Properties of the YAP : Ce scintillator

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Received 22 April 1997; received in revised form 29 August 1997

Abstract

Light yield, light pulse shape due to γ-rays and α-particles, energy and time resolutions for a 10 × 10 × 5 mm³ YAP crystal coupled to the XP2020Q photomultiplier were studied. The measured light output of 17 000 ± 850 photons/MeV includes a correction for the calibrated quantum efficiency of the XP2020Q. The fast component of the light pulse with the decay time constant of 26.7 ± 0.12 ns also shows a finite rise time described approximately by the time constant of 380 ± 45 ps. The YAP crystal which was studied exhibited an energy resolution of 5.7% for 662 keV γ-rays from a ¹³⁷Cs source. This very good energy resolution is due to a low intrinsic energy resolution of 3.4%. These characteristics, together with a good time resolution for ⁶⁰Co γ-rays of 160 ps measured at the threshold of 1 MeV, suggest a broad range of applications for the YAP scintillator. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: Scintillation detectors; Ce doped scintillators; Energy resolution; Fast timing

1. Introduction

The last few years have seen renewed effort to develop new scintillation materials characterized by a high light output, a fast light pulse and a good detection efficiency for nuclear radiation. A recent study [1] identifies several crystalline compounds as potential new scintillator materials.

Among the number of recently proposed new inorganic scintillators [2] the YAP : Ce (yttrium aluminum perovskite, YAlO₃) is of a special interest in many applications. According to Refs. [3–7], YAP : Ce is characterized by a high light output of 40% relative to NaI(Tl), a short decay time constant of 25 ns and a moderate efficiency for γ-ray detection (density of 5.37 g/cm³, and atomic number of yttrium, Z = 39). These properties of YAP and the fact that the peak of its emission spectrum, at 370 nm, is well coupled with the sensitivity of typical photomultipliers suggest potential applications of YAP in γ-ray spectroscopy, nuclear medicine, etc. It is important to note that, according to Refs. [8,9] the energy resolution of YAP is comparable to that of NaI(Tl) and CsI(Tl) with a photodiode. For YAP, this is because the light yield is almost independent of the energy deposited by gamma rays in the crystal [8]. This light yield proportionality is not common for majority of the
scintillators. Indeed, the energy resolution of 7.2% for $^{137}$Cs was reported in Refs. [9,10]. Even better energy resolution (5.9%) was measured recently for YAP coupled to a large area avalanche photodiode [11].

The YAP properties presented above are confirmed by the first applied study of this crystal in nuclear medicine. A YAP matrix has been proposed as a potentially good detector for an animal PET [12] and gamma cameras [13]. A spatial resolution of 1.2 mm FWHM was achieved with a $0.2 \times 0.2 \times 3.0$ cm$^3$ matrix of crystals coupled to position-sensitive photomultipliers [12].

Another important area of application for new scintillators is the detection of charged particles in the study of nuclear reactions [14]. The GSO crystal was recently chosen for the CHICSI project [15], as a part of telescopes used to identify different particles and heavy ions of intermediate energy. The different light pulse shapes of YAG when excited by $\gamma$-rays and $\alpha$-particles [16] result in a pulse shape discrimination capability [17]. In addition, the YAG emission spectrum is well coupled to the sensitivity of Si photodiodes [16]. Thus, YAG can be used for light-charged-particle detection, particularly, in telescopes with $\Delta E$ Si detectors [18]. Consequently, it should also be interesting to study the properties of the YAP for detection of $\alpha$-particles.

While further studies of the YAP crystal are important for its future application, YAP is also the parent scintillator for the newest heavy-crystal LuAP:Ce [19–21] which will likely be of the great importance for applications in $\gamma$-ray spectroscopy and nuclear medicine (PET). All the information gathered on YAP could be applied in the further development of LuAP.

