Performance of Ce-Doped (La, Gd)$_2$Si$_2$O$_7$ Scintillator with an avalanche photodiode

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Abstract

Scintillation properties of Ce-Doped (La, Gd)$_2$Si$_2$O$_7$ (Ce:La-GPS) crystal were measured with Si avalanche photodiode (APD, Hamamatsu S8664-55). Since Ce:La-GPS is a novel scintillator, its scintillation properties have been evaluated using the APD for the first time. This crystal grown by floating zone method had a good light output of 41,000 ± 1,000 photons/MeV and FWHM energy resolution at 662 keV was 4.4 ± 0.1% at 23.0±0.2°C. The photon non-proportional response (PNR) of Ce:La-GPS was approximately 65% at 32 keV, where light output at 662 keV was normalized to 100%. Moreover, the temperature dependence of the light outputs was determined to be approximately 0.15 %/°C from -10 to 30 °C.

Keywords: Scintillator; Ce-Doped (La, Gd)2Si2O7; Photon non-proportional response; Temperature dependence; APD

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1. Introduction

Scintillators are used in various fields such as medical imaging, astronomy [1, 2], and their energy resolution is one of the most important properties, especially when they are applied in radiation monitors, Compton cameras etc. [3]. Although some halide scintillators have excellent energy resolution (FWHM) of 3-5% at 662 keV, these crystals have are significantly hygroscopic [4-7]. Here, some Compton cameras or positron emission tomography (PET) cameras consist of pixel array of scintillation crystals. However, it becomes technically difficult to assemble such an array for the camera using the halide scintillator, because the strong hygroscopic nature degrades the crystal surface and light output [8, 9].

Many oxide scintillators such as Ce:Lu_2SiO_5 (Ce:LSO) [10], Ce:$(Lu_{1-x},Y_x)_2SiO_5$ (Ce:LYSO) [11], Ce:Gd_2SiO_5 (Ce:GSO) [12,13] and Ce:$Gd_3(Al,Ga)_5O_{12}$ (GAGG) [14] have been studied and applied in PET cameras. However, the Lu-containing crystals have intrinsic background due to ^{176}Lu decay, and the background noise degrades the angular resolution of Compton cameras. Moreover, such Lu-containing scintillators cannot be applied in radiation monitoring of low activity levels, which is a case of food monitoring around Fukushima. Thus, the Lu-free Ce:GSO or Ce: GAGG crystals can be successfully applied in the Compton camera [8,15] and food monitor. In particular, Ce:GAGG has a good FWHM energy resolution of 4.9% at 662 keV when coupled to a silicon avalanche photodiode (Si-APD) [14].

Recently, we have reported the scintillation properties of a $(Ce_{0.01},Gd_{0.90},La_{0.09})_2Si_2O_7$ (Ce:La-GPS: Cerium-doped lanthanum-gadolinium pyrosilicate) novel scintillator using a photo multiplier tube (PMT) [16]. It was found that the Ce:La-GPS has a high energy resolution (FWHM) of ~5% at 662 keV measured with the PMT, even if this sample was grown by floating zone (FZ) method and therefore its quality was a bit lowered with respect to crystals grown by Czochralski method or others [17-19]. Other optical and scintillation properties of this material are listed in Table 1. The energy resolution of this new scintillator can be improved to reach the same or better energy resolution with respect to Ce:GAGG.

The Ce:La-GPS has an emission wavelength of approximately 390 nm, and some Si-APDs are also available for detecting scintillation light from Ce:La-GPS instead of a PMT. Since the Si-APD is more compact and can be operated with lower voltage than the PMT, Ce:La-GPS coupled to the Si-APD is expected to be used in various applications. Moreover, this crystal can have a good energy resolution with the Si-APD due to the higher quantum efficiency (QE) of Si-APD than PMT. Thus, we report the scintillation properties of Ce:La-GPS measured by the Si-APD for the first time in this paper.

Table1 Luminescent properties of Ce:La-GPS	
Emission wavelength	390 nm
Decay time	46 ns (79%)
	346 ns (21%)
Light output	3,6000 ph/MeV

Although photon non-proportional response (PNR) of many scintillators was investigated and summarized in the previous works [20-25], in the pyrosilicate group ($Ln_2Si_2O_7$, Ln = Lu, Gd, etc.), only that of Ce:$Lu_2Si_2O_7$ (Ce:LPS) was reported. The PNR is found to be dependent on some factors such as crystal structure, dopant etc. Thus, PNR of the Ce:La-GPS scintillator is also evaluated, and compared with Ce:Gd_2SiO_5 (similar components) and Ce:$Lu_2Si_2O_7$ (the same crystal structure type) in this paper.

