Applications of Cerium-Doped Gadolinium Silicate Gd_2SiO_5:Ce Scintillator to Calorimeters in High Radiation Environment

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We have investigated applications of cerium-doped GSO (Gd_2SiO_5:Ce, or GSO:Ce) scintillating crystals to high precision electromagnetic calorimeters in high energy physics experiments. Basic properties of GSO:Ce such as the decay time, light yield and radiation hardness were measured using short (~4 cm long) sample crystals. The radiation hardness tests include the scintillator stability under ^{60}Co exposure. The characteristics of the observed residual phosphorescence due to radiation were studied in detail. These properties are dependent on the Ce concentration. Between the two concentrations we tested, 0.5 and 1.5 mol%, GSO:Ce scintillator with 1.5 mol% Ce is better in various aspects. Recently, large ingots with 1.5 mol% Ce concentration have been grown successfully. We report optical properties of $2.5 \times 2.5 \times 26$ cm^3 crystals. We discuss the feasibility of GSO:Ce scintillator based calorimeters in operating under high radiation such as at future hadron collider experiments.

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1. Introduction

Cerium-doped gadolinium silicate (GSO:Ce) is a fast and high-Z scintillator discovered by Takagi and Fukazawa [1], and developed by Hitachi Chemical [2]. Excellent scintillator characteristics are reported since then including the light yield as large as about 20% of that of NaI(Tℓ) [3] and radiation hardness up to an order of 1 Grad [4,5]. A table of mechanical and optical properties of GSO:Ce is given elsewhere [2,5]. Among the known scintillating crystals GSO:Ce is extremely radiation resistant. Also the decay time is fast, $\sim$60 ns for 0.5 mol% Ce concentration, among the crystals which yield large light output. Growing of crystals for total absorption electromagnetic (EM) calorimeters has been established [2] for the Ce concentration of $\sim$0.5 mol%. This Ce concentration has been selected because the maximum light yield is achieved at 0.6 mol% as described in [3].

In applications of GSO:Ce to high precision EM calorimeters in future hadron collider experiments where the beam crossing is a few times 10 ns an LHC example being described below we prefer slightly faster decay time. As described in [3], the decay time is made faster with increasing the Ce concentration but is saturated above $\sim$1.5 mol%. At larger Ce concentrations, the crystal turns yellowish and substantial light absorption reduces the available light output. Therefore, we have selected the Ce concentration of 1.5 mol% and have succeeded in growing 1.5 mol% GSO:Ce crystals with 28 cm in length and 8 cm in diameter. Technical aspects of crystal growing are reported in [6,7].

The CMS group [8] at the LHC proton-proton collider experiments at CERN is designing a total absorption type EM calorimeter using lead tungstate $PbWO_4$ scintillating crystals in order to achieve an EM energy resolution of $2\%/\sqrt{E}$. This requirement is motivated to detect the Higgs particle in the $H \rightarrow \gamma\gamma$ decay mode. In the LHC experiments, the maximum dose in the EM calorimeter will be 0.5 Mrad (5 kGy) in the barrel region and 7 Mrad at $|\eta|$=2.6 in the endcap for an integrated luminosity of 5×10^5 pb^{-1}, corresponding to the first ten years. Resistance to such a high radiation level and fast response comparable to the 25 ns beam crossing interval are main issues in designing the EM calorimeter. In the following we have evaluated the GSO:Ce characteristics taking these LHC conditions as our benchmarks.

The characteristics described below are given in comparing the results obtained for 0.5 and 1.5 mol% Ce samples. Section 2 briefly reviews some of the basic scintillator properties such as the decay time, light yield, and light transmittance. Radiation hardness tests using ^{60}Co γ-rays are described in Section 3. In the previous studies [4,5] the radiation hardness was evaluated from degradation in the light transmittance. We have measured, in addition, the stability of the light yield under irradiation. A large increase in the light yield was observed under irradiation for the 0.5 mol% Ce samples. Detailed studies of this effect including the thermoluminescence properties are covered in this section. The above studies were carried out using small crystals of $1\times1\times4$ cm^3. From these studies we conclude that 1.5 mol% GSO:Ce is superior to 0.5 mol% in view of the decay time and light yield stability under irradiation. From one of the recently grown long ingots with 1.5 mol% Ce, two samples were taken with dimensions of $2.5\times2.5\times26$ cm^3, the length corresponding to 18.8 radiation lengths. Optical properties of these large samples are given in Section 4. Section 5 summarizes the results.

2. Basic Properties of GSO:Ce Crystal

2.1 Decay constant

The decay time spectrum was measured using two photomultiplier tubes (PMT) to detect two successively emitted γ-rays from ^{60}Co decay. The start signal for a TDC was produced from a PMT where a fast plastic scintillator block was attached, and the delayed stop signal was from another PMT where a sample GSO:Ce crystal was attached. In order to suppress the effect of multiple scintillating lights, a neutral density filter was inserted between the PMT and the GSO:Ce sample to reduce the light intensity such that the GSO:Ce signal emerging from the photocathode was, at most, one photoelectron.

The TDC distributions in the first 400 ns are shown in Fig. 1. The distribution was fitted well by a two-exponential function in the whole TDC range of 1 μs. We extracted the dominant and faster decay constant to be 28 ns and 53 ns for 1.5 mol% and 0.5 mol% samples, respectively. The slower components showed the decay constant of 0.26 $\pm$ 0.04 μs (0.40 $\pm$ 0.28 μs) with the integrated intensity of 13% (10%) relative to the faster one for the 1.5 (0.5) mol% Ce sample. We note that the rise time to the peak is not negligibly small: 12 ns for 1.5 mol% and 21 ns for 0.5 mol% samples. The time jitter in the measurement apparatus is substantially small as it can be evaluated from the width of the sharp peak seen right after the start signal (at $\sim$90 ns in the plot), which corresponds to γ-rays hitting the photocathode directly. The time structure of the GSO:Ce scintillation is discussed in [9-11] through the mechanism of energy transfer from Gd^{3+} to Ce^{3+}.

2.2 Light yield

Typical charge distributions for ^{137}Cs and ^{22}Na γ sources are shown in Fig. 2. We used a Hamamatsu R329 PMT which had a quantum efficiency of about 20–25% in the emission band of GSO:Ce, to which the 1×4 cm^2 face was coupled via optical grease. The ADC gate of 200 ns was generated by another PMT which was also coupled to the sample. All the other faces were wrapped in Al foil. The number of photoelectrons calculated from the widths of the peaks is approximately 500 photoelectrons (PEs) per 1 MeV energy deposition.

The light yield was measured for three samples each for 0.5 and 1.5 mol% Ce concentrations. The three 0.5 mol% samples showed the light yield with a maximum-to-minimum variation of $\pm$1.9%. They were taken from different GSO:Ce ingots which were grown from Gd_2O_3 and SiO_2 powders with different lot numbers. The three 1.5 mol% samples showed the light yield with a maximum-to-minimum variation of $\pm$2.3%. These three were taken from the same ingot. The difference could be attributed to the different surface condition and to, at least in part, the different Ce concentration along the ingot. The distribution of the Ce concentration along the ingot is quoted in Section 4. The average light yield of 1.5 mol% samples was systematically smaller by 6.0$\pm$1.8% than of 0.5 mol% samples. This number, however, is dependent on the gate width since the decay time is dependent on the Ce concentration as discussed above.

2.3 Transmittance, excitation and emission spectra

The transmittance spectrum is shown in Fig. 3 for the two Ce concentrations. The transmittance through 1 cm is measured relative to air. The absolute value depends on the surface condition of the samples. The error bars represent the rms variation among the samples, three for each concentration. Compared to the 0.5 mol% samples, the cutoff wavelength is nearly 10 nm longer for the 1.5 mol% samples. We note that the scintillation lights populate also in this wavelength region (see below).