The aim of this work was to study the detection properties of the YAP for $\gamma$-rays and $\alpha$-particles. The light output, light pulse shape for $\gamma$-ray and $\alpha$-particle excitation (including inspection of the rise time of the pulses), energy spectra for $\gamma$-rays and $\alpha$-particles, and finally timing capabilities of YAP were investigated. Special attention was paid to measure light output of the YAP. The quoted light output of 40% relative to NaI(Tl) was measured by comparison with that of thin X-ray NaI(Tl) crystals [3,5] which very often exhibit lower light yield. The number of photoelectrons (phe) measured for the YAP crystal in Ref. [4] of $2700 \text{ phe/MeV}$ corresponds to 25–30% of a good, small NaI(Tl) crystal ($N = 9000 \text{ phe/MeV}$) (see Ref. [22]). The most recent papers show the light output of $18000 \pm 900 \text{ photons/MeV}$ [23] and $16600 \pm 1300 \text{ photons/MeV}$ [24]. An accurate knowledge of the number of photoelectrons from tested samples allows the discussion of the energy and time resolutions of the studied crystals. According to Ref. [25], the YAP crystal is among the fastest of inorganic crystals and its expected time resolution should be comparable to those of LSO [26] and LuAP [21]. Thus, a precise evaluation of the YAP timing properties seems to be important.

2. Experimental details

All studies were carried out using two samples of YAP with dimensions of $10 \times 10 \times 5$ and $5 \times 5 \times 5$ mm$^3$. The first sample had a Ce concentration of 0.56 mol% and was delivered by Preciosa Co (Turnov, Czech Republic) while the second one had a Ce content of 0.5 mol% and was grown by Union Carbide (USA). The crystals were wrapped with Teflon tape as an optical reflector.

The measurements were performed using an XP2020Q photomultiplier working with the C voltage chain, calibrated for its radiant sensitivity by Philips Photonics [27]; see Fig. 1.

3. Results

3.1. Light output

To determine the light output of the tested crystals, the number of photoelectrons per unit energy (phe/MeV) was measured by comparing the position of the 662 keV full-energy peak from a $^{137}$Cs source with that of the single photoelectron peak. The photoelectron yields of both the crystals and corresponding light outputs are summarized in Table 1. Note, a high photoelectron yield of $4300 \pm 100 \text{ phe/MeV}$ was measured for the sample delivered by Preciosa Co. This yield reflects the significant progress in the quality of YAP achieved
Fig. 1. Quantum efficiency of the XP2020Q photomultiplier no. 40979 as measured by Philips Photonics. The emission spectrum of YAP crystal, as reported in Ref. [13], is shown in relative scale of intensity.

Table 1
Photoelectron yield and light output of YAP and BGO crystals

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Photoelectron yield [phe/MeV]</th>
<th>Light output [ph/MeV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>YAP, 10 x 10 x 5 mm&lt;sup&gt;a&lt;/sup&gt;</td>
<td>4300 ± 100</td>
<td>17000 ± 850</td>
</tr>
<tr>
<td>YAP, 5 x 5 x 5 mm&lt;sup&gt;b&lt;/sup&gt;</td>
<td>3300 ± 80</td>
<td>13200 ± 700</td>
</tr>
<tr>
<td>BGO, 9 x 1 mm&lt;sup&gt;c&lt;/sup&gt;</td>
<td>1190 ± 24</td>
<td>8100 ± 200</td>
</tr>
</tbody>
</table>

<sup>a</sup> Manufactured by Preciosa Co. 0.56 mol% Ce.
<sup>b</sup> Manufactured by Union Carbide, 0.5 mol% Ce.
<sup>c</sup> Manufactured by Bicron, see Ref. [23].
<sup>d</sup> Light output seen by the XP2020Q PMT calculated from the quantum efficiency of the XP2020Q: Q.E. = 25% for YAP crystal and 14.8% for BGO crystal, see Fig. 1.

by this company in comparison to the earlier crystals [3-7]. The value of 3300 ± 80 phe/MeV measured for the other crystal is significantly lower despite the fact that it is doped with a comparable amount of Ce. This decreased yield could be associated with the parasitic absorption of the light which has been observed for YAP samples from Union Carbide and also for LuAP crystals reported in Ref. [28]. This effect seems to be much weaker in the case of the Preciosa sample. The photoelectron yield was confirmed by measuring the number of photoelectrons for the Preciosa sample coupled to the photomultiplier by the side surface of the crystal. The photoelectron yield of 3970 ± 100 phe/MeV, lower by 7%, was observed. Such reduction could be expected generally for standard scintillators because of the less favourable geometry for light collection.

