

THE PHOTON NONPROPORTIONAL RESPONSES OF CaMoO₄ AND GAGG:Ce SCINTILLATION MATERIALS

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Abstract

The photon response properties of the GAGG:Ce and the CaMoO₄ scintillation materials have been studied in the energy range from 356 keV to 1,332 keV and were measured with a photomultiplier tube (PMT) number R1306 by Hamamatsu Photonics. The energy resolution, light yield and photon non-proportional responses of CaMoO₄ and GAGG scintillation materials were measured and characterized. The results showed that for all energies, the energy peaks of the GAGG:Ce crystal yielded higher number of counts than those of the CaMoO₄ crystal. Regarding the energy resolution, the light yield and photon non-proportional responses were also determined. It was found that the energy resolutions were inverse proportional to the square root of energy for both crystals. The light yield results for both crystals showed the increased with increasing the energy. Moreover, the results showed that the light yield of the GAGG:Ce crystal was much higher than that of CaMoO₄ crystal. It was also found that both crystals demonstrated good proportional property in the energy range of 356-1,332 keV. The photon non-proportional responses is about 2% for GAGG:Ce, which is better than that of about 4% for CaMoO₄.

Keywords: Scintillation materials, Energy resolution, Light yield, Photon non-proportional response

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Introduction

Abundance of scintillators have been wide-ranging make use of various fields including high energy physics, medical imaging, homeland security, astrophysics and industrial control. The consequential mechanism of a scintillators when high energy photon cross through in the materials, they have taken place ionization which can be generated of light from scintillators. The discovery of new scintillators has been an important subject of research for years (Derenzo *et al.*, 2003). In the present, the requirements in terms of performance of scintillator are more demanding. The energy resolution, light yield, non-proportionality of light yield, time resolution, decay time and determination of the mass attenuation coefficient those were considerably required. These parameters are very important of the research activity and it can be used for development of new scintillation materials (So *et al.*, 2012). The photon non-proportional responses (photon-nPR) means that the total light output of a scintillation material is not precisely proportional to the energy of the absorbed photon energy. The shape of the photon-nPR curve and degree of the photon-nPR is studied as a function of chemical composition, dopant type, index of refraction and other properties of the materials (Khodyuk and Dorenbos, 2012).

This study, the calcium molybdate (CaMoO_4) and cerium doped gadolinium aluminum gallium garnet ($\text{Gd}_3\text{Al}_2\text{Ga}_3\text{O}_{12}:\text{Ce}$ or GAGG:Ce) inorganic scintillation materials were selected for characterization because of still no report about the photon-nPR of both crystals. The calcium molybdate (CaMoO_4) inorganic scintillation material is an issue of interest in searches for neutrinoless ^{100}Mo double beta decay (DBD) because offers very

high Q-value (3,034 keV) (Belogurov *et al.*, 2005). There are several experiments technique to search for the double beta decay processes but scintillation detectors processes was used in these experiment because of easy to fabrication and cost effectiveness. (Dujardin *et al.*, 2018). These experiment were carried out and ongoing at the YangYang underground Laboratory (Y2L), which was originally established for dark matter searches in Korea (Annenkov *et al.*, 2008). For GAGG:Ce scintillation materials, this is a newly developed single-crystal scintillator which has a large light output, high density, longer emission light wavelength and short decay time. That is the widespread to used in the medical imaging technique (PET and SPECT) and gamma spectroscopy where higher energy radiation has to be detected (Yamamoto *et al.*, 2013).

In this research is to report a detailed study on energy resolution, light yield and photon-nPR of CaMoO_4 and GAGG scintillation materials in the energy range of 356 keV to 1,332 keV. The different radioactive source of ^{133}Ba , ^{22}Na , ^{137}Cs and ^{60}Co were chosen for this experiment.

Experiment and Measurements

The study of CaMoO_4 and GAGG:Ce scintillators with the size of $10 \times 10 \times 8 \text{ mm}^3$ and $10 \times 10 \times 10 \text{ mm}^3$, respectively, were measured with a photomultiplier tube (PMT) number R1306 by Hamamatsu Photonics. The background of optical and mechanical properties of both crystal were shown in Table 1. From Figure 1, show a schematic representation of the experimental setup used to measure photoelectron yield and energy resolution.

