Characterisation of Cerium-Doped Lanthanum Bromide scintillation detector

Etim Iniobong Prosper[1], Obu Joseph Abebe[2], Ushie James Ogri[3]
[123]Department of Physics, University of Calabar. P.M.B. 1115, Calabar, Cross River State Nigeria.

E-mail: Ini2etim@yahoo.com

(Received 12 December 2011, accepted 6 March 2012)

Abstract

Cerium-doped lanthanum Bromide $LaBr_3$(Ce) crystals is one of the new scintillating detectors that has been developed in recent years which has proven to be superior to other scintillating materials in terms of resolution and efficiency. In this study we have carried out the characterisation of a 25mm X 25mm Brillance^{TM} 380 $LaBr_3$(Ce) detector through the laboratory measurement of its energy resolution, intrinsic photo-peak, total intrinsic, total absolute efficiency and timing resolution. In addition, the energy dependence of the resolution has been studied using a variety of gamma ray sources with variable energy in the range (122-1408keV). The study showed that $LaBr_3$(Ce) detector has an excellent energy resolution of 2.6% full-width-at-half maximum (FWHM) at 662keV photons using a Caesium-137 (^{137}Cs) source at room temperature as compared to NaI(Tl) detector. Also, it was determined that the detector has a full-energy peak efficiency ranging from 90.1% to 4.3% in the energy range of 122-1408keV and for a source-to-detector distance of 150mm.

Keywords: Brillance^{TM} 380 $LaBr_3$(Ce), energy resolution, FWHM, intrinsic photo-peak, total intrinsic and total absolute efficiency.

Resumen

El cerio-lantano dopado Bromuro $LaBr_3$(Ce) cristales es uno de los nuevos detectores de brillantes que se ha desarrollado en los últimos años el cual ha demostrado ser superior a otros materiales de brillantes en términos de resolución y eficiencia. En este estudio se ha llevado a cabo la caracterización de un 25mm x 25mm Brillance^{TM} 380 LaBr3(Ce) a través del detector de medición en el laboratorio de esta resolución de energía, de fotos-máxima actividad intrínseca, total intrínseco, total absoluto de la eficiencia y resolución de tiempo. Además, la dependencia de energía de la resolución se ha estudiado usando una variedad de fuentes de rayos gamma con energía variable en el rango (122-1408keV). El estudio mostró que el detector $LaBr_3$(Ce) tiene una excelente resolución energética del 2.6% total-ancho-de-la mitad máxima (FWHM) en 662keV fotones usando una de Cesio-137 (^{137}Cs) fuente a temperatura ambiente en comparación con el detector NaI(Tl). Además, se determinó que el detector tiene una eficiencia de energía pico-completa que van desde 90.1% a 4.3% en el rango de energía de 122-1408keV y una fuente-para-la distancia del detector de 150mm.

Palabras clave: Brillance^{TM} 380 $LaBr_3$(Ce), resolución de energía, FWHM, fotos-máxima actividad intrínseca, intrínseca total y eficiencia absoluta total.

PACS: 29.25.RM, 29.30.KV, 29.30.MC **ISSN 1870-9095**

I. INTRODUCTION

One of the best tools in obtaining experimental information on nuclear structure is the spectroscopy of gamma rays emitted in the de-excitation of a nucleus or in nuclear reactions [6].The full understanding of these nuclear processes requires observations with instruments having excellent energy resolution together with reasonable efficiency. The use of spectrometers give information both on energy and intensity of the radiation emitted from the source. These spectrometers often referred to as detectors which are categories into gas detectors, scintillation detectors and semi-conductor detectors.

Scintillation crystals have been one of the earliest and popular methods used for the detection of gamma rays [2]. A scintillator is generally a material that emits low-energy (in the visible range) photons when they are struck by high-energy charge particle. Scintillator detectors are divided into organic and inorganic and the types of materials used depend on the application. One of the most commonly used scintillator detector is the inorganic one, where alkali halide salts such as sodium iodide (NaI) and caesium iodide (CsI) are used. An activator in the form of impurity (*e.g*. thallium) is introduced into the inorganic material in order to improve their performance.