The last column of Table 1 presents the light output (photons/MeV) of the tested crystals as calculated by taking the integral quantum efficiency (convoluted with emission spectrum) of the XP2020Q photomultiplier for the YAP (see Fig. 1) to be 25%. In the last row of the Table 1, the results for the reference BGO crystal (9 mm in diameter and 1 mm thick) are also given following Ref. [23], measured under the same conditions. The calculated light output for the BGO crystal of 8100 ± 200 photons/MeV was corrected further in Ref. [23] for the light collection to be 8500 ± 350 photons/MeV which agrees well with that of 8200 ± 350 ± 400 photons/MeV reported by Holl et al. [29]. The light output for the Preciosa YAP of 17000 ± 850 photons/MeV corresponds to that seen by the photomultiplier for the 5 mm thick sample. To get the absolute light output one has to apply a further correction for the light collection efficiency. For the 5 mm thick YAP sample, the light collection efficiency of 95% was estimated in Ref. [23]. Thus, an absolute light output of 18 000 ± 900 photons/MeV was determined [23], corresponding to about 50% of the light output of NaI(Tl) as measured by Holl et al. [29]. This light yield shows that the quality of YAP crystals has recently improved, as compared to previously tested samples [3-7].

3.2. Light pulse shape

Light pulse shape studies were performed by means of the single-photon method [30, 31] using an XP2020UR photomultiplier [32]. The time response of the system was calibrated by means of the Cherenkov radiation produced in the glass window of the photomultiplier by <sup>60</sup>Co γ-rays following the method used in Ref. [32]. The FWHM of this prompt spectrum was measured to be 450 ps.

An uncoated YAP crystal was optically coupled to the XP2020Q photomultiplier in the reference counter and irradiated from the side by γ-rays from
a $^{137}$Cs source or by $\alpha$-particles from an $^{241}$Am source. The time distributions of the light pulses from the YAP crystal delivered by Preciosa are presented in Fig. 2 for $\gamma$-rays and $\alpha$-particles. Note, a very intense fast component with a decay time constant of $26.7 \pm 0.12$ ns and an intensity of $89 \pm 2\%$, followed by a slow component with the decay time constant of $140 \pm 10$ ns were measured for $\gamma$-ray excitation. The light pulse due to $\alpha$-particles shows the fast component of $24.8 \pm 0.12$ ns with the intensity of $85 \pm 2\%$ and the slow component with the decay time constant of $100 \pm 5$ ns. While the fast component of $\gamma$ and $\alpha$ pulses is almost the same, the decay time constant of the slow component under $\alpha$-particle excitation is faster by about 30\%. A similar effect was observed recently for LuAP [21].

Table 2 shows a comparison of the decay time constants and intensities of both the components of the light pulse measured under $\gamma$-ray excitation with those published in earlier papers. While agreement for the fast component is relatively good (within about 10\%), the agreement is less satisfactory for the slow one. This behaviour suggests that the contribution of the slow component depends on different admixtures of trapping centres associated with the quality of tested crystals.

In Fig. 3 the initial part of the light pulse from the Preciosa YAP sample is shown, as measured with a faster time calibration (48 ps/ch). To the distribution of the experimental data the synthetic pulse of the following form was fitted:

$$i(t) = i_0(t) \left[ -A_1 \exp(-t/T_1) + A_2 \exp(-t/T_2) + A_3 \exp(-t/T_3) \right]$$

(1)

Table 2
Decay-time constants of the light pulse from YAP for $\gamma$-ray excitation