Fig. 4 shows a two dimensional plot of the relative light yield as functions of the excitation wavelength (the ordinate) and the emission wavelength (the abscissa), measured with a fluorescence spectrophotometer, Hitachi F4500. The Ce concentration of this sample is 0.5 mol%. As illustrated, the photodetector is positioned at 90° with respect to the monochromatic photon beam injected to the sample at an angle of 30°. Because of this configuration, the obtained spectrum can be considered to represent the emission spectrum of GSO:Ce with least absorption. There are at least two excitation bands peaking at around 280 nm and 340 nm. The emission spectrum is similar in shape for the both excitation bands: it ranges from 380 to 520 nm (at 10% of the peak) or from 410 to 480 nm (at half maximum) with the peak at 434 nm. Since the emission spectrum extends to wavelengths shorter than the cutoff of the transmittance, some fraction of the emitted scintillation is absorbed. The absorption should be therefore larger in the range from 400 to 420 nm for the 1.5 mol% samples.

3. Radiation Hardness

Excellent radiation hardness of GSO:Ce has been reported previously. The deterioration in the transmittance is less than 1–2% per 1 X_o (=1.38 cm) in the radiation environment as high as 10^9 rad for low energy γ-rays [4], or 10^7 rad for 12 GeV protons [5]. The critical neutron flux is as high as 10^{14} n/cm^2 for thermal neutrons or 10^{13} n/cm^2 for fast neutrons [5]. Radiation hardness is 2–3 orders of magnitude better than any other well-known scintillation crystals.

We carried out radiation hardness tests using ^{60}Co γ sources at Japan Atomic Energy Research Institute, Tokai. In addition to the transmittance, we measured the stability of the light output under irradiation and the recovery after irradiation. We noticed that the irradiated samples showed residual phosphorescence and that some samples showed substantial light yield increase during irradiation. The characteristics of these phenomena were studied in detail. The dose rate was in the range from 0.5 krad/h to 67 Mrad/h for 0.5 mol% samples and from 0.5 krad/h to 100 krad/h for 1.5 mol% samples. The maximum dose rate expected in the LHC endcap calorimeter region is $\sim$0.1 krad/h on the average (7 Mrad in ten years). The instantaneous dose rate during the LHC experiment may be as high as $\sim$1 krad/h.

3.1 Transmittance degradation

The transmittance curves measured before and after the ^{60}Co irradiation were compared for the accumulated dose up to 1 Grad. Because the surface conditions of the sample could change each time we set it in the spectrophotometer, the absolute value of the transmittance is reliable to about 2%. The difference in the transmittance curves was 4$\pm$2% in the region below 440 nm in wavelength and negligibly small above it for all the samples. The shape of the transmittance curve normalized at longer wavelengths is usually sensitive to specific absorption bands created by irradiation. In comparing the shape of the curves, we noted a small drop in transmittance of 1-4% in the wavelength range from 390 nm to 420 nm. We have irradiated two other samples to 1 Grad: both have showed similar or smaller changes in the transmittance. The present results are consistent with the previously reported results [4,5], confirming that the radiation damage to the transmittance is small up to at least 1 Grad.

3.2 Stability during γ exposure

In the course of these radiation tests we noticed that the light output of GSO:Ce scintillator with 0.5 mol% Ce increased [12] and that there existed radiation-induced residual phosphorescence. In order to investigate the nature of this phenomenon in more detail, we have measured the changes in scintillation light yield and residual phosphorescence as follows. We interrupted ^{60}Co exposure after 1.5 hours and took the samples out of the irradiation cave. The measurement took typically 20 minutes per cycle. We repeated this cycle of exposure and measurement four times. Four samples with 1.5 mol% Ce were irradiated at dose rate ranging from 0.5 krad/h to 45 krad/h to cover the dose rate expected at LHC. Two other samples with 0.5 mol% Ce concentration were also irradiated at dose rates 0.8 krad/h and 45 krad/h.

Fig. 5 shows the charge distributions for two typical samples, one with 1.5 mol% Ce irradiated at 5 krad/h and another with 0.5 mol% Ce irradiated at 45 krad/h. The accumulated doses are 30 krad and 270 krad, respectively. The charge was measured by detecting the 0.662 MeV peak of a ^{137}Cs source with an ADC in a 500 ns gate width. The position and width of the pedestals were affected by irradiation due to the residual phosphorescence: The pedestal position in Fig. 5a is shifted by $\sim$200 counts and the width is made wider due to irradiation. In addition, the light yield defined as the pedestal-subtracted ADC count for the 0.662 MeV peak showed substantial increase especially for the 0.5 mol% Ce samples. The results of the light yield so measured are summarized in Fig. 6. The 0.5 mol% sample irradiated at 45 krad/h showed an increase in the light yield of 30–35% within three hours. The increase in 1.5 mol% Ce samples was at most 8% at 45 krad/h and smaller ($<$2%) in the range from 0.5 krad/h to 5 krad/h. The increase was also small for the 0.5 mol% sample irradiated at 0.8 krad/h. As for the pedestal shift, we noticed residual signals with an oscilloscope even when the source was taken off. We therefore measured the average PMT anode current at an increased PMT gain. The 1×4 cm^2 face of the sample was precisely positioned on the window of the PMT. As shown in Fig. 6 the residual phosphorescence intensity evaluated in this way increased rapidly in a few hours and stayed nearly constant after. The intensity is dependent on the dose rate and the Ce concentration.

The above results can be summarized as follows. The gain instability (increase) is dependent on the dose rate and the Ce concentration. The gain increase can be maintained small at dose rate less than 5 krad/h for 1.5 mol% samples. The pedestal shift due to residual phosphorescence is typically $\sim$40 keV for the 1.5 mol% sample irradiated at 5 krad/h, which can be evaluated from the ADC count distribution (e.g. in Fig. 5) taking into account that the peak corresponds to 0.662 MeV. For larger crystals we expect an enhanced influence since the residual phosphorescence itself should increase with the volume. If we assume that the volume of the real size crystal is larger by a factor of 20, the pedestal shift would be ~1 MeV, which can be safely ignored in most of calorimeters for high energy experiments.

3.3 Recovery of residual phosphorescence

Although we have seen from the ADC distributions that the intensity of the residual phosphorescence is at a manageable level for calorimeter applications, we have carried out the following measurements in order to study its characteristics in more detail.

The time structure of the residual phosphorescence was measured with an oscilloscope. The phosphorescence consists of frequent fast signals with the individual pulse being consistent with that of a single photoelectron signal. The wavelength spectrum of the residual phosphorescence was measured by inserting successively the sharp-cut filters: It turned out that the spectrum is similar to that of GSO:Ce. From these observations the residual phosphorescence is most likely due to frequent radiative transitions as in the scintillation of GSO:Ce.

We measured the decay of the phosphorescence intensity. In the measurement which was initiated right after the irradiation was stopped, the typical time constant was ~12 min. The samples were brought to University of Tsukuba in order to study the decay in a longer time scale. A measurement for one of the samples (0.5 mol% Ce, irradiated at 45 krad/h to 2.5 Mrad) was initiated 3 hours after the irradiation was finished. The phosphorescence intensity decreased initially with a time constant of ~80 hours in the time range from 3 to 30 hours after, and then stayed nearly constant at a level about 2% of the intensity measured right after the irradiation was stopped. This sample showed an increase in the light yield of 30–35% during the exposure. The increase was 18% after 60 hours and slowly diminished to 15% after two weeks. The increase in the light yield and the intensity of the residual phosphorescence are therefore not directly related.

3.4 Residual increase in the light yield caused by irradiation

As described above, some samples (0.5 mol% samples in particular) showed a substantial increase in the light yield during exposure, which recovered to some extent in time but remained even after two weeks. In order to investigate whether the residual increase recovers more slowly in a longer time scale, we continued to measure the light yield of three samples with 0.5 mol% Ce for an extended period. These samples were irradiated at dose rates of 80, 6700 and 67000 krad/h to the accumulated doses of 12, 100 and 1000 Mrad, respectively. The light yields of these samples were measured 1, 4 weeks, and 4 months after the irradiation. The samples were kept in air at room temperature. In order to evaluate the systematic stability of the measurement system, the light yield of non-irradiated crystals was also measured as the reference. The systematic uncertainty turned out to be at most ~4%. The samples irradiated to 12 and 100 Mrad showed a stable light yield within 2% in this 4-month period. The sample irradiated to 1000 Mrad recovered by 5% in this period; the ratio of the light yield after irradiation to that before was 1.03 after 1 week, 1.02 after 4 weeks and 0.98 after 4 months. We conclude that the recovery is very slow at room temperature in the time scale one week to four months after the irradiation.