The growing interest in scintillator detectors has led to the emergence of the cerium doped halide scintillator,

$LaBr_3$(Ce).This scintillator has demonstrated a high light output (~63,000photons/MeV) with a fast decay time of 16ns [7]. These attractive properties have distinguished it among other scintillators for selected applications in which the demand for energy resolution is a dominant factor over density and attenuation length. In this work, we have carried out a characterisation of the gamma-ray detection properties of $LaBr_3$(Ce), where the optimum operating voltage of the photomultiplier tube (PMT), shaping time of the amplifier, energy resolution and the detection efficiency of the detector was determined.

II. THEORY OF GAMMA-RAY INTERACTIONS IN SCINTILLATORS

There are three major interaction mechanisms that play vital roles in radiation measurements. These include photoelectric absorption, Compton scattering and pair production [2]. In each of these processes there is a partial or complete transfer of gamma-ray photon energy to the electron, resulting in a sudden change in the history of the gamma-ray photon, *i.e.*, the photon either disappears entirely or is scattered through a significant angle.

A. Photoelectric Absorption

This is a process in which a photon undergoes an interaction with an absorber atom and is completely absorbed, while an energetic photoelectron is concurrently ejected from one of the bound shells of the atom [2]. The interaction invariably occurs with the most tightly bound or K-electron of the atom, and does not occur with free electrons. The energy of the ejected photoelectron, E_e is given by

$$E_{e^-} = hv - E_b, \tag{1}$$

where E_b is the binding energy of the photoelectron in its original shell, and hv is the energy of the gamma-ray photon. Photoelectric absorption leads to the creation of an ionized absorber atom as a result of the vacancy left in one of the bound shells. A free electron from the medium is quickly captured to fill the vacancy and there is a rearrangement of electrons from other shells of the atom. This could result in the emission of the characteristic X-ray while in some cases the emission of an Auger electron may occur. Photoelectric process occurs predominantly for gamma rays (or X-rays) of relatively low energy [5].

B. Compton scattering

Compton scattering is an interaction process where the photon transfers a portion of its energy to the electron (assumed to be at rest) leading to the deflection of the photon through an angle θ with respect to its original direction and a recoil of the target electron. The energy change during the process is given by [2]:

$$E_{e^-} = hv - hv' = hv\left(\frac{\left(\frac{hv}{m_oc^2}\right)(1-cos\theta)}{1+\left(\frac{hv}{m_oc^2}\right)(1-cos\theta)}\right), \tag{2}$$

where m_oc^2 is the rest mass energy of the electron, E_{e^-} is the recoil energy of the electron, ν and ν' are the frequencies of the incident and deflected photon respectively.

C. Pair Production

Pair production occurs in the intense electric field created by protons in the nuclei of the absorbing material. This process is energetically possible only if the gamma-ray energy exceeds twice the rest mass energy of an electron, and so, the process is only limited to high-energy gamma rays since its probability of interaction increases at high energy. During Pair production the gamma ray photon disappears and an electron-positron pair is produced. The excess energy of the photon above the required threshold appears in the form of kinetic energy shared by the electron-positron pair, *i.e.*

$$E_{e^-} + E_{e^+} = hv - 2m_oc^2. \tag{3}$$

After production, the positron slows down and annihilate in the absorbing medium producing two annihilation photons as secondary products of interaction. It is this annihilation radiation that plays an important part in the response of gamma-ray detectors.

III. THEORY OF ENERGY CHARACTERISATION OF SCINTILLATION DETECTORS

A. Energy Resolution

Energy resolution of a detector is a measure of its ability to resolve small differences in the energy of incident γ-rays. It is defined as [3]:

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

where FWHM is the Full width of the full-energy peak at half maximum height expressed as the number of channels, and H_o is the channel number corresponding to the peak centroid.

