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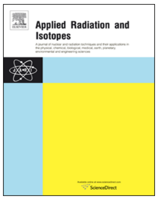

Non-proportionality study of $CaMoO_4$ and GAGG:Ce scintillation crystals using Compton coincidence technique

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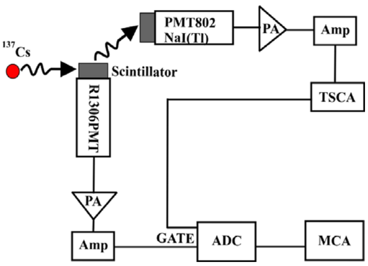
HIGHLIGHTS

- The electron response of CMO and GAGG:Ce crystals were studied using Compton coincident technique (CCT).
- CMO and GAGG:Ce were primary detectors and NaI(Tl) was secondary detector.
- The electron energy resolutions are inverse proportional to the square root of energy.
- The electron energy resolution of GAGG:Ce better than CMO.
- At the energy range from 100.5–435.4 keV, the electron response was slightly decreased at approximately 5% for both crystals.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, the CCT technique and nuclear instrument module (NIM) setup for the measurements of coincidence electron energy spectra of calcium molybdate ($CaMoO_4$) and cerium doped gadolinium aluminium gallium garnet ($Gd_3Al_2Ga_3O_{12}$:Ce or GAGG:Ce) scintillation crystals were carried out. The ^{137}Cs irradiated gamma rays with an energy (E_γ) of 662 keV was used as a radioactive source. The coincidence electron energy spectra were recorded at seven scattering angles of 30°–120°. It was found that seven corresponding electron energies were in the range of 100.5–435.4 keV. The results show that, for all electron energies, the electron energy peaks of $CaMoO_4$ crystal yielded higher number of counts than those of GAGG:Ce crystal. The electron energy resolution, the light yield and non-proportionality were also determined. It was found that the energy resolutions are inverse proportional to the square root of electron energy for both crystals. Furthermore, the results show that the light yield of GAGG:Ce crystal is much higher than that of $CaMoO_4$ crystal. It was also found that both $CaMoO_4$ and GAGG:Ce crystals demonstrated good proportional property in the electron energy range of 260–435.4 keV.

1. Introduction

Scintillation detectors are of particular interests in recent development utilizations in many applications, i.e., inorganic scintillators in science, industry, high energy physics, medical diagnostics, and medical imaging (Istvan et al., 2012). Especially, the

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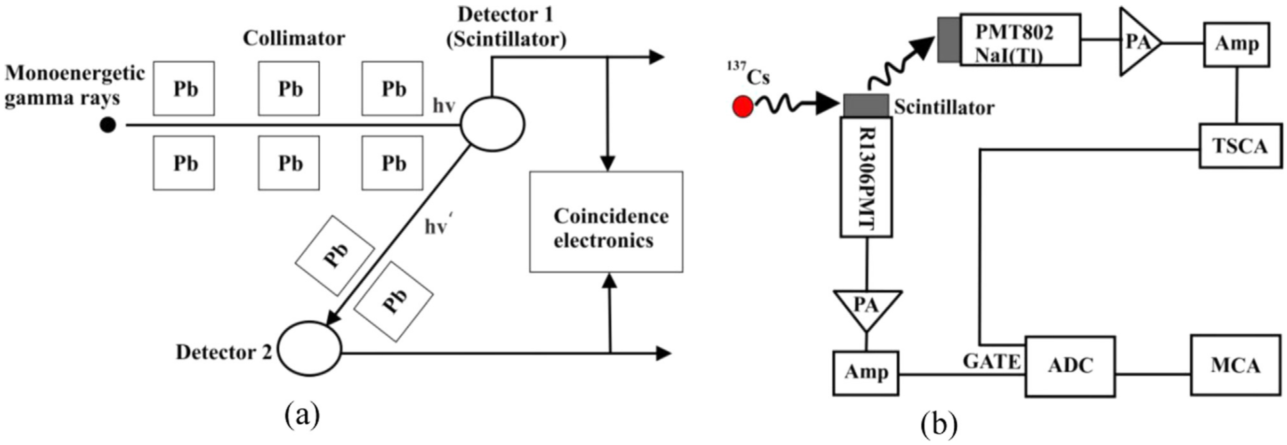