One of the applications of this crystal is radiation monitoring, and the temperature dependence of the light output is an important factor in order to determine the dose more precisely. Using some X-ray source, we can calibrate the APD and preamplifier gain as a function of temperature. Thus, the temperature dependence of the light output and the energy resolution for the Ce:La-GPS scintillator is also investigated.

2. Materials and Experimental Methods

2.1 APD calibration

In order to estimate the light output of the crystal, a Si-APD (Hamamatsu S8664-55)[26] was excited with X rays from several sources, ^{55}Fe (5.9 keV), ^{57}Co(6.4, 14.4 keV), ^{241}Am(13.9, 17.8 20.8 keV), without the crystal as shown in Fig 1 (a) for the first step. This Si-APD was operated at a high voltage of 290 V with a bias supply (Clear-pulse, E6665). The output signal from the Si-APD was preamplified (Clear-pulse, 581K) and shaped with a module (Clear-pulse, 4417) using a shaping time of 1 μs, and digitized with a multi-channel analyzer (Amptek, MCA8000A). Then the pulse height spectra were obtained. The Si-APD and the preamplifier were set in the thermostat chambers (Espec, SU-241), and the above measurement was performed at 23.0±0.2°C.

2.2 Pulse height spectra

The polished sample with a thickness of 1 mm and grown by FZ method was coupled to the Si-APD using optical grease (Oken, 6262A) as shown in Fig 1 (b). Here, Teflon™ tape was used as a reflector, and the crystal sample was excited with gamma rays from selected X/gamma-ray sources: ^{241}Am(59.5 keV), ^{57}Co(122 keV), ^{152}Eu(40.1,122, 245, 344, 1408 keV) ,^{133}Ba (31.0, 81.0, 274, 303, 356, 384 keV), ^{137}Cs (32.2, 662 keV), ^{22}Na (511, 1275 keV), ^{54}Mn(835 keV) and ^{60}Co (1173, 1333 keV). The measurement system and the temperature conditions were the same as described for the above Si-APD calibration system.

We compared the peak channels of pulse height spectra data for this sample irradiated with X-rays (5.9 keV) and gamma rays from ^{55}Fe, and ^{137}Cs sources, respectively, in order to evaluate its light output. We assumed that (i) the APD had a QE of 63% at 390 nm and (ii) the electron/hole pair creation for Si was 3.6 eV [27], and the light output (LO) was calculated using a following equation:

$$LO[\text{photons/MeV}] = C_{Cs}/C_{Fe} \times (1\ \text{MeV}/0.662\ \text{MeV}) \times \{(5.9\ \text{keV}) / (0.0036\ \text{keV})\} / 0.63, \quad (1)$$

where C_{Cs} and C_{Fe} are absorption peak channel at pulse height spectra of the sample irradiated from ^{55}Fe, and ^{137}Cs sources, respectively.

Using the above data and (1), PNR of the Ce:La-GPS scintillator and energy resolution were evaluated. Here, the non-proportionality at energy *E* was defined as follows:

$$PNR(E) = Npe_{APD}(E) / Npe_{APD}(E=662\ \text{keV}), \quad (2)$$

where $Npe_{APD}(E)$ denotes the number of photoelectrons per MeV of absorbed energy observed at energy *E* [20].

2.3 Temperature Dependence

In order to show the temperature dependence of the light output and the energy resolution for Ce:La-GPS, pulse height spectra were obtained at several temperatures from -20 to 30°C with a 5°C-interval using the thermostat chamber. Here, this temperature range is similar to the usual environmental temperature for the dose monitor in Japan or other area. Temperature dependence of the Si-APD and preamplifier gain was also measured using a ^{55}Fe source. After the gain correction for the Si-APD and preamplifier, the light outputs of the scintillator at several temperatures were evaluated.