The light yields measured 2-4 weeks after the irradiation are summarized in Fig. 7 for the samples irradiated in various conditions covering the wide range of dose rates and accumulated doses. The data points are normalized to the light yield before irradiation. Since the recovery is small in this period and after, as described above, the results represent the radiation induced change which does not recover for months. The samples with 0.5 mol% Ce showed an increase in the light yield of 5–15%, whereas the samples with 1.5 mol% Ce did not show any substantial dependence on the dose rate in the range from 0.5 to 100 krad/h. The light yield for the 1.5 mol% samples is systematically smaller by $\sim$5%.

In order to verify the existence of this systematic shift for 1.5 mol% samples, we have carried out another set of measurements where the systematic uncertainty was controlled to 2%. The results are summarized in Fig. 8 for five 1.5 mol% samples: The data for the two 0.5 mol% samples will be referred to in the next section. We note that the light yield increased by 9% also for the 1.5 mol% sample irradiated at higher dose rate of 100 krad/h. Except for this sample, other 1.5 mol% samples were stable 1% during the irradiation. After the irradiation was stopped, the light yield started to decrease and was made stable at $-$(2–5)% in a time scale of a day. It seems that only the sample irradiated to 0.03 Mrad showed a light yield stable to within 1% over the whole measurement period.

3.5 Thermoluminescence

The energy transfer mechanism of GSO:Ce scintillator has been studied in [9-11]. GSO absorbs the energy of the incident particle and the excited Gd^{+3} transfers its energy to Ce^{+3} ion, which is the luminescence center. The increase in the light yield caused by irradiation could be explained through a hypothesis that there exist certain number of intermediate energy levels due to impurities or host ions in the energy gap, and they usually absorb some fraction of the energy. If the excited electrons drop to the intermediate levels and then undergo non-radiative transitions to the ground levels, they do not contribute to scintillation. If such intermediate levels are occupied by irradiation, the energy transfer efficiency to Ce^{+3} increases, resulting in the increase in the light output. The time scale of the recovery is determined by the stability of the occupied states. Since we measured a residual increase of $\sim$10% in the light yield, some of these states should be stable for months in the room temperature.

We have measured the thermoluminescence property of an irradiated GSO:Ce crystal. The sample crystal was fitted into a hole machined in the copper block which was heated using a mantel heater. A PMT was placed in a thermostat chamber, which measured the luminescence out of the crystal through a quartz window of the chamber. The temperature of the sample was controlled using a thermometer inside the copper block.

The measurement results are shown in Fig. 9, where the monitor temperature and the luminescence intensity are plotted as a function of time. The temperature was slowly raised from the room temperature to 360°C, and then brought back to the room temperature. We note that there are several specific temperatures that contribute to the luminescence and that the luminescence disappears on the way the temperature is brought down. Those specific temperatures should correspond to the energy gaps between the intermediate levels and the conduction bands from which the electrons can drop to the luminescence center Ce^{3+}. Once the occupied levels are made free, the luminescence should disappear, which is observed on the way the temperature was brought down. The increase in the light yield should disappear also under the hypothesis described above. The light yield of this sample normalized to that at pre-irradiation recovered to 1.01$\pm$0.02 after heating once to 360°C, while it was 1.10$\pm$0.02 before heating. Depending on which energy levels are contributing to the increase, the amount of recovery could depend on the maximum temperature raised. The recovery was not recognized at 100°C, and was about a half at 200°C.

We can explain qualitatively the time structure of the light yield instability during the irradiation through the same hypothesis. The two processes, emptying and filling the intermediate levels, are competing during irradiation. By assuming a certain number of intermediate levels which is dependent on the Ce concentration, the existence of the threshold dose rate and threshold dose above which the light yield reaches to the plateaux can be understood (see Fig. 6 for 0.5 mol% Ce samples irradiated at 45 krad/h). The increase was not seen for 0.5 mol% samples irradiated at smaller dose rates, and for 1.5 mol% samples in the dose rate range below ~5 krad/h. The increase during the irradiation reached to about 30%. Once the irradiation is stopped, the levels with smaller energy gaps start to be made free: The increase diminished to about 10% in a week. The remaining increase is due to occupation of the levels with larger energy gaps which are hard to be emptied at the room temperature.

In Fig. 8 the light yields after 6.5 Mrad γ-ray exposure are shown for the two 0.5 mol% samples which had been irradiated to 1 Grad about a year ago. The light yields are normalized by those measured before the present exposure. One of the samples was subjected to the above thermoluminescence test, which showed an increase of 30% in the second irradiation whereas the other samples showed a smaller increase of 22%. As described in Section 3.2, 0.5 mol% samples increased the light yield by ~30% during the first exposure. Therefore the sample undergone the thermoluminescence test showed the similar increase to them, while the irradiated sample showed a smaller increase by 10%. This difference could be attributed as the contribution of the occupied energy levels which are stable for a year at the room temperature.

4. Optical Properties of Large Crystals

We have grown ingots of 80 mm in diameter and 280 mm in length with Ce concentration of 1.5 mol% [6,7]. Two specimens of 25$\times$25$\times$260 mm^3 were taken from one of the ingots. All surfaces were polished to the optical grade.

We injected the photon beam off the spectrophotometer into a side of the crystal and measured the transmittance at three positions along the crystal. The results are shown in Fig. 10. The data Bottom and Top were taken at 2.5 cm from the bottom and top of the crystal, respectively, and the data Middle were taken at the midpoint. The transmittance is better at position Top than at Bottom between 400 nm and 420 nm. The crystals were grown with a Czochralski method and the position corresponds to the different ages in the crystal growth - crystals grow from top to bottom. The Ce concentration x in the crystal expressed in terms of $Ce_xGd_{2-x}SiO_5$ varies along the crystal length due to segregation. From an ICP (Inductively Coupled Plasma Atomic Emission Spectrometer) analysis, the Ce concentration is found to vary from 0.93% (top) to 1.39% (bottom) along the crystal for 1.5 mol% samples, and from 0.4% (top) to 0.5% (bottom) for 0.5 mol% samples.

The light yield read out from the end of long crystals is position dependent due to finite attenuation along the light path. We illuminated the GSO:Ce crystal with ^{137}Cs γ-rays through a Pb collimator of 5 mm in diameter, and emerging scintillation light was read out with a PMT (Hamamatsu R329). The light yield of small samples was calculated from the width of the photoelectric peak, while the light yield of long samples was evaluated as the ratio of the ADC count for the photoelectric peaks measured for long samples to that measured for small samples.

The light yield uniformity is summarized in Table 1, which shows the relative light outputs measured at 4 cm and 26 cm from the readout end. The uniformity was measured from either end, top or bottom. We note that the results are similar for the two samples. The nonuniformity read out from the bottom end ($\sim$29%) is better than that read out from the top end ($\sim$45%), which can be understood from the difference in transparency (see Fig. 10); when the crystal is illuminated from the far end, the transparency across the crystal is essentially the same in both cases, whereas it is dependent on the end to read out when the near end is illuminated.

Use of a reflector sheet improves the light yield and the uniformity as shown in Fig. 11. The abscissa is the distance of the γ source from the readout end, and the ordinate is the light yield in terms of the number of photoelectrons per 1 MeV energy deposition. The two groups shown in the figure are results with and without a reflective sheet. Note that the light yield is approximately 500 PEs per 1 MeV for small samples with 1.5 mol% Ce. The light yield with reflector is about 37% of the small sample at 4 cm and 29% at 26 cm. The reflective sheet we used was 100 μm thick PET (polyethyrene terephtharate containing TiO_2) which was wrapped around the crystal. The light yield increased by 27% at 4 cm and 40% near the far end, and the resulting nonuniformity along the length was reduced to 22%. With this reflective sheet, we can collect about 130 PEs/MeV from the far end of the crystal (QE $\sim$ 20%). The stochastic term of the energy resolution arising from the photo-statistics is $0.28\%/\sqrt{E}$, with E expressed in GeV.

5. Summary

Cerium-doped gadolinium silicate (GSO:Ce) scintillator has excellent properties especially in its large light yield and radiation hardness. Some of basic characteristics of GSO:Ce can be summarized as follows:

- The scintillation decay time and the rise time are, respectively, 28 ns and 12 ns for the samples with 1.5 mol% Ce concentration. These time constants depend on the Ce concentration; they are 53 ns and 20 ns, respectively, for 0.5 mol% samples.