Fig. 1. (a) Schematic of CCT technique and (b) nuclear instrument module (NIM) setup.

development of new scintillation cystals leads to the fast response time (10–100 ns) for time resolution, high light yield, high density, good energy resolution, cost-effectiveness, and high effective atomic number for efficient gamma ray detections. Several techniques can be used for the developments of scintillation detectors. Among these techniques, the Compton coincidence technique (CCT) is one of most precise techniques which the non-proportionality of converting deposited energy to the light is measured. Such technique was developed by Rooney and Valentine in 1994–1996 for measuring the light yield non-proportionality of NaI(Tl) (Rooney and Valentine, 1994,1996). After the implementation of CCT, Taulbee et al. (1997) measured the electron responses with this technique for two “classic” scintillators, i.e., $CaF_2(Eu)$ and $Bi_4Ge_3O_{12}$ (BGO), and two newer scintillators, i.e., $Lu_2SiO_5{:}Ce^{3+}$ (LSO) and $Gd_2SiO_5{:}Ce^{3+}$ (GSO), over the energy range of 5–450 keV. Mengesha et al. (1998) studied the light yield non -proportionality of CsI(Tl), CsI(Na), and YAP, which also based on the measurement of electron responses from the CCT technique. The advantage of CCT technique used in the past decades is due to it allows highly accurate measurement of electron response because the energetic electrons are produced internally in the scintillator by gamma rays.

In collaborations for the Advanced Molybdenum-based Rare process Experiment (AMoRE), the most important material is calcium molybdate ($CaMoO_4$) scintillation crystals. The $CaMoO_4$ scintillation crystal is a subject of interest in searches for neutrinoless ^{100}Mo double beta decay (DBD) (Annenkov et al., 2008; Belogurov et al., 2005). This is because $CaMoO_4$ offers high Q-value (3034 keV) and a reasonable natural abundance (9.63%) that results in manageable costs for enrichment. The enriched crystal can be directly grown from the raw $^{40}Ca^{100}MoO_4$ in the pallet form (So et al., 2012). On the other hand, the $Gd_3Al_2Gl_3O_{12}$: Ce:(GAGG:Ce) scintillation material is the most recent multicomponent garnet-type crystal that demonstrates very high light yield with moderately fast scintillation response. Based on the properties of GAGG:Ce scintillation crystal previously reported by Yeom et al. (2013), the GAGG:Ce crystal is the most commonly used in medical imaging technique and gamma spectroscopy where higher energy radiation has to be detected.

This study, therefore, focused on the CCT technique and nuclear instrument module (NIM) setup for the measurements of coincidence electron energy spectra of $CaMoO_4$ and GAGG:Ce scintillation crystals. The electron energy resolution, the light yield and non-proportionality were also determined and discussed.

2. Theory

2.1. Compton coincidence technique and Compton scattering

The CCT has a unique feature of measuring the light yield from internally generated electrons within a scintillator and has thus helped overcome problems related to the surface effects observed when using other techniques (Mengesha and Valentine, 2002). In order to measure the electron scintillation response, a scintillator is exposed to the collimated beam of monoenergetic gamma rays. The interaction process of Compton scattering takes place between the incident gamma ray photon and an electron in the absorbing material. It is the most predominant interaction mechanism for gamma ray of radioisotope sources. The relation of the energy transfer and the scattering angle for any given interaction can be expressed from simultaneous equations for the conservation of energy and momentum (Glenn, 2000). The Compton scattering requires that the light is viewed as a particle and not just a wave because it is the collision of photon with electron and the exchange of energy, which accounts for the shift in energy. The energy imparted to the recoil electron is given by Compton (Trousfanidis, 1983) according to the equation:

$$E_{\gamma'} = \frac{E_\gamma}{1 + \frac{(1 - \cos\theta)E_\gamma}{mc^2}} \tag{1}$$