In scintillation detectors the important factor affecting resolution is the statistical fluctuation in the number of photoelectrons produced in the PMT, *i.e.* the variation in the amplitude of successive events [4]. Another additional factor is the non-uniformity in light output. The amplitude of the output pulse is proportional to the number of photoelectrons created.

$$R = \frac{FWHM}{H_O} = \frac{K\sqrt{E}}{E} = \frac{K}{\sqrt{E}}, \tag{5}$$

where *E* is the Photon energy and *K* is a constant.

B. Detection efficiency

It describes the proportion of radiation events recorded by a detector, and includes both the geometric and radiation interaction effects. The efficiency of a detector is a measure of how many pulses occur for a given number of gamma rays. There are three categories of detection efficiency, namely, absolute total efficiency, intrinsic total efficiency and intrinsic full-energy peak efficiency.

IV. EXPERIMENTAL SET-UP

The characterisation of LaBr(Ce) was carried out using a 25mm X 25mm Brilliance^{TM} 380 Crystal (Type: B380) LaBr3(Ce) detector coupled to a PM:XP2060 B02 PMT. These were connected to a Canberra HV supply and a Canberra amplifier 2022 which was then linked to an Ortec Easy MCA and a Dell PC. The experimental set-up is as depicted in Fig. 1. The amplifier settings were as follow: Fine gain: 0.5x, coarse gain: 30x, input polarity: Positive.

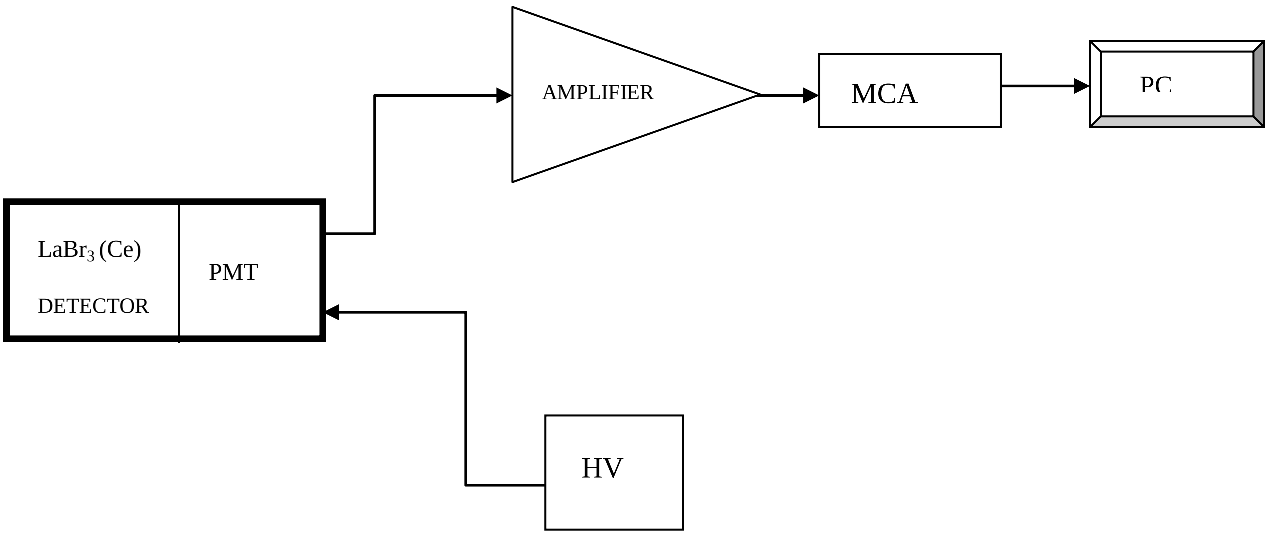


FIGURE 1. Experimental set-up for characterisation LaBr3(Ce) detector.