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Fast component</th>
<th>Slow component</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$T$ (ns)</td>
<td>Intensity (%)</td>
</tr>
<tr>
<td>YAP, 10 x 10 x 5</td>
<td>26.7 $\pm$ 0.12</td>
<td>89 $\pm$ 2</td>
</tr>
<tr>
<td>Ref. [3]</td>
<td>27 $\pm$ 2</td>
<td></td>
</tr>
<tr>
<td>Ref. [4]</td>
<td>31.4 $\pm$ 0.1</td>
<td>98</td>
</tr>
<tr>
<td>Ref. [33]</td>
<td>25</td>
<td>87</td>
</tr>
</tbody>
</table>

Fig. 2. Light pulse shape from the YAP crystal with 10 x 10 x 5 mm$^3$ dimensions from Preciosa Co. excited by $\gamma$-rays and $\alpha$-particles.
where \( f(t) \) is the prompt time spectrum of the instrumentation, \( T_1 \) is the rise-time constant of the emission. \( T_2 \) is the decay-time constant of the fast component and \( T_3 \) is the decay-time constant of the slow component. The decay-time constants of both the components found above and their intensities (see Fig. 2), were held constant in the fit. The solid curve in Fig. 3 represents the result of the fit to Eq. (1) using the rise-time constant of 380 ± 45 ps. This result is in a good agreement with the 350 ps rise-time constant measured recently by Derenzo et al. [33]. However, a visual comparison of the shape of the synthetic pulse to the measured one suggests that the description of the light pulse is more complex than that assumed in the calculation.

3.3. Energy spectra

Fig. 4 shows the energy spectrum obtained from the YAP crystal delivered by Preciosa Co, when irradiated with 662 keV \( \gamma \)-rays from a \(^{137}\)Cs source. Note the excellent energy resolution of 5.7\% in FWHM, it is one of the best achievable with scintillation counters. Figs. 5 and 6 present the energy spectra of \( \gamma \)-rays from \(^{22}\)Na and \(^{241}\)Am sources, respectively. Again an excellent energy resolution is observed in the spectra.

The good energy resolution of the YAP crystal seems to be due to a small contribution of the intrinsic resolution of the scintillator itself. In
Fig. 7, the energy resolution of the YAP crystal is plotted versus energy of γ-rays together with the contribution coming from photoelectron statistics, $R_{\text{phe}}$, calculated according to the equation

$$R_{\text{phe}} = 2.35N^{-1/2}(1 + \varepsilon)^{1/2},$$  

where $N$ is the number of photoelectrons (see Table 1), and $\varepsilon$ is the variance of the electron multiplier gain, typically equal to 0.1 for the XP2020Q PMT equipped with the enhanced gain first dynode, see Ref. [9]. The bottom curve represents the contribution of the scintillator itself calculated as the geometric difference of the measured energy resolution and that from photoelectron statistics; see Eq. (2). The low atomic number of yttrium and moderate density of YAP limit the full-energy peak efficiency. This efficiency is dramatically lower than those observed for LSO (a photofraction of 25% for the crystal of 0.29 cm$^3$ volume) [26] and LuAP (a photofraction of 13% for the crystal of 0.05 cm$^3$ volume) [21]. In this respect, YAP is recommended for spectrometry of low-energy γ-rays. For a high-energy range, one has to look for further development of the LuAP crystal [19–21] expecting to get spectroscopic properties similar to YAP.

The excellent spectroscopy properties of YAP are somewhat limited by a low photofraction of 4% observed in Fig. 4. The low atomic number of yttrium and moderate density of YAP limit the full-energy peak efficiency. This efficiency is dramatically lower than those observed for LSO (a photofraction of 25% for the crystal of 0.29 cm$^3$ volume) [26] and LuAP (a photofraction of 13% for the crystal of 0.05 cm$^3$ volume) [21]. In this respect, YAP is recommended for spectrometry of low-energy γ-rays. For a high-energy range, one has to look for further development of the LuAP crystal [19–21] expecting to get spectroscopic properties similar to YAP.