3. Results and discussion

3.1 APD calibration

The pulse height spectra of the Si-APD under excitation with X rays from ^{55}Fe (5.9 keV), ^{57}Co (6.4, 14.4 keV) and ^{241}Am (13.9, 17.8, 20.8 keV) sources without the scintillator were obtained, and the absorption-peak-MCA channel position as a function of the X-ray energy is shown in Fig. 2 (a). From this figure, the APD and other readout system had good linearity within ±3% from a generated-photoelectron number of 1,600 to 6,000 (Fig. 2 (b)). This photoelectron number region was corresponded to a photon number of 2,500 to 9,400 assuming that the quantum efficiency of the Si-APD is 64% at 390 nm. Best-fit line for the data points in Fig. 2 (a) is described as follows

$$Ex = a \text{ (MCA channel) } + b, \quad (3)$$

where *a, b* are fitting parameters, and *Ex* denotes the X-ray Energy. Here,

$$Ex = Q\, w\, N_{phe} \quad (4)$$

where, *Q, w* and N_{phe} are QE, electron/hole pair creation and number of photoelectrons, respectively. Thus, we can obtain the number of photoelectrons.

3.2 Pulse height spectra

Pulse height spectrum of Ce:La-GPS excited by ^{137}Cs was obtained as shown in Fig. 3, and the escape peak, originated from Gd Kα-line (42.7 keV) mainly, was also observed due to the good energy resolution. The light output of this crystal was determined to be 41,000 ± 1,000 photons/MeV at 23.0±0.2°C using ;

The pulse-height spectra for the other sources were also obtained, and the PNR of this crystal is shown in Fig. 4. This crystal had a PNR of 0.9 – 1.05 above 300 keV. The PNR of Ce:La-GPS was approximately 64% at 32 keV, and those of Ce:GSO, Ce:LPS and Ce:LSO are listed in Table 2. Here, the PNRs at 32 keV for the Ce:GSO, Ce:LPS and Ce:LSO were estimated from the figure in ref [20].

Table2 PNR of several scintillators

	PNR@10 keV [%]	PNR@32 keV [%]	Ref
Ce:La-GPS	n/a	65	This work
Ce:GSO	68.8	~88	[20-22]
Ce:LPS	~42	~65	[20, 22-24]
Ce:LSO	56.7	~77	[20-22,24, 25]

Ce:La-GPS had almost the same PNR as Ce:LPS which is the same crystal structure type. Our sample consists of 9 mol%-La, and also the PNR may depend on the concentration of La.

The FWHM energy resolution of Ce:La-GPS at 662 keV was 4.4 ± 0.1%, and Fig. 5 shows the FWHM energy resolutions from 30 to 1333 keV. Best-fit line for the data points is represented by α=4.6±0.1, β=0.52±0.01 using the following equation:

$$\text{resolution }(\%) = \alpha \times \left(\frac{E}{662\text{ keV}} \right)^{-\beta} . \qquad (5)$$

This crystal had excellent energy resolution. β was close to 0.5, and this suggests that the light output is large and the photoelectron statistics is the main contribution to the energy resolution.

Moreover, we also measured the same Ce:La-GPS sample using a PMT (Hamamatsu, R7600-200), as the APD measurement, and the FWHM energy resolution was 4.7 ± 0.1 % at 662 keV. The energy resolutions measured with the PMT and APD were almost the same value.

3.3 Temperature Dependence

In order to measure the temperature dependence of the APD and preamplifier gain, pulse height spectra of the APD, under excitation with5.9 keV X-ray from a ^{55}Fe source, were obtained without the scintillator.

Figure 6 shows the APD signal excited by the ^{55}Fe source as a function of temperature, where the light output and the peak ADC channel at -20°C were normalized to 1. Typical Si-APD gain data from the data sheet [26] is also shown in Fig. 6.

Figure 7 shows the temperature dependence of the light output of the Ce:La-GPS crystal after the correction for the APD and preamplifier gain. Moreover, the temperature dependence of the FWHM energy resolution at 662 keV of Ce:La-GPS is shown in Fig. 8. Below -10°C, the light output decreased, and the energy resolution was worse than the values above -10°C. Moreover, the temperature dependence of the light outputs (temperature coefficient of light output) was determined to be approximately 0.15 %/°C from -10 to 30°C using a following equation:

$$(\text{Temperature coefficient}) = \{(LO_{30} - LO_{-10}) / LO_{10}\} / \{30 - (-10)\}, \qquad (6)$$

where LO_T is light output at T [°C]. The temperature coefficients of other scintillators are listed in Table 3, and Ce:La-GPS had stable light output around the room temperature.

