Investigation of some scintillation properties of YAG : Ce crystals

T. Ludziejewski, M. Moszyński, M. Kapusta, D. Wolski, W. Klamra, K. Moszyńska

*Soltan Institute for Nuclear Studies, PL 05-400 Świerk-Otwock, Poland

University of Warsaw, Institute of Experimental Physics, Hoża 69, PL 00-681 Warsaw, Poland

Royal Institute of Technology, Department of Physics, Frescati, S-104 05 Stockholm, Sweden

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Abstract

A light output expressed in the photoelectron (phe) number, a relative light yield for α-particles and light pulse shapes for γ-rays and α-particles excitation of the YAG : Ce crystals with different Ce doping between 0.012 and 1.08 mol% were studied. The highest light output of 1420 ± 70 phe/MeV was measured for the sample doped with 0.21 ± 0.03 mol% Ce, while the fastest pulse was observed for the sample doped with 1.08 ± 0.08 mol% Ce showing, however, lower light yield by 10%. The relative light yield for α-particles of 0.21 ± 0.03 as compared to γ-rays was approximately the same for all the samples studied. In conclusion, the YAG : Ce crystal doped with about 1 mol% of Ce is the most appropriate for different applications. For this crystal the α–γ discrimination by digital charge comparison method was studied. A clear separation was observed with the YAG crystal coupled to the XP2020Q photomultiplier for 5.49 MeV α-particles from 241Am source and 6.04 MeV as well as 8.77 MeV from ThC source. However, this separation was inferior compared to that known for CsI(Tl).

1. Introduction

In our previous work [1] we have studied the basic properties characterising Ce-doped yttrium–aluminum garnet (YAG) scintillator as a detector for γ-rays and α-particles. The host material, well known from laser physics, has a good mechanical properties, is chemically inert and not hygroscopic [2]. The relatively high light yield of the order of 20 000 photons/MeV, the two-component decay of the scintillation pulse with the decay time constants of 88 and 300 ns, respectively, and the significant difference of the light pulse response for γ-rays and α-particles suggested good performance of the YAG : Ce crystal in various applications, mainly for detection of light charged particles and their expected identification by the pulse shape discrimination method.

Simultaneously, the studies of Ref. [1] indicated possible application of the YAG crystal in experiments with photodiode light read out. Due to the strong crystal field caused by the high symmetry of the YAG, its emission spectrum is shifted to the green region (maximum wavelength of emission at 550 nm) [2,3]. This rather unique feature within the other Ce-doped scintillators predestines YAG : Ce crystal for the measurements with silicon photodiodes for which the region of maximum sensitivity matches the emission spectrum. Therefore, the light released by the crystal can be effectively converted to the electron–hole pairs; see Ref. [1]. An important advantage of the YAG : Ce crystal over CsI(Tl), conventionally used in this case, is a much faster light pulse. The potential applications of YAG scintillator are, however, limited by its relatively low density of 4.55 g/cm³ and low atomic number of yttrium (Z = 39) and, consequently, the low stopping power for γ-rays [4]. Nevertheless the scintillation properties investigated in Ref. [1] indicate that YAG : Ce can serve as a useful scintillator for light charged particles detection in nuclear physics experiments and is potentially a versatile substitute of CsI(Tl) and GSO crystals. The high stopping power of new crystals for charged particles makes them attractive in nuclear reaction studies [5]. GSO was recently chosen in the Scandinavian project CHICSI [6] as the E detector in telescopes used to identify different particles and heavy ions of intermediate energy.

The aim of this work was twofold. First, by investigating the scintillation properties of the YAG crystal, which depend on the Ce concentration, we wanted to establish the condition for the optimal speed and light output of

* Corresponding author. Tel.: +48 22 779 9531; fax: +48 22 779-3481; e-mail: marek@sunp3.cyf.gov.pl.

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YAG scintillator. It was also expected that an analysis of the light pulse shapes for γ-rays and α-particles would help to establish the best conditions for particle identification by the pulse shape discrimination method. Then, in the second part of the work, the pulse shape discrimination capabilities of the YAG crystal were studied.

2. Experimental details

Four samples of the Ce-doped YAG (Y₃Al₅O₁₂) crystal were investigated in the present study. Three of them were obtained from the Lebedev Physical Institute in Moscow, while the last sample was delivered from the Preciosa Co (Turnov, Czech Republic). All crystals were grown by the Czochralski method from melts to which CeO₂ was added. Three pieces with the dimension 10 × 10 × 2 mm³ were cut from the single crystal and polished from one side. The last sample with the same dimensions was polished on two sides.