where $E_{\gamma'}$ is the scattered gamma ray energy, E_γ is the incident gamma ray energy, θ is the scattering angle, and mc^2 is the rest-mass-energy of electron (511 keV). From Eq. (1), we can determine the electron energy (E_e) by conservation of energy:

$$E = E_\gamma - E_{\gamma'} \tag{2}$$

This equation demonstrates the response of scintillator to Compton electrons as recorded in coincidence with the Compton scattered photon at a given angle defined by the second collimator (see Fig. 1). In CCT technique, the Compton electrons in the primary scintillation detector generate a monoenergetic internal electron and it can be used to characterize the non-proportionality of scintillator light yield as a function of electron energy by varying the angle of the secondary detector (Limkitjaroenporrn et al., 2010).

2.2. Full width at half maximum (FWHM)

The full width at half maximum of electron energy spectra is related to the standard deviation (σ) and given by (Trousfanidis, 1983)

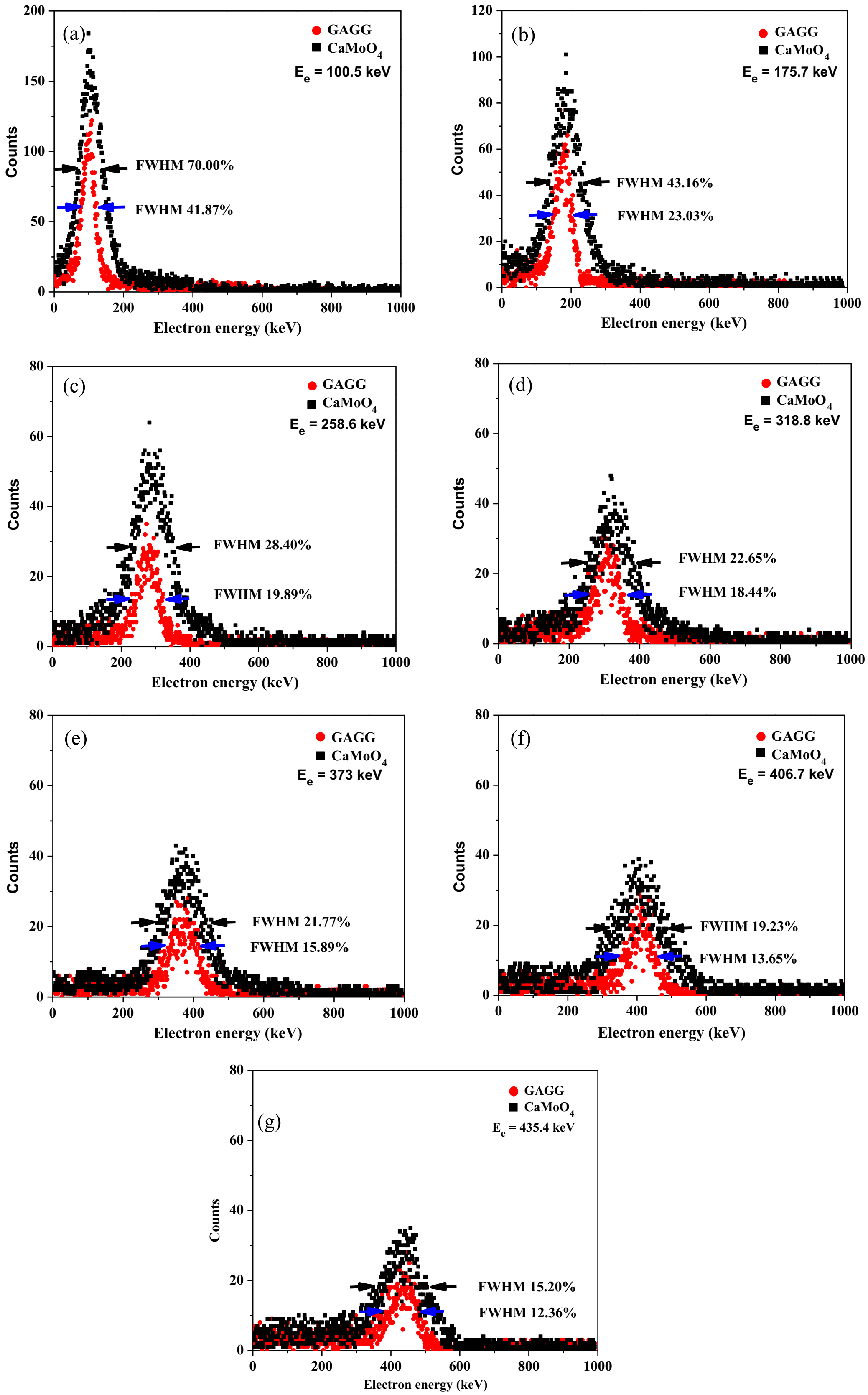

Fig. 2. (a)–(g) Electron energy spectra of $CaMoO_4$ and GAGG:Ce scintillation crystals in the energy range of 100.5–435.4 keV.

$$FWHM = 2\sigma\sqrt{2\ln 2} \tag{3}$$

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

$$R = \frac{FWHM}{E_0} \times 100\% \tag{4}$$

where E_0 is the electron energy peak centroid.

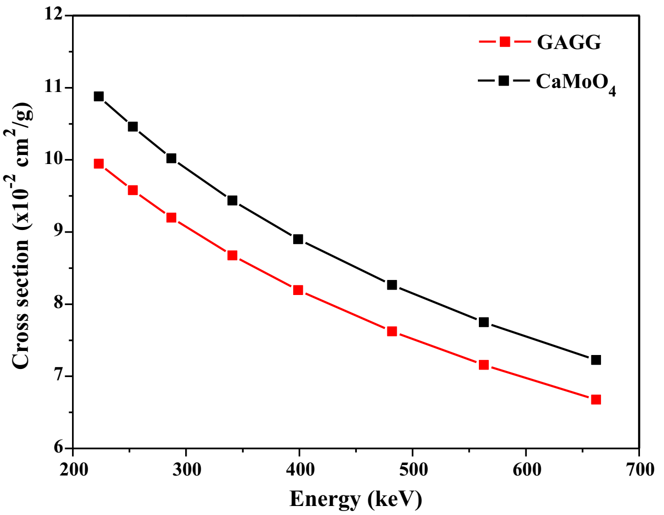

Fig. 3. Compton scattering interaction cross sections of CaMoO$_4$ and GAGG:Ce crystals in the energy region from 223.02 to 662 keV. (Calculated by WinXcom program).

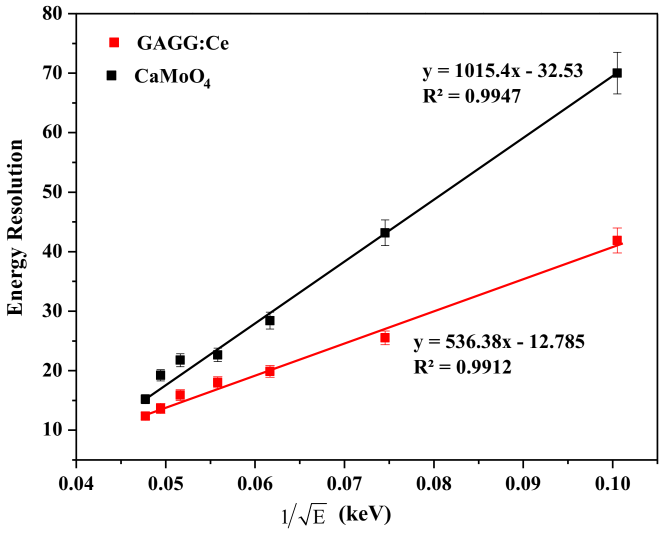

Fig. 4. Energy resolution of electron response of CaMoO$_4$ and GAGG:Ce crystals measured with the CCT in the energy range 100.5–435.4 keV.

Table 1
Light yield and non-proportionality of CaMoO$_4$ and GAGG:Ce crystals.