Table3 Temperature Coefficient of Light Output

	Temperature Coefficient [%/°C]	Temperature Range [°C]	Reference
Ce:La-GPS	~0.15	-10 to 30	This work
$Bi_4Ge_3O_{12}$(BGO)	-0.9	5 to 35	[28]
Ce:LSO	-0.2	5 to 35	[28]
Ce:GSO	-0.8	-20 to 20	from Table in [29]

4. Conclusions

The (La, Gd)$_2$Si$_2$O$_7$ (Ce:La-GPS) crystal grown by floating zone had a good light output of 41,000 ± 1,000 photons/MeV and FWHM energy resolution at 662 keV was 4.4 ± 0.1% at 23.0 ± 0.2°C measured with a Si-avalanche photodiode. The photon non-proportional response of Ce:La-GPS was approximately 65% at 32 keV. The temperature coefficient of light output was approximately 0.15 %/°C from -10 to 30 °C. Although this crystal had similar photon non-proportional response compared with other oxide scintillators such as Ce:$Lu_2Si_2O_7$, Ce:Gd_2SiO_5, it had an excellent light output and energy resolution.

Acknowledgments

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References

[1] Carel W E van Eijk, Phys. Med. Biol. 47 (2002) R85.

[2] C. Rozsa, R. Dayton, P. Raby, M. Kusner, R. Schreiner, IEEE Trans. Nucl. Sci., 37 (1990) 966.

[3] T. Tanimori et al., “MeV γ-ray imaging detector with micro-TPC,” New Astron. Rev., 48 (2004) 263.

[4] E. V. D. vanLoef, P. Dorenbos, C. W. E. van Eijk, K. W. Krämer,and H. U. Güdel, Appl. Phys. Lett., 79,

(2002) 1573.
[5] K. S. Shah, J. Glodo, M. Klugerman, W. Higgins, T. Gupta, P. Wong, W. W. Moses, S. E. Derenzo, M. J. Weber, and P. Dorenbos, IEEE Trans.Nucl. Sci., 51, (2004). 2302.
[6] J. T. M. d. Haas and P. Dorenbos. Advances in Yield Calibration of Scintillators. IEEE Trans. Nucl Sci., 55 (2008)1086.
[7] E. V. D. van Loef, P. Dorenbos, K. Kramer and H. U. Gudel, IEEE Trans. Nucl. Sci., 48 (2001) 341.
[8] H. Nishimura, K. Hattori, S. Kabuki, H. Kubo, K. Miuchi, T. Nagayoshi, Y. Okada, R. Orito, H. Sekiya, A. Takada, A. Takeda, T. Tanimori, K. Ueno, Nucl. Instr. and Meth. A573 (2007) 115.
[9] S. Kurosawa, H. Kubo, K. Hattori, C. Ida, S. Iwaki, S. Kabuki, K. Miuchi, H. Nishimura, Y. Okada, J. D. Parker, A. Takada, M. Takahashi, T. Tanimori, K. Ueno, and Y. Yanagida IEEE Trans. Nucl. Sci., 56 (2009) 3779.
[10]C. L. Melcher and J. S. Schweitzer. IEEE Trans. Nucl. Sci., 39 (1992) 502.
[11]L. Pidol, A. Kahn-Harari, B. Viana, E. Virey, B. Ferrand, P. Dorenbos, J. T. M. de Haas and C. W. E. van Eijk. IEEE Trans. Nucl. Sci., 51 (2004)1084.
[12] K. Takagi and T. Fukazawa, Appl. Phys. Lett., 42 (1983) 43.
[13] H. Ishibashi, K. Shimizu, K. Susa, and S. Kubota, IEEE Trans. Nucl. Sci., 36 (1989) 170.
[14] K. Kamada, T. Yanagida, T. Endo, K. Tsutumi, Y. Usuki, M. Nikl, Y. Fujimoto, A. Fukabori, A. Yoshikawa, Journal of Crystal Growth 352, (2012) 88.
[15] A. Takada, H. Kubo, H. Nishimura, K. Ueno, K. Hattori, S. Kabuki, S. Kurosawa, K. Miuchi, E. Mizuta, T. Nagayoshi, N. Nonaka, Y. Okada, R. Orito, H. Sekiya, A. Takeda, T. Tanimori ,The Astrophysical Journal (2011) 733:13
[16] A. Suzuki, S. Kurosawa, T. Shishido, J. Pejchal, Y. Yokota, Y. Futami and A. Yoshikawa, Appl. Phys. Express 5 (2012) 102601.
[17] J. Czochralski, Z. phys. Chem. 92, 219 (1918).
[18]C.D. Brandle, Journal of Crystal Growth 264, 593 (2004).
[19]L.A. Boatner et al., Journal of Crystal Growth 379, 63 (2013).
[20]I. V. Khodyuk and P. Dorenbos, IEEE Trans. Nucl. Sci., 59 (2012) 3320.
[21]M. Balcerzyk, M. Moszynski, M. Kapusta, D. Wolski, J. Pawelke, C. L. Melcher, IEEE Trans. Nucl. Sci., 47 (2000) 1319.
[22] Ivan V. Khodyuk, Johan T. M. de Haas, and Pieter Dorenbos, IEEE Trans.Nucl. Sci., 57 (2010) 1175.
[23] D. Pauwelsl, N. Le Massod, B. Vianna', A. Kahn-Hararil, E.V.D. van Loef, P. Dorenbos, and C.W.E. van Eijk, IEEE Nuclear Science Symposium, 1999. Conference Record. 1 (1999) 102.
[24]P. A. Cutler, C. L. Melcher, M. A. Spurrier, P. Szupryczynski, and L. A. A. Eriksson IEEE Trans.Nucl. Sci., 56 (2009) 915.
[25] P.Dorenbos, J. T.M. de Haas, and C. W. E. van Eijk, IEEE Trans. Nucl. Sci., 42 (1995) 2190.
[26] “Si APD”, Hamamatsu photonics, http://www.hamamatsu.com/resources/pdf/ssd/si_apd_kapd0001e04.pdf