- The light yield emerging from the 1×4 cm^2 face of the samples of $1 \times 1 \times 4$ cm^3 is estimated to be 500 photoelectrons per 1 MeV energy deposition with a bialkali phototube. Dependence on the Ce concentration is small for the samples of this size. Large crystals of $2.5 \times 2.5 \times 26$ cm^3 with 1.5 mol% Ce concentration showed the light yield of 37% (29%) of that in the small crystals at a position 4 cm (26 cm) from the readout end.

- Degradation of the light transmittance due to irradiation by low energy γ-rays is at most $4 \pm 2\%/\mathrm{cm}$ in the wavelength range below $\sim$420 nm up to at least 1 Grad. The degradation is smaller for longer wavelengths.

- The light yield under γ-ray irradiation showed an increase of $\sim$30% for 0.5 mol% Ce samples and $\sim$5% for 1.5 mol% Ce samples at a dose rate of 45 krad/h. This increase recovers partially in a few days after the irradiation, giving a residual increase of 5–15% for 0.5 mol% Ce samples. It was typically $-(2\text{–}5)\%$ for 1.5 mol% Ce samples almost independent of the dose rate up to at least 100 krad/h and of the accumulated dose up to at least 12 Mrad. No significant instability was observed for 1.5 mol% Ce samples irradiated at 0.5–5 krad/h, and for 0.5 mol% Ce samples irradiated at 0.8 krad/h.

- Irradiated samples showed residual phosphorescence, whose intensity is dependent on the dose rate and the Ce concentration. The residual increase in the light yield and the residual phosphorescence could be considered to be caused by intermediate energy levels from which electrons drop to the ground state non-radiatively. Once such levels are occupied by irradiation, the energy transfer directly from Gd to Ce^{3+} occurs more often, increasing the light output. Heating the irradiated crystal up to 360°C recovered the light yield to a level before irradiation.

Because of the fast decay time, large light output and other excellent properties, GSO:Ce (1.5 mol%) can be considered as a good material for a high precision-calorimeter in high radiation environment. The light yield instability by irradiation described above is too large to ignore for 0.5 mol% Ce crystals but is much smaller for 1.5 mol% Ce crystals. The light yield of 1.5 mol% Ce samples is stable to 1% during exposure and seem to decrease by (2–5)% in a day after irradiation is stopped. Improvement of this instability is under consideration, for example, by use of Gd with smaller impurities [13] and doping of other rare earth elements.

We have succeeded in growing 1.5 mol% Ce crystals of 28 cm length, which corresponds to $\sim$20 radiation lengths. The light yield difference along the length is at most 22% when the signal is readout from the end. Slightly longer crystals are necessary for calorimeters of total absorption type.

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Table 1: Relative light yield of the two samples measured at 4 cm and 26 cm from the readout end of the crystal. Top or Bottom in the readout end column refers to the end where the signal was read out. The crystal was grown from Top end to Bottom with a Czochralski method. No reflective sheet was used in this measurement.

	readout end	4 cm	26 cm
Sample 1	Bottom	1	0.71
	Top	1.18	0.71
Sample 2	Bottom	0.99	0.72
	Top	1.15	0.70

Figure Captions

Fig. 1 Decay time distributions of GSO:Ce crystals: 1.5 mol% (hatched) and 0.5 mol% (clear) Ce concentration.

Fig. 2 Typical pulse height distributions in the charge mode of a 0.5 mol% sample for ^{137}Cs (hatched) and ^{22}Na (clear) γ sources.

Fig. 3 Transmittance for 0.5 mol% (filled circles) and 1.5 mol% (open squares) Ce concentrations.

Fig. 4 Excitation and emission spectra measured for the sample with 0.5 mol% Ce concentration. The equi-potential curves represent the relative light output. The sample crystal was set at 30° and 60° with respect to the excitation beam direction and the photodetector direction, respectively.

Fig. 5 Pulse height distributions in the charge mode for a ^{137}Cs γ source measured before (hatched) and after (clear) the irradiation. a) The sample with 0.5 mol% Ce irradiated at 45 krad/h to 270 krad, and b) the sample with 1.5 mol% Ce irradiated at 5 krad/h to 30 krad.

Fig. 6 Change of the light yield (top) and the intensity of residual phosphorescence (above) as a function of the irradiation time. The irradiation was interrupted for 20 minutes (not included in the irradiation time given above) at the measurements. The phosphorescence intensity changes by $\sim$30% depending on the measurement order whereas the order was circulated among the samples.

Fig. 7 The light yields normalized to that before irradiation as a function of the accumulated dose. Samples with 0.5 and 1.5 mol% Ce concentration are shown. Post-irradiation measurements were made 2–4 weeks after the irradiation. The number attached to each data point is the dose rate in krad/h.

Fig. 8 The light yields normalized to that before exposure to ^{60}Co γ-rays. The two 0.5 mol% samples had been irradiated to 1 Grad about a year ago (one of them was subjected to a thermoluminescence test), for which light yields are normalized by those measured before the present irradiation.

Fig. 9 Thermoluminescence out of an irradiated sample (0.5 mol% Ce concentration). (top) Monitor temperature and (above) PMT current as a function of the elapsed time.

Fig. 10 Transmittance of a large 1.5 mol% GSO:Ce crystal measured across the crystal (2.5 cm) at three positions along the crystal.

Fig. 11 Light yield of a large 1.54 mol% GSO:Ce crystal measured at positions along the crystal. The signal is read out from the Bottom end (see text).

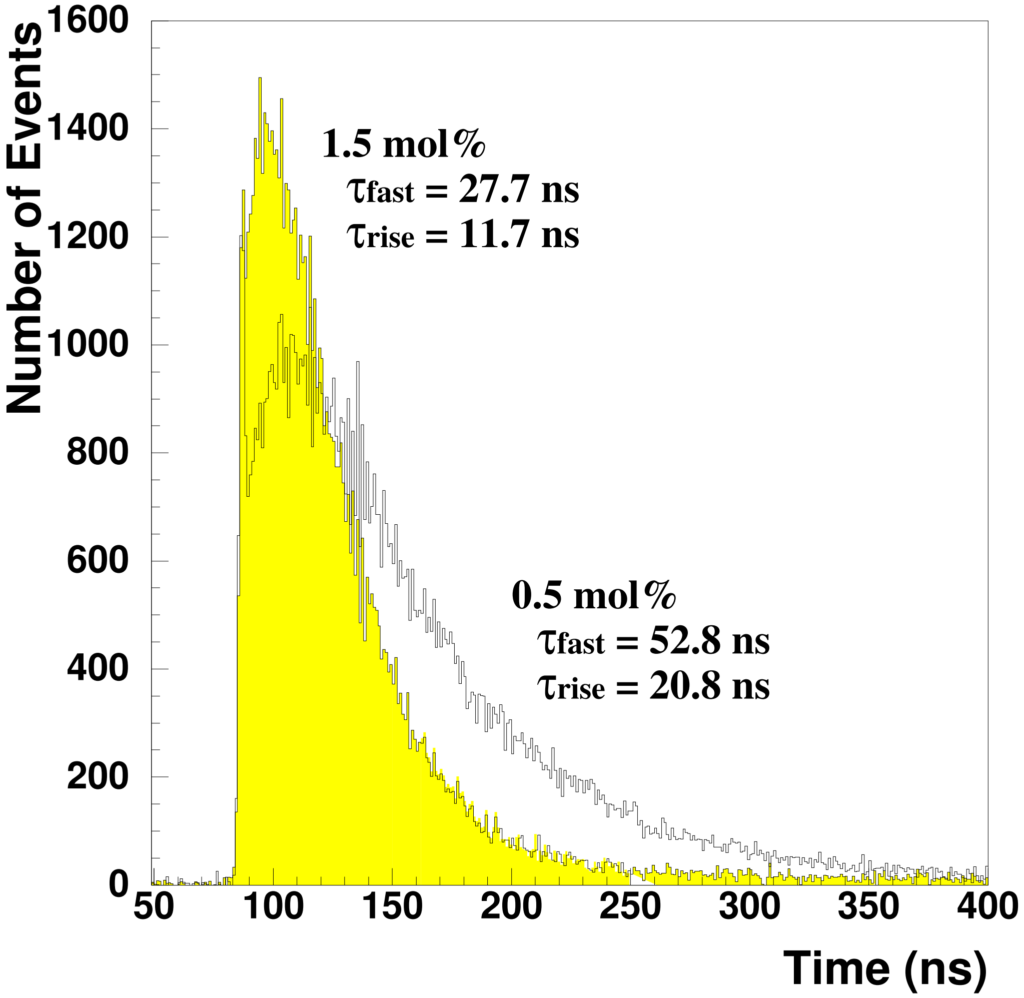

Figure 1: Decay time distributions of GSO:Ce crystals: 1.5 mol% (clear) and 0.5 mol% (hatched) Ce concentration.