In spite of the fact that the relative amount of Ce added to the melt by the manufacturers was known, the optical properties (different yellow colouring) of samples cut from the opposite sides of the single crystal suggested an inhomogeneous distribution of Ce dopant within the crystal volume. Therefore, we decided to recheck the nominal Ce concentration given by the manufacturer, through direct measurement. The crystals were analysed for Ce content using a standard X-ray fluorescence analysis technique. For the excitation of characteristic X-rays of cerium and yttrium the 59.6 keV γ-line from 241Am source was used. The Ce concentration was found to be 1.08 ± 0.08, 0.21 ± 0.03, 0.015 ± 0.07 and 0.012 ± 0.08 mol%, respectively.

In the measurements characterising the basic properties of YAG scintillation (e.g., the light output or α/γ light output ratio), the XP2020Q photomultiplier with the radiant photocathode sensitivity of 74 mA/W at 401 nm was used. For the measurements with γ-rays the crystals were wrapped with Teflon tape, while for α-particles the source simultaneously served as a reflector.

Table 1
The number of photoelectrons measured with the tested YAG:Ce crystals

<table>
<thead>
<tr>
<th>Ce content [mol%]</th>
<th>YAG:Ce</th>
<th>YAG 1</th>
<th>YAG 2</th>
<th>YAG 3</th>
<th>YAG 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.012 ± 0.08</td>
<td>1000 ± 50</td>
<td>1180 ± 60</td>
<td>1420 ± 70</td>
<td>1270 ± 60</td>
<td></td>
</tr>
<tr>
<td>0.015 ± 0.07</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.21 ± 0.03</td>
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<td></td>
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<td></td>
</tr>
<tr>
<td>1.08 ± 0.08</td>
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</tbody>
</table>

1000 phe/MeV for x = 0.012 mol% of Ce to the maximum of 1420 ± 70 phe/MeV for x = 0.21 ± 0.03 mol% and is slightly lower for the maximum investigated Ce content of 1.08 ± 0.08 mol%. Such saturation of the light output is characteristic for doped scintillators [7], and indicate the existence of efficient energy transfer from the primary electron–hole excitation to the active cerium centres. The observed decrease of the light output for the 1.08 mol% doped crystal is similar to that reported in Ref. [8] for GSO:Ce.

The number of photons emitted by the tested scintillator can be determined assuming the 100% light collection efficiency in the crystal, a full photoelectron collection in the photomultiplier and exploiting the quantum efficiency characteristic of the XP2020Q photomultiplier. Light yield was determined recently for a number of inorganic scintillators [9] including YAG crystal doped with 1.08 mol% Ce. The light yield of this sample was determined as 16700 ± 660 photons/MeV, somewhat lower than that found in Ref. [1]. However, for our discussion we use the number of photoelectrons per unit energy, since this quantity is more easily measured and reflects well relative light yield of different samples.

The moderate number of photoelectrons produced by YAG crystals is the result of the emission spectrum peaking at 550 nm, in the region where the quantum efficiency of the bialkali photocathode is low. However, in the case of Si-photodiodes the situation is more preferable. The number of electron–hole pairs (e–h) measured in Ref. [1] (15000 ± 750 e–h/MeV) places YAG crystal among the best scintillators. Its absolute light output measured in Ref. [9] of 16 700 ± 660 ph/MeV corresponds to about 43% of that of NaI(Tl) [10].

3. Results

3.1. Light output

The number of photoelectrons produced by the tested scintillators under the γ excitation was determined by comparing the position of the full energy peak of 661.6 keV γ-rays from ⁵⁸⁷Cs source with that of the single photoelectron peak. The results of the measurements are tabulated in Table 1. It shows that the largest light output, expressed in terms of photoelectron yield, is observed for the samples with 0.21 ± 0.03 mol% Ce content. The measured light output increases from about

3.2. Relative light output of α-particles

The relative α/γ light output was measured comparing the position of the peak due to 5.49 MeV α-particles from a ²⁴⁴Am source and that of 661.6 keV full energy peak of γ-rays from a ⁵⁸⁷Cs source measured in the same conditions. For the investigated range of Ce doping, the α/γ ratio was found to be approximately constant and equal.
to 0.21 ± 0.03. This rather low $\alpha/\gamma$ ratio for YAG as compared to CsI(Tl) or NaI(Tl) crystals (equal to 50% and 45%, correspondingly) can be attributed to the concentration quenching caused by the high ionisation density of the $\alpha$-particles and is typical for the new inorganic crystals. The $\alpha/\gamma$ ratio equal to 0.22 was found recently for a LuAP crystal [11], a similar quantity of 0.21 ± 0.03 was measured for GSO:Ce crystal [12] and 0.24 for GSAG crystal [13]. An even more significant reduction of the light output for $\alpha$-particles has been observed recently for LSO:Ce [14] where the $\alpha/\gamma$ ratio is of the order 0.12.