[27]J.R. Fiebiger and R.S. Muller, J. Appl. Phys. 43, 3202 (1972).
[28]R. Mao, L. Zhang, R.-Y. Zhu, IEEE Trans. Nucl. Sci., 55 (2008) 2425.
[29] N. Tsuchida, M. Ikeda, T. Kamae, M. Kokubun, Nucl. Instr. and Meth. A, 385 (1997) 290.

Figure captions

Figure1. Schematic view of the setup for the APD gain calibration (a), and measurement of pulse height spectrum for the Ce:La-GPS (b).

Figure2. APD calibration: (a) X-ray energy as a function of MCA channel and the solid line denotes the best fit line described as (2). (b) its residual error between the above data and fitting line.

Figure3. Pulse height spectrum of the Ce:La-GPS crystal irradiated with gamma rays from a ^{137}Cs source.

Figure4. Photon non-proportional response as a function of X/gamma-ray energy.

Figure5. Energy resolution (FWHM) at 662 keV as a function of X/gamma-ray energy. The solid line denotes the best fit line described as (4).

Figure6. Gain of readout system including the Si-APD and preamplifier as a function of temperature (closed circles) and the APD gain in [26] (open squares). In these sets, data points at -20 °C are normalized to 1

Figure7. Light output of Ce:La-GPS scintillator at 1 MeV as a function of temperature.

Figure8. Energy resolution (FWHM) at 662 keV as a function of temperature.