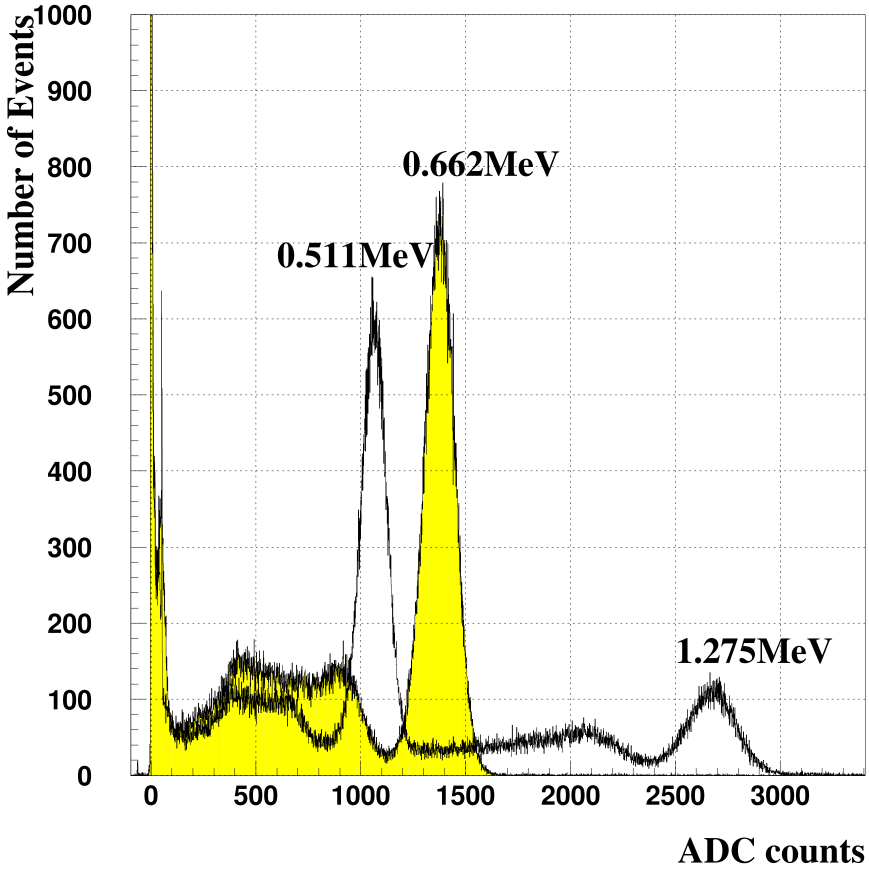

Figure 2: Typical pulse height distributions of a 0.5 mol% sample for ^{137}Cs (hatched) and ^{22}Na (clear) γ sources.

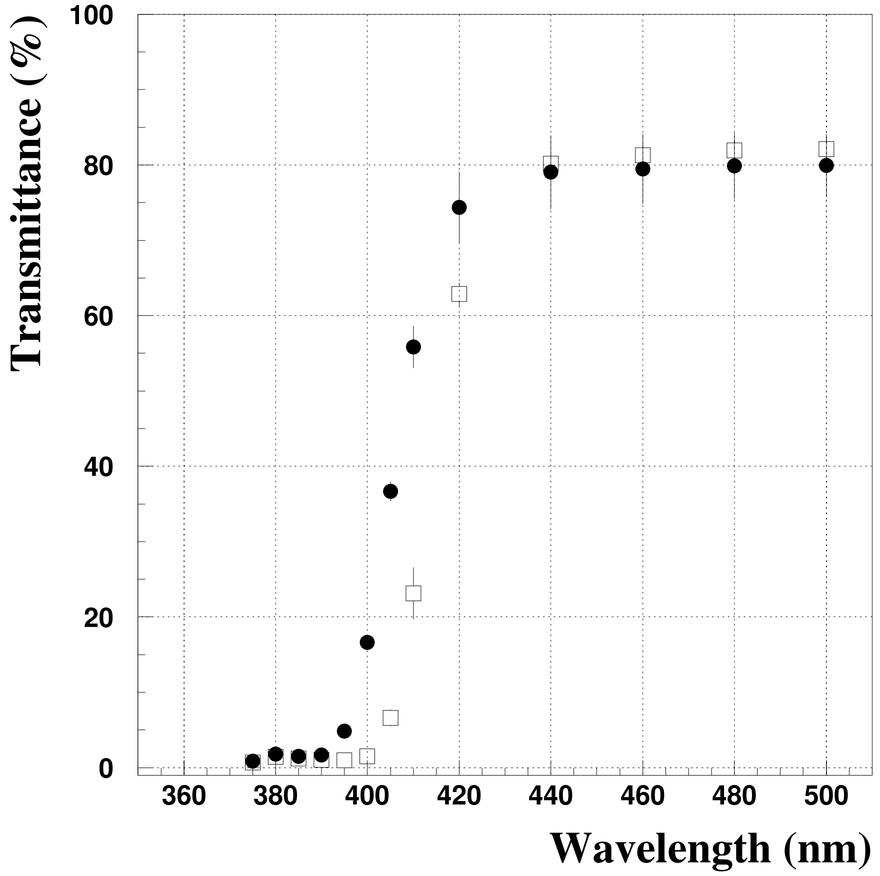

Figure 3: Transmittance for 0.5 mol% (filled circles) and 1.5 mol% (open squares) Ce concentrations.