### 3.3. Light pulse shape study

The light pulse shape studies were performed by means of a single photon method. The XP2020UR photomultiplier was used to record single photons. The uncoated YAG:Ce crystals were optically coupled to the XP2020Q photomultipliers and irradiated by 661.6 keV $\gamma$-rays from $^{137}$Cs source or 5.49 MeV $\alpha$-particles from $^{241}$Am source.

Fig. 1 shows the light pulse shapes measured for the YAG crystal doped with 1.08 mol% of Ce under $\gamma$-ray and $\alpha$-particle excitation. Both pulse shapes are normalised to match the intensities of the far tail of the light pulse.

$$I(t) = A_1 \exp(-t/T_1) - A_2 \exp(-t/T_2) + A_3 \exp(-t/T_3)$$

(1)

where $T_f$ is the decay time constant of the fast component, $T_r$ the rise time constant of the initial part of the light pulse shape, $T_s$ the decay time constant of the slow component and $A_1$, $A_2$ and $A_3$ are the corresponding intensities of the pulse shape components. The dashed lines in Figs. 2 and 3 represent the decays of both the components.

In Figs. 4 and 5 the decay time constants of both the components are plotted versus Ce concentration, for the light pulses measured with $\gamma$-rays and $\alpha$-particles excitation, respectively. Fig. 6 presents the relative intensities of the fast component versus Ce content for $\gamma$-rays and $\alpha$-particles excitation. A general trend of faster pulses with the growing Ce concentration is observed. The decay time constants of both the components are decreasing while the relative intensity of the fast component is increased. The last effect is particularly important for $\gamma$-rays. The YAG crystal doped with about 1 mol% of Ce will be preferred in some applications because it results in the fastest light pulse, in spite of the fact that somewhat lower total light output is observed for this sample (see Section 3.1). This crystal also shows the largest difference in the intensity of the fast component between $\gamma$-rays and $\alpha$-particles. Thus, it is the best sample for the tests of the YAG crystal capabilities in pulse shape discrimination method.

Fig. 1 shows light pulse decay curves for $\alpha$- and $\gamma$-excitations measured for the sample 4, normalised in the...
Fig. 2. Light pulse shapes from YAG:Ce doped with 0.21 mol% Ce due to γ-rays and α-particles. The solid lines show the results of the fit of Eq. (1). The dashed lines represent the decay of both the components.

Fig. 3. Light pulse shapes from YAG:Ce doped with 0.015 mol% Ce due to γ-rays and α-particles. The solid lines show the results of the fit of Eq. (1). The dashed lines represent the decay of both the components.

Fig. 4. The decay time constant of the fast component versus Ce concentration in YAG crystals for γ-ray and α-particle excitation.

Fig. 5. The decay time constant of the slow component versus Ce concentration in YAG crystals for γ-ray and α-particles excitation.

region of slow components. A simple subtraction of those two curves is presented in Fig. 7 and seems to indicate an exponential light decay described well by the two exponential equations. The effect of this same procedure for
Fig. 6. The relative intensity of the fast component of the light pulse versus Ce concentration.

the crystal doped with 0.21 mol% of Ce is exhibited in Fig. 8. In both cases, a rise time constant of about 10 ns was extracted, while the decay time constant approaches those of the fast component found above for α-excitation. Fig. 9 shows the relative contribution of this component to the γ-pulse plotted versus Ce concentration. The figure exhibits a continuous increase of this component intensity with the Ce content. The physical meaning of the observed component is difficult to understand. By assuming that the fast component of the light pulse due to α-particles corresponds only to the primary light emission from the YAG crystals, we can speculate that the observed components (see Figs. 7 and 8) represent the delayed component of the light induced by the energy transfer process from the crystal lattice-to-activator. The same process is known to be responsible for the slowing down of the light pulse from GSO as it was discussed in Ref. [15]. The observed component of the light pulses may suggest that it presents the time structure of this delayed component, as expected in Ref. [15].