Fig. 1

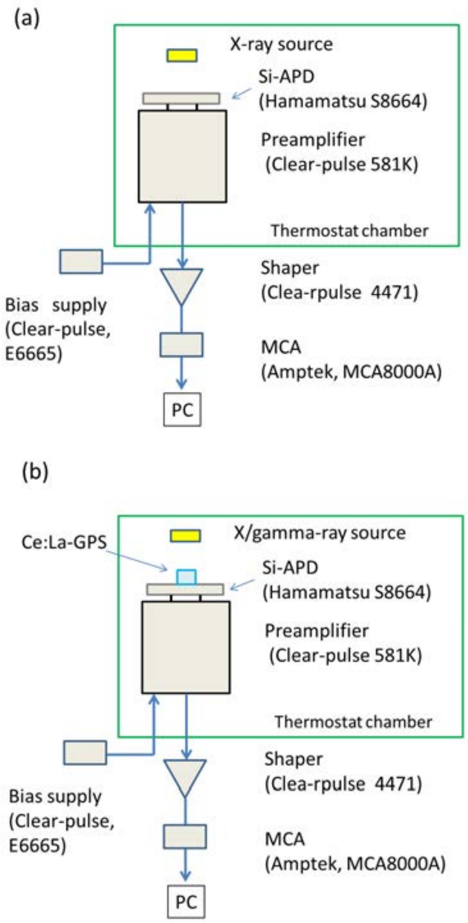

(a)
X-ray source
Si-APD
(Hamamatsu S8664)
Preamplifier
(Clear-pulse 581K)
Thermostat chamber
Shaper
(Clea-rpulse 4471)
Bias supply
(Clear-pulse,
E6665)
MCA
(Amptek, MCA8000A)
PC
(b)
Ce:La-GPS
X/gamma-ray source
Si-APD
(Hamamatsu S8664)
Preamplifier
(Clear-pulse 581K)
Thermostat chamber
Shaper
(Clea-rpulse 4471)
Bias supply
(Clear-pulse,
E6665)
MCA
(Amptek, MCA8000A)
PC

Fig. 2

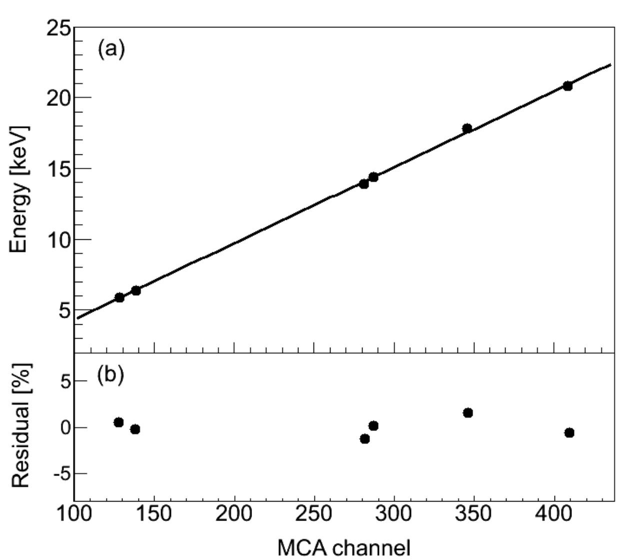

(a)
Energy [keV]
25
20
15
10
5
(b)
Residual [%]
5
0
-5
100
150
200
250
300
350
400
MCA channel