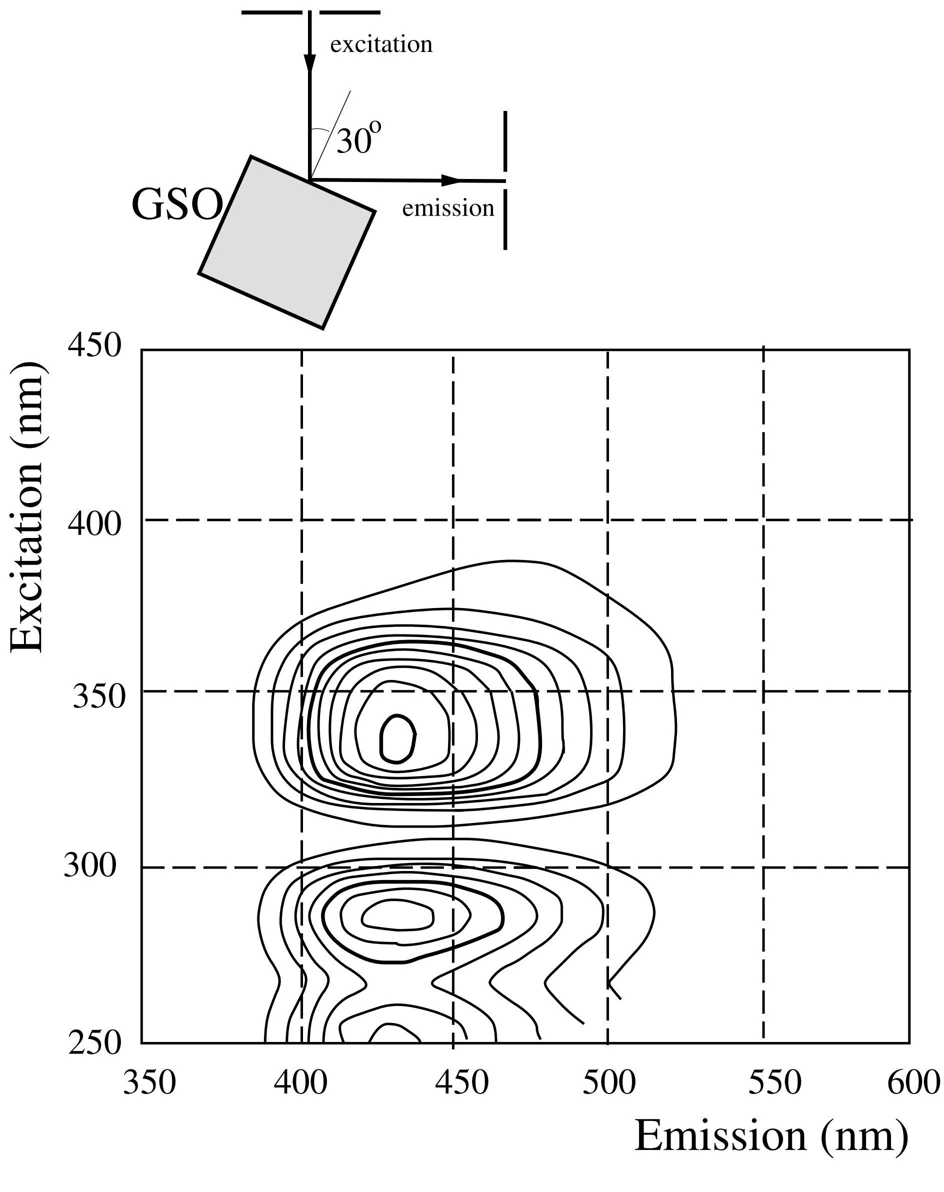

Figure 4: Excitation and emission spectra measured for the sample with 0.5 mol% Ce concentration. The equi-potential curves represent the relative light output. The sample crystal was set at 30° and 60° with respect to the excitation beam direction and the photodetector direction, respectively.

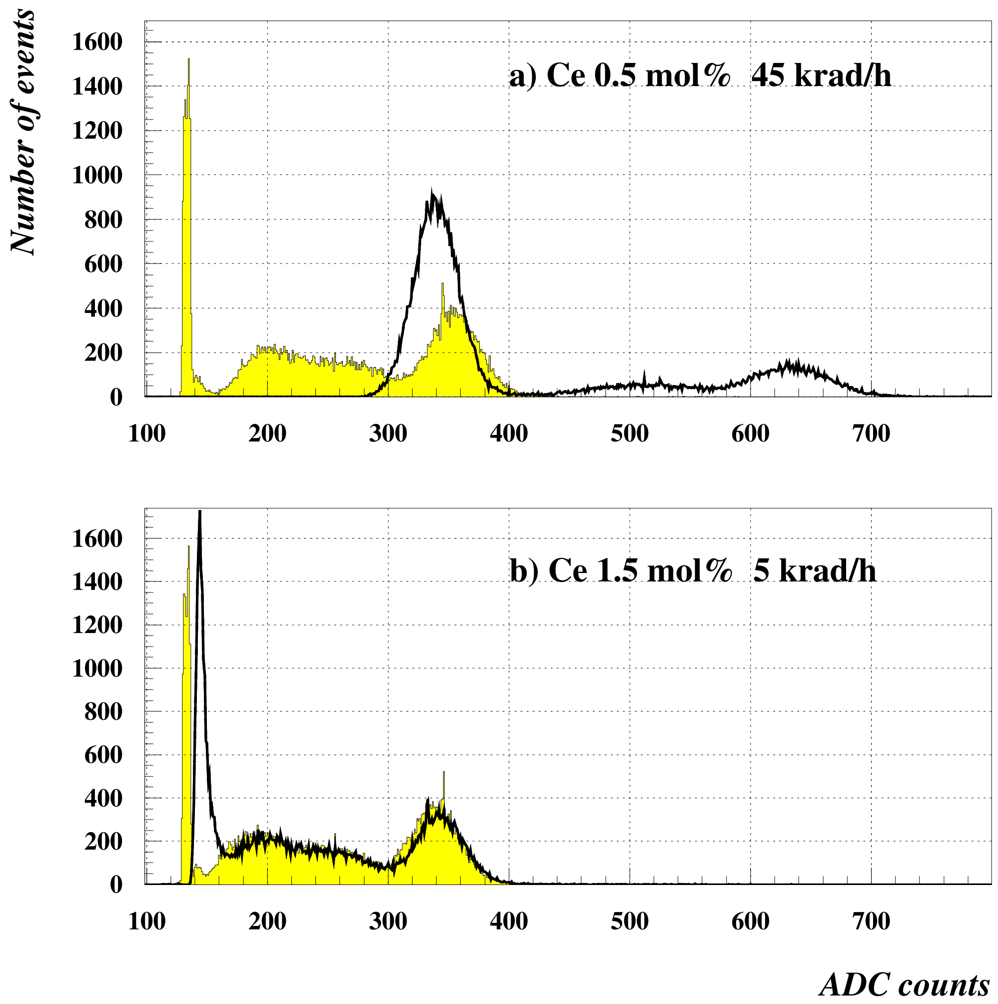

Figure 5: Pulse height distributions in the charge mode for a ^{137}Cs γ source measured before (hatched) and after (clear) the irradiation. a) The sample with 0.5 mol% Ce irradiated at 45 krad/h to 270 krad, and b) the sample with 1.5 mol% Ce irradiated at 5 krad/h to 30 krad.

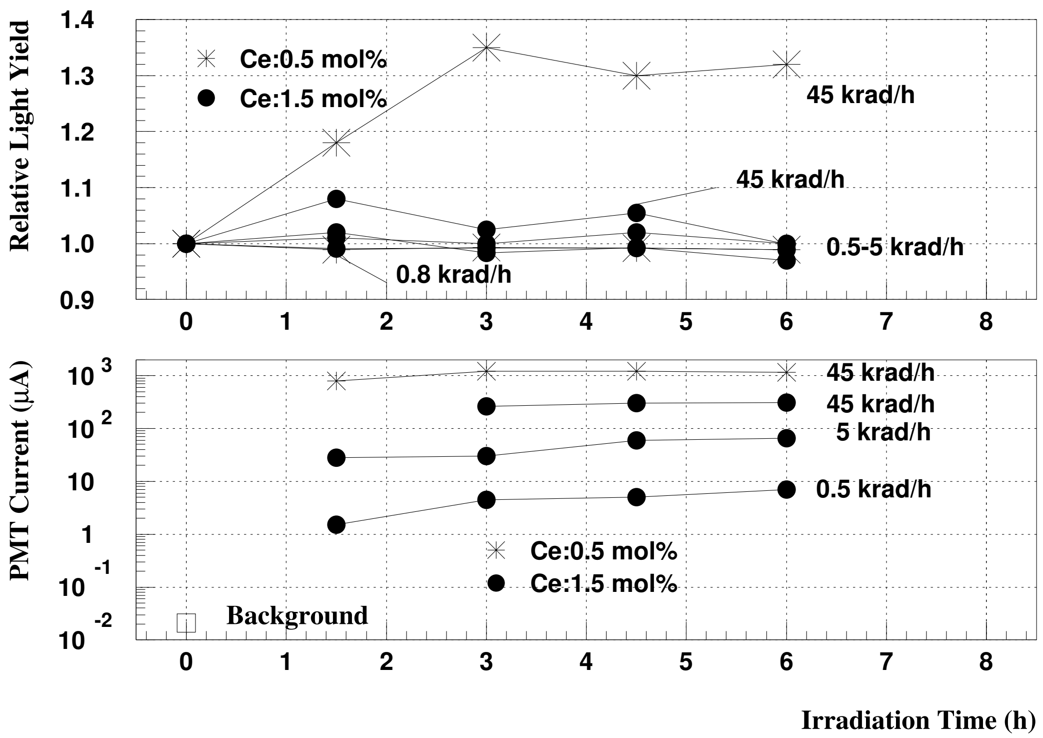

Figure 6: Change of the light yield (top) and the intensity of residual phosphorescence (above) as a function of the irradiation time. The irradiation was interrupted for 20 minutes (not included in the irradiation time given above) at the measurements. The phosphorescence intensity changes by $\sim$30% depending on the measurement order whereas the order was circulated among the samples.