Electron energy (keV)	Light yield (electron/MeV)		Non-proportionality of electron response (% of 435.4 keV)	
	CaMoO$_4$	GAGG:Ce	CaMoO$_4$	GAGG:Ce
100.5	20146.8 ± 1800	152405.4 ± 13700	94.6 ± 2.84	94.9 ± 2.85
175.7	20817.9 ± 2080	156308.8 ± 14000	97.8 ± 2.45	97.3 ± 2.40
258.6	21064.5 ± 2100	158676.1 ± 14280	98.9 ± 1.97	98.8 ± 1.98
318.8	21163.7 ± 2110	159624.1 ± 14360	99.4 ± 1.98	99.4 ± 1.99
373.0	21226.1 ± 2120	160250.4 ± 14400	99.7 ± 1.50	99.8 ± 2.00
406.7	21274.7 ± 1910	160477.2 ± 14440	99.9 ± 1.90	99.9 ± 1.89
435.4	21292.9 ± 2130	160659.2 ± 14450	100.0 ± 1.50	100.0 ± 1.50

2.3. *Light yield and non-proportionality*

The light yield of crystal is defined by the number of electrons (N_e) divided by the average quantum efficiency (QE) of PMT (Bertolaccini et al., 1968):

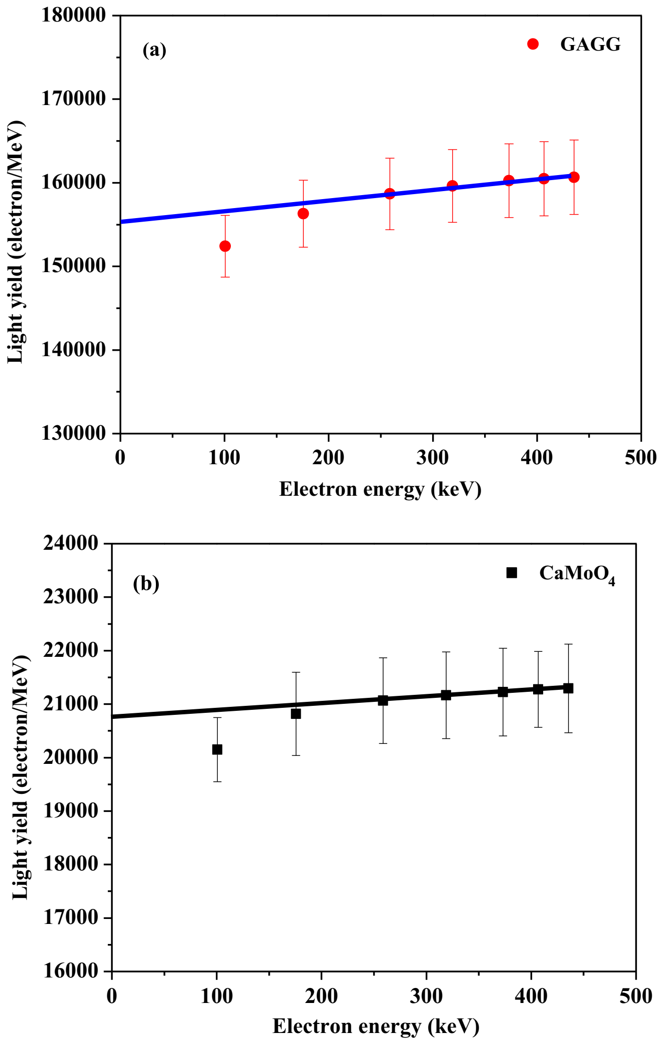

Fig. 5. Light yield of: (a) CaMoO$_4$ and (b) GAGG: Ce crystals as a function of electron energy. The solid line demonstrates the fit curve to the energy point above 260 keV.

$$\text{Light yield} = N_e(\text{MeV})/QE \tag{5}$$

where QE is the average quantum efficiency of the PMT at the peak wavelength of the emission spectrum for each scintillator.

The non-proportionality of crystal in percent (%) is the normalized light yield at an electron energy of 435.4 keV, that is the light yield for each electron energy divided by the light yield at an electron energy of 435.4 keV.