4. Pulse shape discrimination

The pulse shape discrimination capabilities of YAG crystal were tested for α-γ separation with the sample doped with 1.08 mol% Ce coupled to the XP2020Q photomultiplier. The pulse shape discrimination was studied by means of digital charge comparison method, following that used for n-γ discrimination with liquid scintillators [16,17]. Fig. 10 presents the block diagram of the experimental arrangement. The anode signal was split and sent to a constant fraction discriminator (CFD) and to a charge-to-voltage converter (QVC) [18] used previously for n-γ discrimination [16]. The output signal of the CFD was used to produce a gate with adjustable delay and width. A typical adjustment of the gate in relation to the anode signal at the inputs of the QVC is presented in the bottom of Fig. 10. The best conditions for α-γ discrimination were achieved when selecting the charge corresponding to the fast component of the light pulse. To get a signal proportional to the energy, the last dynode signal was sent to a standard spectroscopy amplifier. Both output signals were sent to the two-parameter data acquisition system based on CAMAC and a PC computer [19].

All tests were carried out with α-particles from 241Am and ThC sources. Fig. 11(a) presents the two parameter
Fig. 8. The result of the subtraction of the light pulse due to α-particles from that of γ-rays for the YAG crystal doped with 0.21 mol% Ce. Both light pulses were normalised at the late tail. The solid line is the result of the fit of a two exponential equation.

Fig. 9. The relative intensity of the component showed in Figs. 7 and 8 versus the Ce concentration.

Fig. 10. The block diagram of the experimental arrangement used for the digital charge comparison method for α-γ discrimination. At the bottom of the figure, a typical position of the delayed gate in relation to the anode pulse is presented.

spectrum observed with 5.49 MeV α-particles from $^{241}$Am source and γ-rays from $^{60}$Co source. Fig. 11(b) shows a one parameter spectrum of α-γ discrimination corresponding to the gate at the 5.49 MeV α-peak. The evidence of α-γ discrimination is, however, much weaker than that measured with CsI(Tl) [20]. No doubt that the quality of α-γ discrimination is limited by the low number of photoelectrons produced by YAG scintillator in bialkali photocathode of XP2020Q. Thus, better results are expected when a photomultiplier with extended red S20R photocathode having maximum sensitivity shifted to the green region of the wavelengths is used.

The difference between the time constants for the two components of the decay in the YAG crystal is much smaller than for crystals like CsI(Tl) or BaF$_2$, crystals known for very good charged-particle separation properties. Consequently, a gate on the fast YAG component may also contain a contribution from the slow component, which in turn may result in worse pulse shape discrimination.

Somewhat better pulse shape discrimination is shown in Fig. 12, where results of the measurements done with ThC source are presented. Both α-particle peaks (6.04 and 8.77 MeV) are well separated from the γ-ray component, see Figs. 12(a) and (d), due to higher number of photoelectrons. To exhibit separation of 8.77 MeV α-particles the γ-component was produced due to the
Fig. 11. Two-parameter spectrum of the charge under the fast component versus total charge showing separation of α-particles from a $^{241}$Am source and γ-rays from a $^{60}$Co source (a). One parameter spectrum corresponding to the gate at the 5.49 MeV α-peak (b).

Fig. 12. Two-parameter spectrum of the charge under the fast component versus total charge showing separation of α-particles from a ThC source and γ-rays from Pu-Be neutron source. (a) Two-parameter spectrum, (b) and (d) one-parameter spectra corresponding to the gates at the 8.77 and 6.04 MeV α-peaks, respectively, (c) energy spectrum of α-particles and γ-rays.

4.4 MeV γ-rays from a Pu-Be neutron source detected in YAG. Fig. 12(c) presents an energy spectrum of α-particles and γ-rays.

5. Conclusions

The present study shows that YAG doped with about 1 mol% Ce is the most appropriate for a broad range of applications as a scintillator. It shows the fastest light pulse with the main decay time constant of 88 ± 4 ns for γ-rays and a high light output of 1270 ± 60 phe/MeV as measured with the XP2020Q photomultiplier. This crystal shows moderate pulse shape discrimination capabilities, as indicated by observing α–γ separation with the digital charge comparison method for α-particles from $^{241}$Am and ThC sources.

References