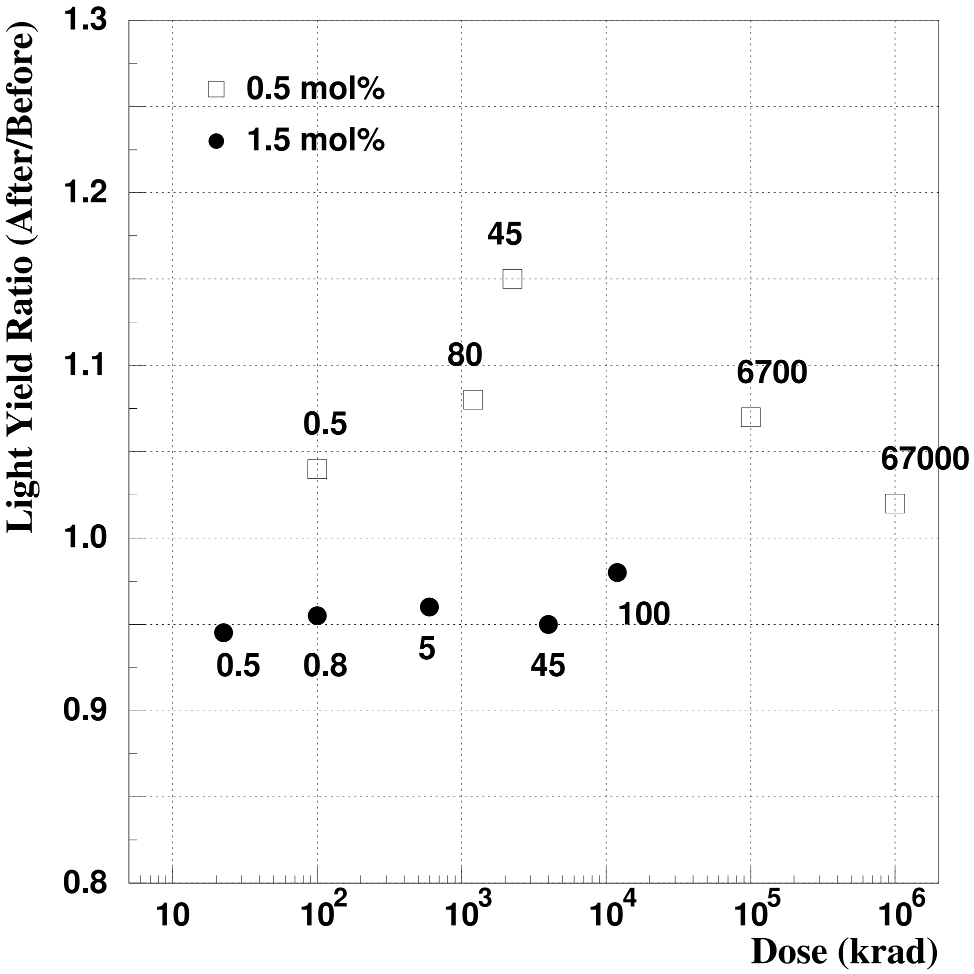

Figure 7: The light yields normalized to that before irradiation as a function of the accumulated dose. Samples with 0.5 and 1.5 mol% Ce concentration are shown. Post-irradiation measurements were made 2–4 weeks after the irradiation. The number attached to each data point is the dose rate in krad/h.

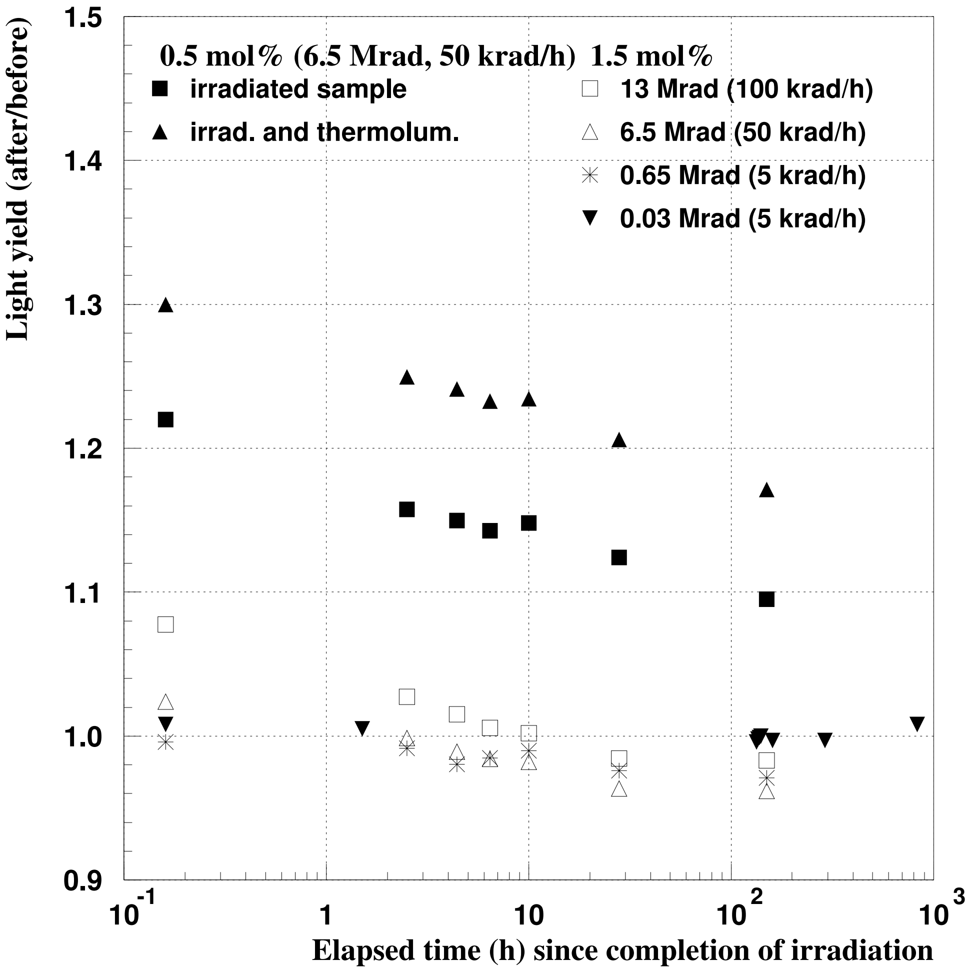

Figure 8: The light yields normalized to that before exposure to ^{60}Co γ-rays. The two 0.5 mol% samples had been irradiated to 1 Grad about a year ago (one of them was subjected to a thermoluminescence test), for which light yields are normalized by those measured before the present irradiation.

