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Multifunctional scintillation materials of the garnet structure for non-homogeneous detecting cells of electromagnetic calorimeters to operate in a harsh irradiation environment

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Abstract. The scintillator gadolinium aluminium gallium garnet (Gd₃Al₂Ga₃O₁₂, GAGG) was found to be an excellent material for application in non-homogeneous detecting cells for future calorimeters, operating in a harsh irradiation environment. GAGG, activated by cerium ions, can be used to detect γ -quanta and to absorb efficiently neutrons in a wide energy range. The capture of neutrons is accompanied by the emission of relatively soft γ -quanta which can be ignored at calorimetric measurements above 10 MeV. These findings create prospects to construct compact detectors for different purposes, from particle physics to industry.

1. Introduction

The irradiation environment of experiments to be expected at novel colliders will be harsh enough to limit the long term operation of homogeneous detector calorimeters. This will occur due to the accumulation of the damage caused by various effects, particularly due to the hadron component of the irradiation. Heavy crystalline materials are widely used in homogeneous detecting cells of calorimetric detectors of the current and planned high energy physics experiments. Note, bismuth germanate Bi₄Ge₃O₁₂ (BGO) and lead tungstate PbWO₄ (PWO), were successfully used to construct and exploit the electromagnetic calorimeters of the L3, CMS and ALICE collaborations at LEP and LHC accelerators at CERN [1]. However, self-activated scintillation materials show significant damage under hadron irradiation [2, 3]. Non-homogeneous calorimetric detecting cells, consisting of absorber and low-volume scintillation elements as plates or fibers become very promising in this view. So far, such detecting modules used plastic scintillators, what made detecting cells relatively cheap and competing to construct large area devices. However, the damage of the plastic scintillator matrixes remains tolerable for fluences below 10^{14} p/cm² [4], before a dramatic change of the optical transmission of the plastic scintillators is observed. Therefore, a replacement of the plastic scintillator with more radiation hard material has to be considered. Here we just mention the two most popular designs: "spaghetti" and "shashlyk" type detecting cells, combining heavy metal absorber with scintillators.

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Among considered alternative materials, scintillators of garnet structure show several preferences. They can be produced in a single crystalline or ceramic form; their atomic composition can be varied to meet the requirements of the individual application. Gadolinium aluminum gallium garnet $Gd_3Al_2Ga_3O_{12}$, (GAGG), activated by cerium ions, can be used to detect electromagnetic probes and to absorb neutrons in a wide energy range. The capture of neutrons is accompanied by the emission of relatively soft γ -quanta which can be practically ignored. Comparison of the properties of radiation hard scintillation crystals is listed in Table 1. Further details about selected scintillators are described elsewhere in [2].

Material	Density	Z _{eff} / photo absorp.	Yield	Decay time	Luminescence
	g/cm ³	coeff. at 511 keV / cm ⁻¹	photons/MeV	τ_{sc} / ns	maximum
		radiation length / cm			$\lambda_{max} \ / \ nm$
Gd ₃ Al ₂ Ga ₃ O ₁₂ :Ce (GAGG)	6.67	50.6/0.12/1.61	46 000	80/800	520
(Gd-Y) ₃ (Al-Ga) ₅ O ₁₂ :Ce	5.8	45/0.08/1.94	60 000	100/600	560
Y ₃ Al ₅ O ₁₂ :Ce (YAG)	4.55	32.6/0.017/3.28	11 000	70	550
YAlO ₃ :Ce (YAP)	5.35	32/0.019/2.2	16 200	30	347
(Y _{0.3} -Lu _{0.7})AlO ₃ :Ce	7.1	60/0.21/1.3	13 000	18/80/450	375
(LuYAG)					
Lu ₂ SiO ₅ :Ce (LSO)	7.4	66/0.28/1.1	27 000	40	420
(Lu-Y) ₂ SiO ₅ :Ce (LYSO)	7	60/0.20/1.35	30 000	37	420

Table 1. Scintillation properties of inorganic scintillators suitable to construct non-homogeneous detecting cells of electromagnetic calorimeters.

In addition to measurements of the shower energy these materials can be used for MIP detection to improve timing measurements. A comparison of MIP signals created in a scintillator of 1 mm thickness is shown in Table 2. One can see that the highest yield per MIP is obtained in GAGG crystals.

Material	Yield,	dE/dx for	Yield,		
	photons/MeV	10 GeV e ⁻ , MeV/mm	photons per 1 mm per MIP		
Plastic scintillator	10 000	0.154	1540		
(vinyltoluene based)					
$Y_{3}Al_{5}O_{12}$ (YAG)	11000	0.591	6500		
Y ₃ (Al _{0.5} -Ga _{0.5}) ₅ O ₁₂ (YAGG)	30000	0.614	18420		
YAlO ₃ (YAP)	16000	0.708	11350		
$Gd_{3}Al_{2}Ga_{3}O_{12}$ (GAGG)	46000	0.808	37200		
Lu ₂ SiO ₅ (LSO)	27000	0.879	23700		
(Lu _{0.8} -Y _{0.2}) ₂ SiO ₅ (LYSO)	30000	0.85	25500		

 Table 2. Light output per MIP (10 GeV e⁻) per 1 mm thickness in different scintillation materials.

Moreover, multi-doped GAGG crystals were found to be suitable to operate at temperatures below 0° C, which is well adapted to operate with silicon photo-multipliers (SiPM) to reduce the intrinsic noise level [5]. On the other hand co-doping of the crystal with Mg and Ti leads to strongly suppression of the phosphorescence and afterglow is reduced by factor 100 to the GAGG crystal solely doped with Ce. The scintillation decay is nearly 60 ns. So, one should not expect saturation up to a few tens MHz of the incident flux of the particles. It is reasonably better than for 3He based counters, for example.

