitted by a simple function $I = I_0 + A \times exp(-t/\tau)$. Two fits were attempted, with $I_0$ being free (FIT1) and $I_0$ fixed at 1 (FIT2),
Table 2. Best fit parameters for the recovery time profiles in figure 6. The numbers in the parentheses below $\chi^2$/DOF and fitting parameters are the probabilities of having larger $\chi^2$ and statistical errors, respectively.

|       | FIT1          | FIT2          |
|-------|---------------|---------------|
|       | $\chi^2$/DOF | I$_0$         | A             | $\tau$ (sec) |
| DAY1  |              |               |               |              |
| GSO-A | 54.6/46      | 1.129         | 0.086         | 12,500       |
|       | (0.18)       | (0.005)       | (0.006)       | (2,700)      |
|       | GSO-B        | 72.3/44       | 1.126         | 0.086        | 10,700       |
|       | (4.5 x 10$^{-3}$) | (0.008)       | (0.008)       | (2,900)      |
| DAY2  |              |               |               |              |
| GSO-A | 50.1/98      | 1.137         | 0.045         | 5,600        |
|       | (>0.99)      | (0.002)       | (0.002)       | (700)        |
|       | GSO-B        | 70.2/98       | 1.119         | 0.054        | 8,800        |
|       | (0.98)       | (0.004)       | (0.003)       | (1,800)      |

and best fit curves are plotted in figure 6 as solid and dashed lines, respectively. The results are summarized in table 2. In table 2, $\chi^2$, degrees of freedom and best fit parameters for FIT1 and FIT2 are shown for each data set. The probabilities to have larger $\chi^2$ and statistical errors are given in parentheses below $\chi^2$ and best fit parameters, respectively. FIT1 yielded statistically satisfactory results but not FIT2. It is interesting that all 4 measurements showed I$_0$=1.13±0.01. In [6] it was reported that a sample with its light intensity increased by 30-35% recovered to 15% after two weeks. In [7] it was reported that a sample with its light intensity increased by 22% recovered to 12% (92% relative to the increased value) using a similar fit like our study. All results showed very similar values of I$_0$, allowing to conclude that the increase does not recover to unity at room temperature. This implies that there are several energy levels for trapping the electrons, and about half of them have energy gaps that cannot be overcome by thermal fluctuations at room temperature.

The timescale of recovery ($\tau$) exhibited a large fluctuation compared with the statistical errors. Relying only on the FIT1 results, we can conclude that $\tau$ is about $10^4$ s, consistent with $1.5 \times 10^4$ s in [7] that was obtained from one GSO sample.

4.2 Active recovery by IR exposure

GSO-A was exposed to the IR light from 4,000 s and 11,000 s after the irradiations on DAY1 and DAY2, respectively. This timing can be identified by the longer interval of data points marked by ‘IR’ in figure 6. There is no indication of accelerated recovery rate compared with the GSO-B results. FIT1 of the GSO-A data, using a single time scale, yields statistically acceptable results and additional shorter time scale is not required to explain the data after IR exposure. We conclude that the exposure to 1.3 $\mu$m (peak) IR light with 860 mW/cm$^2$ x 5 min x 32 = 8.3 kW/cm$^2$ does not accelerate the rate of recovery (the number 32 corresponds to the number of exposures on DAY2).

5 Summary

We studied the increase in light intensity and the recovery of GSO scintillators after irradiations with high-intensity accelerator carbon beam with 290 MeV/n and doses of up to 10 kGy. Our main conclusions are:
1. GSO scintillators (0.4% Ce doped) increased their light intensity up to 23% after irradiation with 7.4 kGy.

2. The light intensity increase recovered naturally at room temperature, in the exponential manner, on the time scale of $10^4$ s.

3. Within our measurements (maximum 17 h on DAY1), the temporal profiles of recovery were captured by a single exponential function with a constant asymptotic level $I_0 = (13 \pm 1)\%$. This was consistent with a previously reported long but sparse measurement, lasting up to two weeks [6], and with a similar measurement in which the scintillator was excited by UV light [7]. We concluded that the thermal energy at room temperature is not sufficient for releasing half of the trapped electrons. We note that in [6] it was reported that the light intensity recovered to its original level after heating a GSO scintillator up to 360°C.

4. One of the two irradiated GSO samples was exposed to the IR light with 1.3 $\mu$m peak wavelength. No indication of accelerated recovery existed, up to the total exposure of 8.3 kW/cm$^2$.

The first three points in our conclusions are consistent with the previous studies although we were the only one to use the accelerator beams for both irradiation and monitoring. We expect that this detailed knowledge on the recovery characteristics of GSO scintillators following irradiation will be useful for designing future experiments at high-intensity and high-luminosity accelerators.

Acknowledgments

The authors would like to thank the HIMAC staff for providing a desired and stable beam during the experiment. The authors also thank the anonymous referee for a careful review of the paper. This research was supported by the Research Project with Heavy Ions at NIRS-HIMAC (H325) and JSPS KAKENHI Grant Number 24105704.

References

[1] M. Kobayashi and M. Ishii, Excellent radiation-resistivity of cerium-doped gadolinium silicate scintillators, Nucl. Instrum. Meth. B 61 (1991) 491.

[2] M. Kobayashi et al., Radiation hardness of cerium doped gadolinium silicate Gd-2SiO-5:Ce against high-energy protons, fast and thermal neutrons, Nucl. Instrum. Meth. A 330 (1993) 115.

[3] A.A. Annenkov, M.V. Korzhik and P. Lecoq, Lead tungstate scintillation material, Nucl. Instrum. Meth. A 490 (2002) 30.

[4] V. Dormenev et al., Stimulated recovery of the optical transmission of PbWO$_4$ scintillation crystal for electromagnetic calorimeters after radiation damage, Nucl. Instrum. Meth. A623 (2010) 1082.

[5] E. Auffray, M. Korjik and A. Singovski, Experimental Study of the Lead Tungstate Scintillator Proton-Induced Damage and Recovery, CMS-CR-2011-257 (2011).

[6] M. Tanaka et al., Applications of cerium-doped gadolinium silicate Gd$_2$SiO$_5$:Ce scintillator to calorimeters in high-radiation environment, Nucl. Instrum. Meth. A 404 (1998) 283.

[7] K. Kawade et al., Study of radiation hardness of Gd$_2$SiO$_5$ scintillator for heavy ion beam, 2011 JINST 6 T09004.
[8] J.B. Birks, *The Theory and Practice of Scintillation Counting*, Pergamon Press Ltd. (1964) 70.

[9] T. Suzuki et al., *Performance of very thin Gd$_2$SiO$_5$ scintillator bars for the LHCf experiment*, *2013 JINST* 8 T01007.