3. Experimental setup

Fig. 1(a) shows the schematic of CCT technique and the nuclear instrumentation setup. The ^{137}Cs obtained from the Office of Atom for Peace (OAP), Thailand, with an activity of 555 MBq was used as a monoenergetic gamma ray source with an energy of 662 keV. The CCT system consists of two detectors. Each scintillation crystal was used as a primary detector. A NaI(Tl) with a size of 2 × 2 in.^{2} was used as a secondary detector in order to measure the spectrum of scattered gamma rays.

Fig. 1(b) shows the nuclear instrument module (NIM) setup for the measurement of coincidence electron energy spectra. The scintillation crystal to be measured was mounted on the R1306 PMT, and then exposed to the gamma ray. The primary signal was amplified by an amplifier and converted to digital signal by an

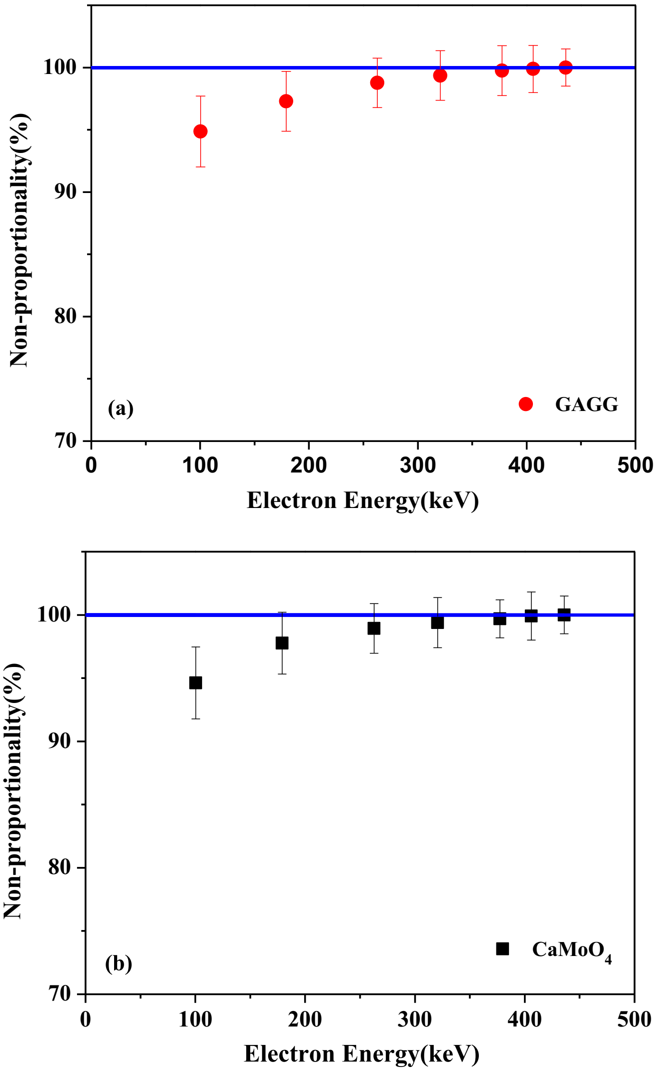

Fig. 6. Non-proportionality of CaMoO$_4$ and GAGG: Ce crystals in the electron energy range of 100.5–435.4 keV as normalized at 435.4 keV.

analog to digital converter (ADC). The signal from the secondary detector was amplified by an amplifier and delayed by a timing single channel analyzer (TSCA). The TSCA output was used to open the gate and analog signal was converted to digital signal by an analog to digital converter (ADC). When the signal from primary detector is coincident with the signal from secondary detector, the gate is opened and the coincidence signal from electron energy was obtained.

In this work, the CaMoO$_4$ and GAGG:Ce scintillation crystals were obtained from Kyungpook National University, Korea. Each crystal was mounted on the R1306 PMT using silicone grease and covered with several layers of teflon tape. The R1306 PMT with the crystal was then wrapped with the black tape and housed within a thin aluminum container. The gamma radiation from ^{137}Cs radioactive source with an energy (E_γ) of 662 keV was then exposed on the samples. The experiments were carried out using standard nuclear instrument modules (NIM). The coincidence electron energy spectra were recorded using a 32 Bit I/O board connected to a computer. A multichannel analyzer (MCA; Canberra) was used to record coincidence electron energy spectra at seven scattering angles of 30°–120° corresponding with electron energy in the range of 100.5–435.4 keV. The data from the coincidence electron energy spectra for both crystal samples were of Gaussian-shaped peaks. For each electron peak, the centroid and full width at half maximum (FWHM) of the full energy peak were obtained using Gaussian fitting software supplied by Canberra (Limkitjaroenporrn et al., 2010).