In this report we describe the scintillation properties and radiation hardness of GAGG scintillators in the single crystalline form. Results of the simulation of the light output for detecting cells of different design are also presented.

2. Samples and measurements

The test samples were supplied by FOMOS Crystals (Moscow, Russia) doped with Ce and co-doped with Mg and Ti [5] and grown by the Czochralski method in iridium crucibles. Samples 1, 2 and 3 had the same amount of Ce and different concentrations of co-dopings. The ingots were annealed on air before production. To characterize the radiation damage in the crystals, the optical transmission was measured before and after irradiation with the spectrophotometer Varian 4000. All measurements were performed at room temperature.

The samples were irradiated with a strong ⁶⁰Co source ($\langle E_{\gamma} \rangle \approx 1.25$ MeV) up to different integral doses with a rate of 1.4 Gy/minute. The transmission has been measured in 2 minutes after γ -irradiation. Similar samples have been irradiated by 24 GeV protons at the CERN Proton Synchrotron (PS) at the flux of 10⁹ p/cm²/sec up to the fluence of $3.1 \cdot 10^{15}$ p/cm² by LHCb collaboration. Crystals showed perfect tolerance to proton irradiation and the detailed results are described in [6].

Samples with a volume of $15x18x7 \text{ mm}^3$ were exposed to a neutron source. Crystal coupled to photomultiplier tube Hamamatsu R2059 was placed in 80 mm distance to the Am-Be neutron source. The activity of the ²⁴¹Am source was 220 GBq corresponding to an estimated neutron yield of $\ge 1.3 \cdot 10^7$ neutron/s.

3. Results and discussion

Figure 1 shows the optical transmission spectra of 7 mm thick samples before and after γ -irradiation. A minor change of the optical transmission is observed. Figure 2 shows the radiation induced absorption coefficient dk, defined as $dk = Ln[T_{before}/T_{after}]/d$ (where T_{before} , T_{after} are the transmittances before and after irradiation and *d* the thickness of the samples, respectively) for the irradiation with an integral dose of 100 and 1000 Gy. All samples demonstrate low induced absorption coefficient at the level below 2 m⁻¹. It corresponds to a reduction of the attenuation length of the material from 1 m (undamaged material) to 0.5 m.





Figure 3 shows the results of the spontaneous recovery of the induced absorption spectrum during one week after irradiation. The crystals were kept in the dark at room temperature. Almost all samples showed full recovery in the range of the emission wavelength.



1000 Gy integral dose and one week of spontaneous recovery at room temperature.

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In order to estimate the impact of the induced absorption on the light output in two different elements of non-homogeneous detecting cells, we performed a LITRANI B simulation of the relative light output of scintillator plates plate with dimensions of 11x11x2 mm and a fiber bar with dimensions 3x3x48 mm at different levels of induced damage and considering photo detectors similar to SiPMs, produced by the FBK company [7], with an sensitive area of 4x4 and 2.8x2.8 mm², respectively.

The relative output, detected by the photosensor at the optical window, was evaluated for different positions of the scintillation excitation along the long side of each element. The distance between positions, where scintillation was excited, was 1 mm in the plate and 5 mm in the bar respectively. The results of the simulation are presented in figures 4 and 5. We did not observe any change of the plate response, when an additional absorption due to irradiation damage was implemented. Therefore, the change of the attenuation length from 1 m to 0.5 m does not significantly affect the light output obviously also due to the small thickness of the element.



It seems that the configuration using a scintillator bar is more sensitive to the optical transmission damage. The non-uniformity of the light output, depending on the position of excitation, is practically not changed like in case of the plate. However, a drop of the light output of nearly 20% is observed when the attenuation length is decreased to 0.5 m.

Finally, figure 6 shows γ -spectra experimentally measured with a GAGG:Ce (15x18x7 mm³) sample irradiated with neutrons from the Am-Be source. The spectra were measured either with lead discs of a total thickness of 4 cm and a 3 mm thick copper disc placed between detector and source or secondly without any material placed in between detector and source. The spectrum of a ¹³⁷Cs source (662 keV gamma line) was recorded for energy calibration.

Besides a continuous spectrum, a few groups of distinct lines at energies of ~90 keV, 190 keV, 511 keV, and in the energy range from 900 keV to 1200 keV have been observed. The 4.44 MeV γ -line from the Am-Be source is seen when no Pb-shield was used. As seen, γ -lines, generated in GAGG material under neutron irradiation are below 1.5 MeV energy and can be easy discriminated in the detector by appropriate choice of the threshold.



Figure 5. Simulated relative light output in the GAGG bar measured from one end with SiPM readout depending on the position of the scintillation light excited by an particle (100 keV e⁻): blue points: attenuation length 1 m, red points: 0.5 m and green points: 0.2 m. A sketch of the simulated detector unit is included.



4. Conclusion

Our modeling and experimental evaluations have shown that GAGG scintillation material, doped with Ce and co-doped with different ions, is suitable to operate in a harsh irradiation environment. It may

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be material of a choice to construct detecting cells of "shashlyk" or "spaghetti" types of nonhomogeneous electromagnetic calorimeters and other detectors, where fast response to MIPs is considered. GAGG is found to be even a prospective material in applications below High Energy Physics to detect and identify neutrons up to moderate energies below 20 MeV. The gamma-lines acquired with GAGG scintillation detector under neutron irradiation are located in the energy range up to 4 MeV with major γ -lines concentrated in the energy range below 0.6 MeV. Finally, the energy deposition of MIPs in 2 mm thick GAGG crystal plate amounts close to 1.5 MeV. Therefore, the overlapping with γ -quanta generated by thermal and low energy neutrons will be negligible.

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