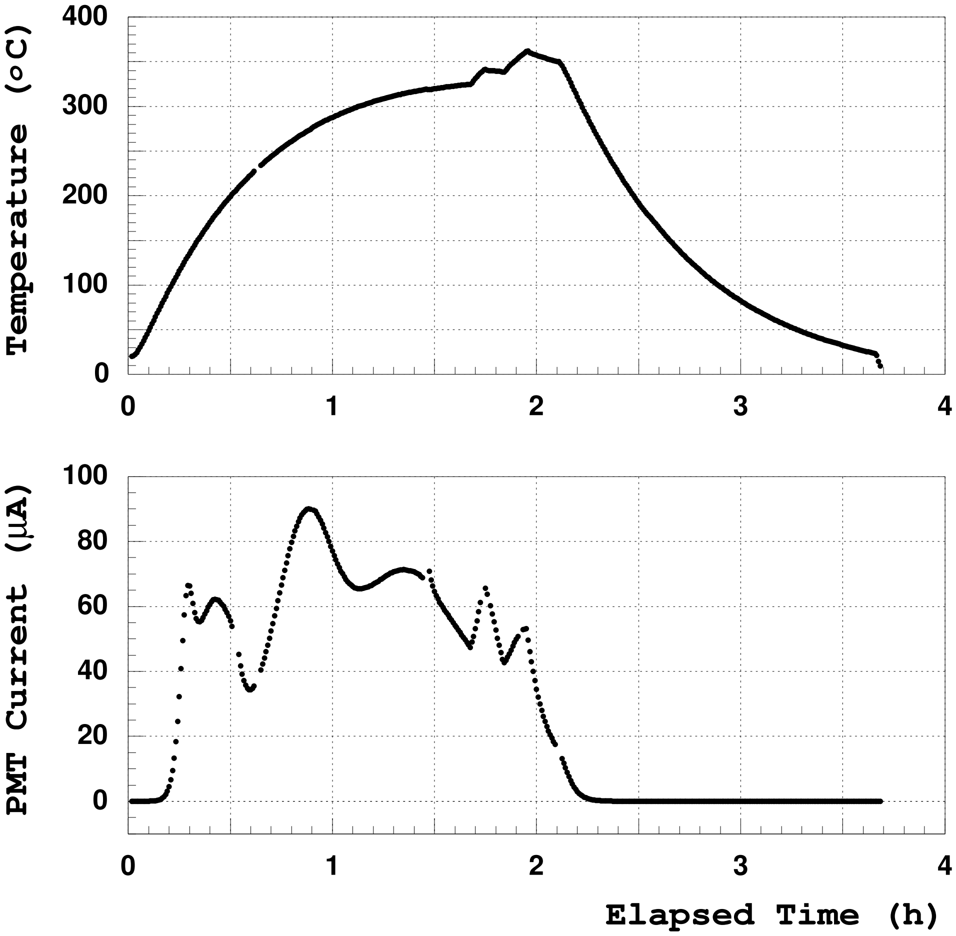

Figure 9: Thermoluminescence out of an irradiated sample (0.5 mol% Ce concentration). (top) Monitor temperature and (above) PMT current as a function of the elapsed time.

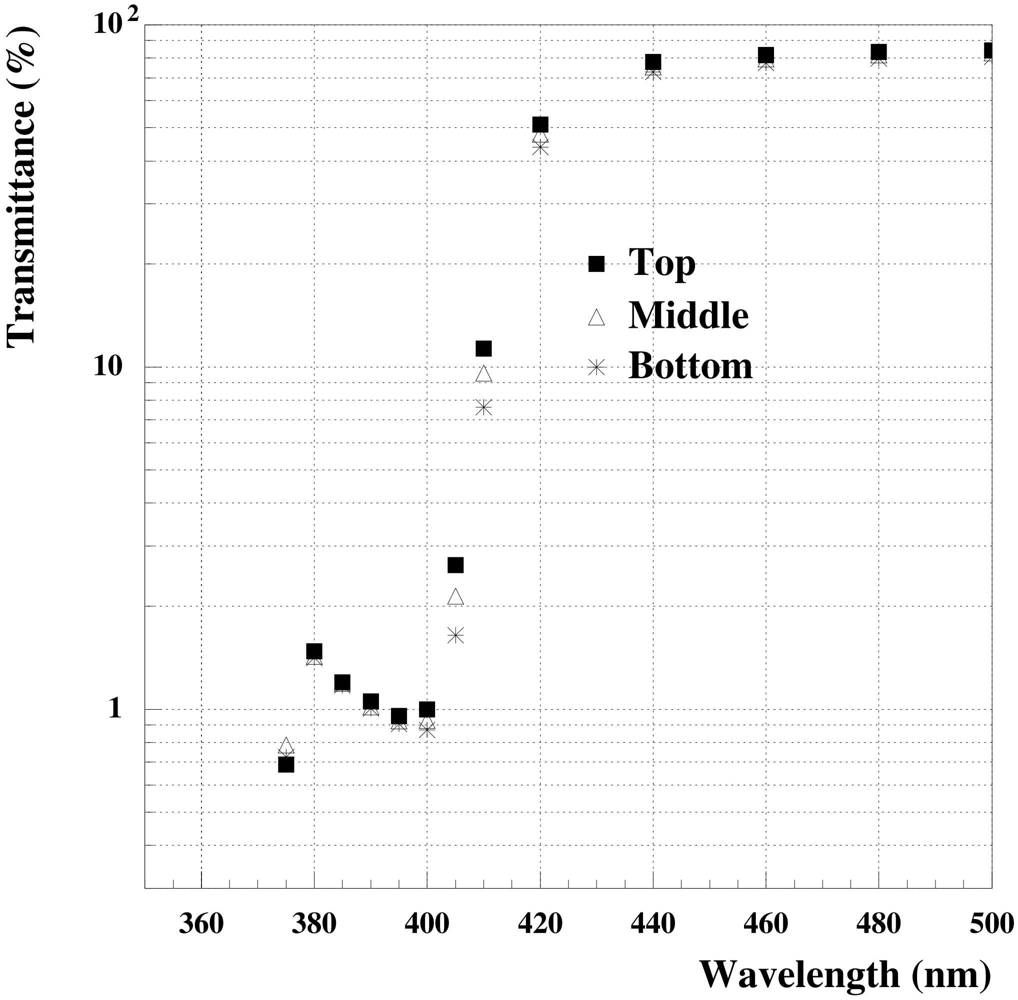

Figure 10: Transmittance of a large 1.5 mol% GSO:Ce crystal measured across the crystal (2.5 cm) at three positions along the crystal.

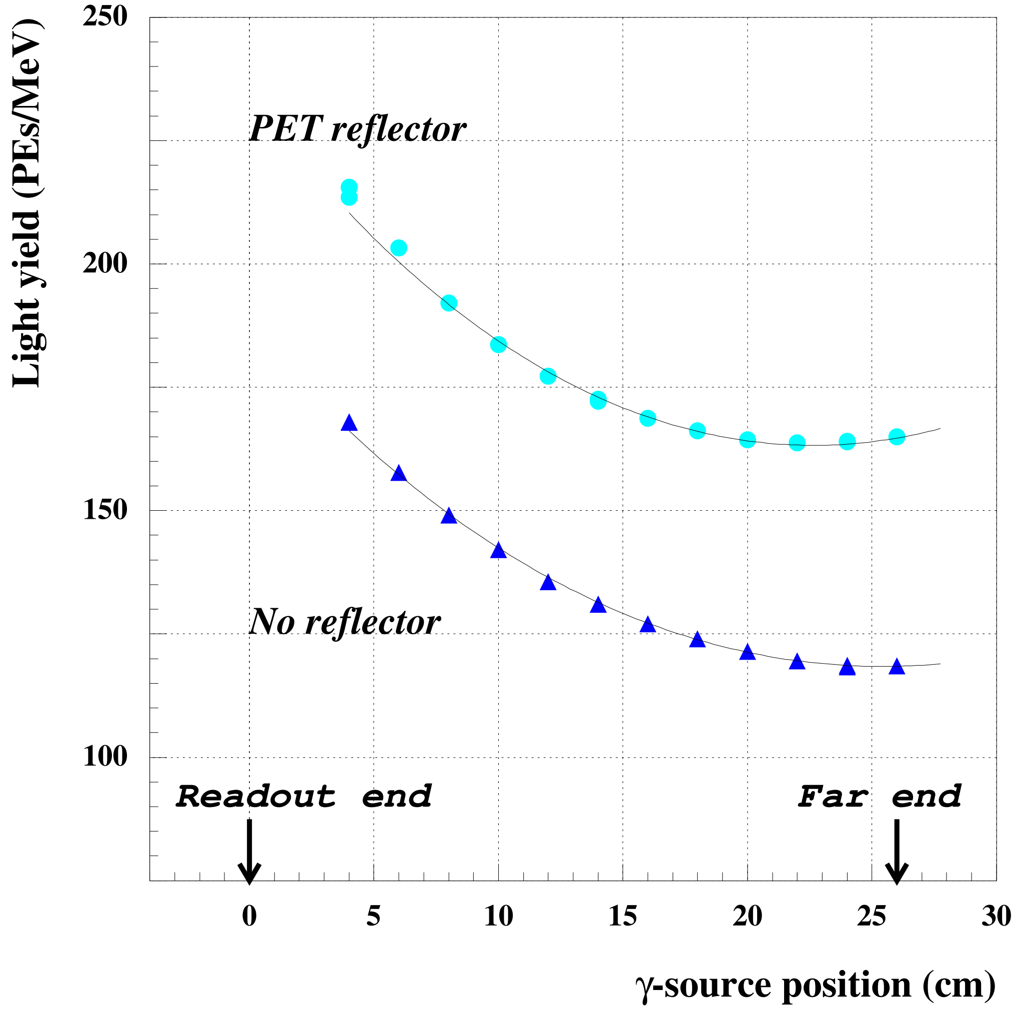

Figure 11: Light yield of a large 1.5 mol% GSO:Ce crystal measured at positions along the crystal. The signal is read out from the Bottom end (see text).