Fig. 3

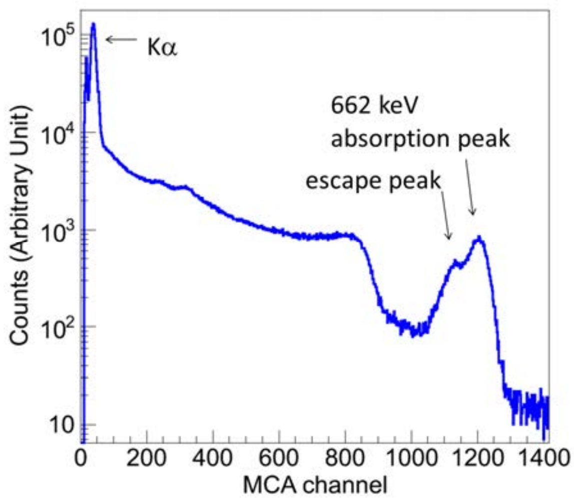


Fig. 4

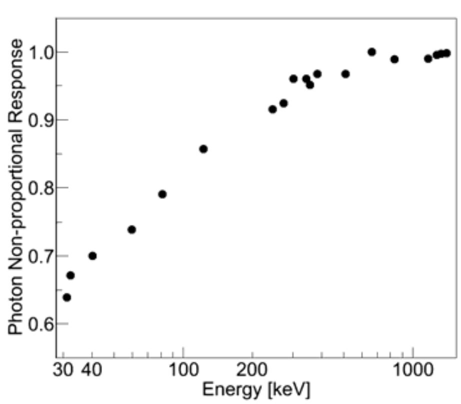
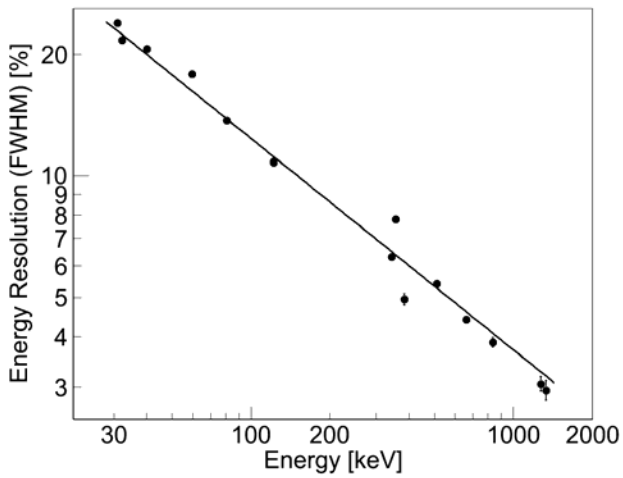
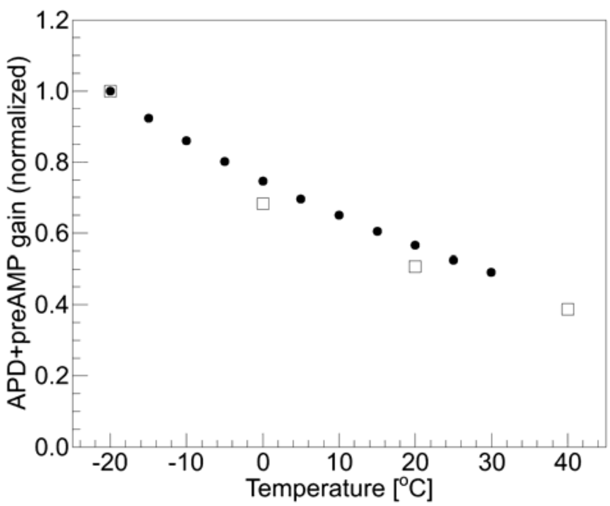


Fig. 5



Energy Resolution (FWHM) [%]
20
10
9
8
7
6
5
4
3
30
100
200
1000
2000
Energy [keV]

Fig. 6



APD+preAMP gain (normalized)
1.2
1.0
0.8
0.6
0.4
0.2
0.0
-20
-10
0
10
20
30
40
Temperature [°C]

Fig. 7

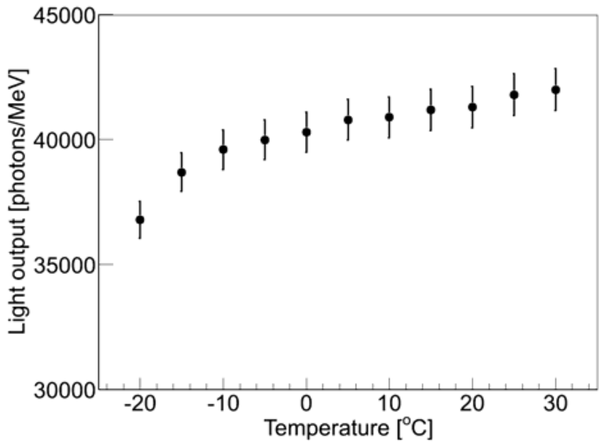
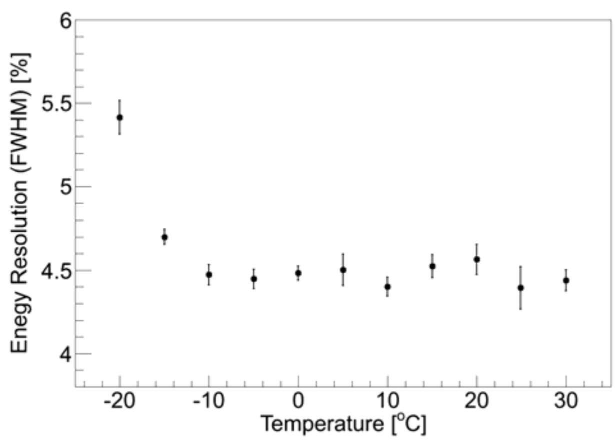

45000
40000
35000
30000
Light output [photons/MeV]
-20
-10
0
10
20
30
Temperature [°C]

Fig. 8


6
5.5
5
4.5
4
Enegy Resolution (FWHM) [%]
-20
-10
0
10
20
30
Temperature [°C]