Fig. 8 presents the energy spectrum of 5.49 MeV α-particles from an $^{241}$Am source. The energy spectrum of 662 keV γ-rays from a $^{137}$Cs source is superimposed on the α-spectrum to find the $\alpha/\gamma$ ratio of the light yield. The $\alpha/\gamma$ ratio was calculated from the ratio of the peak positions corresponding to 5.49 MeV energy for α-particles and 662 keV for γ-peak. This quantity of 0.3 ± 0.03 is the highest among the new crystals doped with Ce; see Refs. [17,21,25,26]. This indicates a good work of YAP in spectroscopy of light charged particles and heavy ions. The energy resolution of the peak of α-particles equal to 8.8% is strongly affected by the quality of the source and the fact that measurement
was not done in vacuum. This effect is confirmed by a tail observed on the low-energy side of the peak.

3.4. Timing studies

The timing studies of the YAP crystal were carried out for the 10 x 10 x 5 mm³ sample delivered by Preciosa Co coupled to the XP2020Q photomultiplier. The measurements were carried out on γ-rays from ⁶⁰Co and ²²Na sources. The reference counter consisted of a 25 mm in diameter and 12 mm high BaF₂ coupled to a XP2020URQ photomultiplier. The resulting time resolution (FWHM) of the reference counter, measured in narrow energy windows for both ⁶⁰Co peaks, was equal to 80 ± 3 ps. The 511 keV peak from ²²Na registered 125 ± 4 ps in this study. To increase the accuracy of the quoted time resolution every measurement was repeated three times and the corresponding mean value is used in the following discussion. The time spectra measured here were compared with those measured on LSO [26] and LuAP [21] crystals.

Fig. 9 presents the time spectrum measured with ⁶⁰Co γ-rays and an energy threshold set at 1 MeV. The resultant total time resolution was found to be 180 ± 6 ps, which, when corrected for the contribution of the reference counter (80 ± 3 ps), yields a time resolution (FWHM) for YAP counter of 160 ± 8 ps. This value is essentially the same as those measured for the LSO (160 ps) [26] and LuAP (160 ps) [21] crystals. Fig. 10 presents the time spectrum measured for the annihilation quanta from the ²²Na source. For these measurements, only 511 keV full-energy peak was included in the energy window. The measured time resolution of 260 ± 10 ps, corrected for the contribution of the reference counter (125 ± 4 ps), gives a time resolution of 230 ± 14 ps for YAP. Again, this value is similar to that found in Ref. [21] for the LuAP crystal (233 ps).

To better understand the measured time resolution for YAP one can make a direct comparison to that measured for LuAP in Ref. [21]. The time resolution is approximately proportional to \((T/N)^{1/2}\), where \(T\) is the decay time constant of the light pulse and \(N\) the number of photoelectrons [34]. Taking the measured values for the decay times of YAP and LuAP as 26.7 ± 0.12 and 16.5 ± 0.5 ps, respectively, and their corresponding photoelectron yields as 4300 ± 100 and 2850 ± 60 phe/MeV, respectively, we estimate the value of
(T/N)^{1/2} of 0.079 ± 0.001 and 0.076 ± 0.002 for the YAP and LuAP, respectively. These quantities are approximately the same. This calculation confirms that the time resolution of both YAP and LuAP crystals in the present phase of the development are similar. A similar estimation of the time resolution based on the Hyman theory of timing [35], as those done in Ref. [26] for LSO crystal and in Ref. [21] for LuAP leads to an estimated time resolution of 110 ps for ^{60}Co. According to the above comparison, this latter calculation predicts the same time resolution as that calculated in Ref. [21] for the LuAP crystal. The observed disagreement of the measured time resolution and that estimated from the Hyman theory is associated with the finite risetime constant exhibited by the light pulse from YAP which affects the measured time resolution. Note that the calculations based on the Hyman theory assumed a pure exponential light pulse for the YAP.

4. Conclusions

This study has confirmed earlier observations that YAP : Ce is a fast scintillator with a high light output. An excellent energy resolution of 5.7% for γ-rays from a ^{137}Cs source (one of the best measured with scintillation detectors) and a very good time resolution of 160 ps for ^{60}Co γ-rays with a threshold of 1 MeV (comparable to those measured recently with LSO and LuAP crystals) make YAP the scintillator of choice for a number of applications.

References