Table 1. Optical and mechanical properties of the GAGG:Ce and the CaMoO_4

Properties	GAGG:Ce	CaMoO_4
Emission peak (nm)	520	520
Melting point ($^{\circ}\text{C}$)	1,850	1,430
Density (g/cm^3)	6.63	4.20
Decay time (ns)	90	14 (μs)
Refractive index	1.9 (540 nm)	1.98

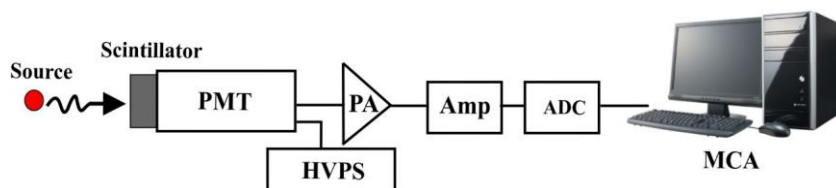


Figure 1. Schematic representation of the experimental setup used to measure photoelectron yield and energy resolution

A photomultiplier tube coupled the crystals to using silicone grease covered with several layers of white Teflon tape and wrapped with black tape, then covered with aluminum housing. The signal output from the PMT anode passed through a Canberra 2007B preamplifier and was sent to Canberra 2022 an amplifier set at 0.5 μ s a shaping time constant. The energy spectra analyzed with software of Canberra MCA to record data. The measurements were carried out with $\times 100$ of CaMoO_4 and $\times 30$ of GAGG:Ce scintillators in the amplifier. The high voltage power supply (HVPS) was used 900 V for CaMoO_4 scintillators and 970 V for GAGG:Ce.

The energy resolution is the ability of a detector to identify particles of different energies, called the energy resolution, $R(E_0)$, is given by:

$$R = \frac{FWHM}{E_0} \times 100\% \quad (1)$$

where E_0 is the energy peak centroid.

The photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) for each gamma peak, was measured by Bertolaccini method (Bertolaccini *et al.*, 1968). In this method the number of photoelectrons is measured by comparing the position of a full energy peak of gamma rays detected in the crystals with that of the single photoelectron peak from the photocathode, which determines the gain of PMT. In order to transform the number of photoelectrons (Nphe) per MeV into an absolute light yield expressed as the number of photons per MeV (ph/MeV) of absorbed gamma ray energy. The overall detection efficiency of the PMT has to be

known. In this study, the number of photons per MeV of absorbed gamma ray energy (Nph/MeV) is calculated by dividing the number of photoelectrons per MeV (Nphe/MeV) of absorbed gamma ray energy by the average quantum efficiency (QE) of the PMT:

$$Nph/MeV = (Nphe/MeV)/QE \quad (2)$$

where QE is the average quantum efficiency of the PMT at the peak wavelength of the emission spectrum for each scintillator. A schematic representation of the experimental setup is shown in Figure 1. The non-proportionality of crystal in percent (%) is the normalized light yield at an energy of 662 keV. That is the light yield for each gamma energy divided by the light yield at gamma energy of 662 keV.

Results and Discussions

The Energy Spectra and Energy Resolution

Figure 2 shows the energy spectra of CaMoO_4 and GAGG:Ce scintillation crystals at the energy of 662 keV. It is seen that the energy peaks of GAGG:Ce crystal yielded higher number of counts than those of CaMoO_4 crystal. This is due to GAGG:Ce crystal has higher mass attenuation coefficient than that of CaMoO_4 crystal over the whole energy region, it can be confirmed by correlating the gamma-ray attenuation as discussed in the Ref. (Chaiphaksa *et al.*, 2016). The energy resolution of CaMoO_4 and GAGG:Ce crystals was determined using Eq. (1) by different radioactive source (^{133}Ba , ^{22}Na , ^{137}Cs , ^{60}Co) in the energy range of 356 keV to 1,332 keV. Figure 3 shows the energy

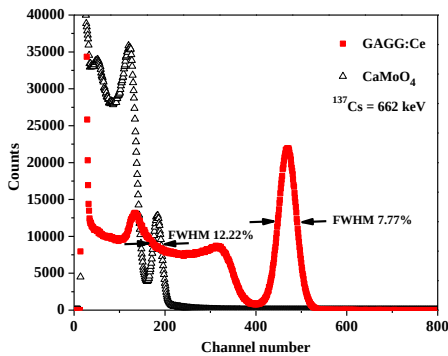


Figure 2. Energy spectra of CaMoO₄ and GAGG:Ce scintillation crystals at the energy of 662 keV

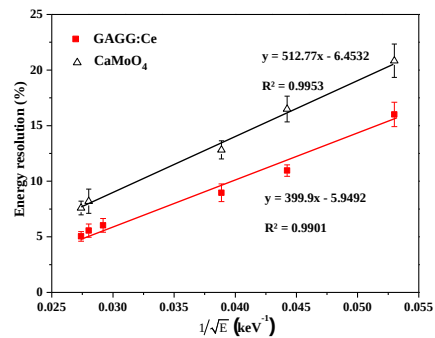


Figure 3. Energy spectra of CaMoO₄ and GAGG:Ce scintillation crystals at the energy of 662 keV

Table 2. Light yield and photon-nPR of CaMoO₄ and GAGG:Ce crystals for photon response