4. Results and discussion

Fig. 2 shows the electron energy spectra of CaMoO$_4$ and GAGG: Ce scintillation crystals, measured at seven different angles from 30°–120°, using CCT technique with the background subtraction to electron energy spectra. It was found that seven corresponding electron energies were in the range of 100.5–435.4 keV. It is seen that, for all electron energies, the electron energy peaks of CaMoO$_4$ crystal yielded higher number of counts than those of GAGG:Ce crystal. This is due to CaMoO$_4$ crystal has higher Compton scattering interaction cross section than that of GAGG:Ce crystal over the whole electron energy region, as can be seen in Fig. 3. The cross section data were obtained from the empirical calculation using WinXcom program.

From Fig. 1, the energy resolution of electron response for CaMoO$_4$ and GAGG:Ce crystals was determined using Eq. (4). Fig. 4 shows the energy resolution of electron response as a function of square root of electron energy. The relations are observed for both crystals indicated that the energy resolutions are inverse proportional to the square root of energy. The results are in good agreement with those of Phunpueok et al. (2012) and Prosper et al. (2012). In addition, we can deduce that the GAGG:Ce crystal has much better energy resolution of electron response.

The light yield as determined by Eq. (5) and the non-proportionality of CaMoO$_4$ and GAGG:Ce crystals in the energy range of 100.5–435.4 keV are shown in Table 1. It is seen that the light yield of GAGG:Ce crystal is much higher than that of CaMoO$_4$ crystal. The plots of the relationship between the light yield and electron energy are presented in Fig. 5. The graphs showed that the light yield slightly increased with increasing the electron energy. Fig. 6 shows the non-proportionality of CaMoO$_4$ and GAGG: Ce crystals in the electron energy range of 100.5–435.4 keV as normalized at 435.4 keV, It can be observed that both CaMoO$_4$ and GAGG:Ce crystals demonstrated good proportional property in the electron energy range of 260–435.4 keV. For the energy range below 260 keV, the electron response was slightly decreased at approximately 5% for both scintillation crystals over the measured energy range. Several studies suggest that the non-proportional response of the crystal is mainly responsible for the intrinsic resolution (Moszynski, 2003). In this work, the good proportional property was observed at higher electron energy, corresponding with lower resolution value or better resolution. This result is general trend for crystals.

5. Conclusion

The authors successfully developed the CCT technique and nuclear instrument module (NIM) setup for the measurements of coincidence electron energy spectra of calcium molybdate (CaMoO$_4$) and cerium doped gadolinium aluminium gallium garnet ($Gd_3Al_2Ga_3O_{12}$:Ce or GAGG:Ce) scintillation crystals. The ^{137}Cs obtained from the Office of Atom for Peace (OAP), Thailand, with an activity of 555 MBq was used as a monoenergetic gamma ray source with an energy of 662 keV. The CCT system consists of two detectors. The coincidence electron energy spectra were recorded using a 32 Bit I/O board connected to a computer. A multichannel analyzer (MCA; Canberra) was used to record coincidence electron energy spectra at seven scattering angles of 30°–120° corresponding with electron energy in the range of 100.5–435.4 keV. The results show that, for all electron energies, the electron energy peaks of $CaMoO_4$ crystal yielded higher number of counts than those of GAGG:Ce crystal. The electron energy resolution, the light yield and non-proportionality were also determined. It was found that the energy resolutions are inverse proportional to the square root of electron energy for both crystals. Furthermore, the results show that the light yield of GAGG:Ce crystal is much higher than that of $CaMoO_4$ crystal. It was also found that both $CaMoO_4$ and GAGG:Ce crystals demonstrated good proportional property in the electron energy range of 260–435.4 keV.

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