Energy (keV)	Light yield (ph/MeV)		photon-nPR of light yield (% of 662 keV)	
	CaMoO ₄	GAGG:Ce	CaMoO ₄	GAGG:Ce
356	5332.0±426	45659.8±3652	98.0±1.96	98.6±2.85
511	5374.5±322	46020.2±2760	99.3±2.97	99.4±2.40
662	0.54603±490	46290.8±4166	100.0±1.50	100.0±1.50
1173	-	47081.6±2824	-	101.7±2.03
1274	5547.7±277	47166.3±2358	103.9±1.50	101.9±1.52
1332	0.55791±390	47313.1±3785	104.0±2.60	102.2±1.89

resolution as a function of square root of photon energy. The linear relations which are observed for both crystals indicated that the energy resolutions are inverse proportional to the square root of energy. It is very clearly value of energy resolution of GAGG:Ce crystal better than CaMoO₄ crystal all the photon energy range that refer to GAGG:Ce crystal is very high ability of a detector to identify particles of different energies.

Light Yield and Photon Non-Proportional Response

The light yield as determined by Equation (2) and the photon-nPR of CaMoO₄ and GAGG:Ce crystals in the energy range of 356-1,332 keV are shown in Table 2. It is seen that the light yield of GAGG:Ce crystal is much higher than that of CaMoO₄ crystal. The plots of the relationship between the light yield and energy are presented in Figure 4. The graphs showed that the light yield increased

when increasing the energy. Figure 5 shows the photon-nPR of CaMoO₄ and GAGG:Ce crystals in the energy range of 356-1,332 keV as normalized at 662 keV. It can be observed that both CaMoO₄ and GAGG:Ce crystals demonstrated good proportional property in the energy range. For the energy range from 356 keV to 1,332 keV, the photon-nPR is about 2% for GAGG:Ce, which is better than that of about 4% for CaMoO₄. The high proportionality of GAGG:Ce is one of the main reasons behind its good energy resolution (Chewpraditkul *et al.*, 2012).

Conclusions

The photon response properties of the GAGG:Ce and the CaMoO₄ scintillation crystals have been studied in the energy range from 356 keV to 1,332 keV. The results showed that for all energies, the energy peaks of the GAGG:Ce

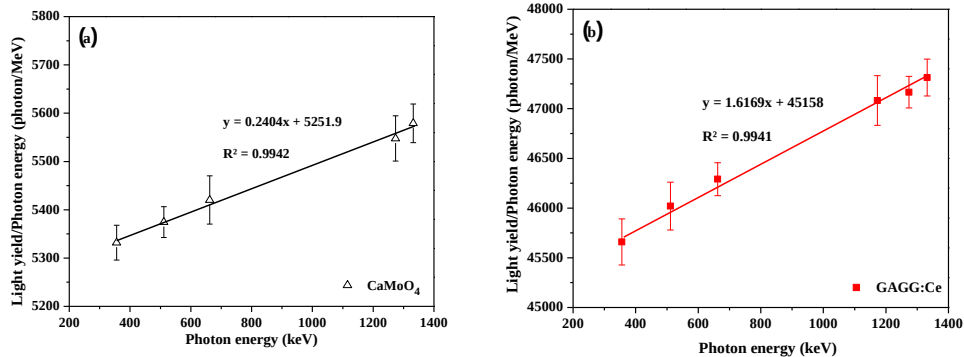


Figure 4. Light Yield of: (a) CaMoO₄ and (b) GAGG:Ce Crystals

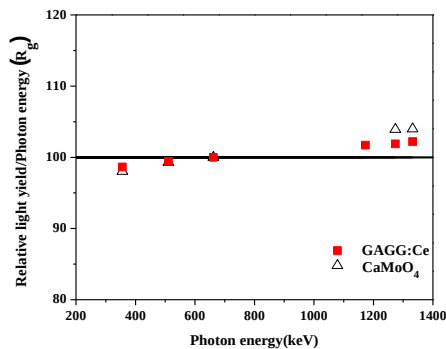


Figure 5. Photon-nPR of CaMoO₄ and GAGG:Ce Crystals in the Energy Range of 356-1,332 keV as Normalized at 662 keV

crystal yielded higher number of counts than those of the CaMoO₄ crystal. Regarding the energy resolution, the light yield and the photon-nPR were also determined. It was found that the energy resolutions were inverse proportional to the square root of energy for both crystals. The light yield results for both crystals showed the increased with increasing the energy. Moreover, the results showed that the light yield of the GAGG:Ce crystal was much higher than that of CaMoO₄ crystal. It was also found that both crystals demonstrated good proportional property in the energy range of 356-1,332 keV. The photon-nPR is about 2% for GAGG:Ce, which is better than that of about 4% for CaMoO₄.

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