V. EXPERIMENTAL PROCEDURE

A. Energy Calibration

Applying an operating voltage of 750V and an optimum shaping time of 1μs, the $LaBr_3$(Ce) detector was calibrated using ^{152}Eu, ^{22}Na, ^{137}Cs, and ^{60}Co sources by collecting energy spectrum and determining the central channel number of the full energy peak by eye via the peak report on the Multi-Channel Analyzer (MCA). A graph of energy against channel number was plotted in order to observe the linearity of the energy response of the detector.

B. Energy resolution

Energy spectrum was taken from ^{152}Eu, ^{22}Na, ^{60}Co and ^{137}Cs for 300 seconds and the FWHM in keV for each photo-peak was recorded. The energy resolution was calculated using Eq. (4).

C. Efficiency measurement

The ^{137}Cs source was used to collect a spectrum in order to determine the various efficiency. A region of interest was set over the whole spectrum and the total count rate in this region was recorded, with the background count rate taken into consideration. The net count rate was collected for the full energy photo-peak. These procedures were repeated for ^{57}Co, ^{22}Na, ^{60}Co and ^{152}Eu sources by varying source detector distance. A graph of intrinsic photo-peak efficiency against energy was then plotted.

VI. RESULTS AND DISCUSSION

Figure 2 shows the energy-channel relation for the $LaBr_3$(Ce) detector. The entire response of the detector covers the energy range of 122KeV-1408KeV and was found to be linear. A linear function of $E = 1.5135C - 32.541$ was obtained from the calibration. Using this relationship channels can be easily converted to energy. As can be seen from the plot, $LaBr_3$(Ce) detector exhibits an outstanding linear response.

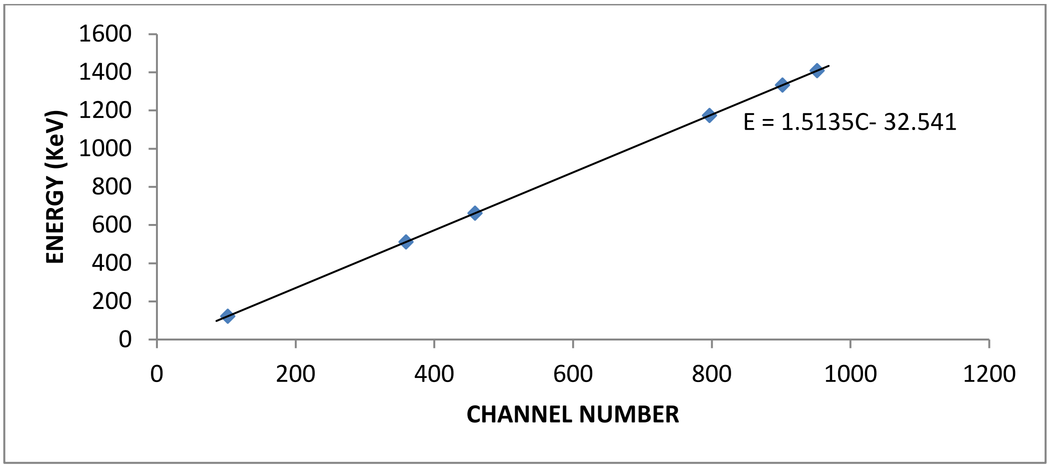


FIGURE 2. Graph of energy against channel number for 25mm X 25mm Brillance™ $LaBr_3(Ce)$ detector.

The response function of the $LaBr_3(Ce)$ shows that the energy resolution improves with increase in gamma-ray energy. For the $LaBr_3(Ce)$ detector an energy resolution of 2.6% at the 662 keV photo-peak at room temperature was recorded. This is graphically depicted in Fig. 3. This value of energy resolution approaches the value for the energy resolution of some semiconductor detectors such as CdTe and CdZnTe at room temperature [5], and this has never been achieved with any established inorganic scintillators.

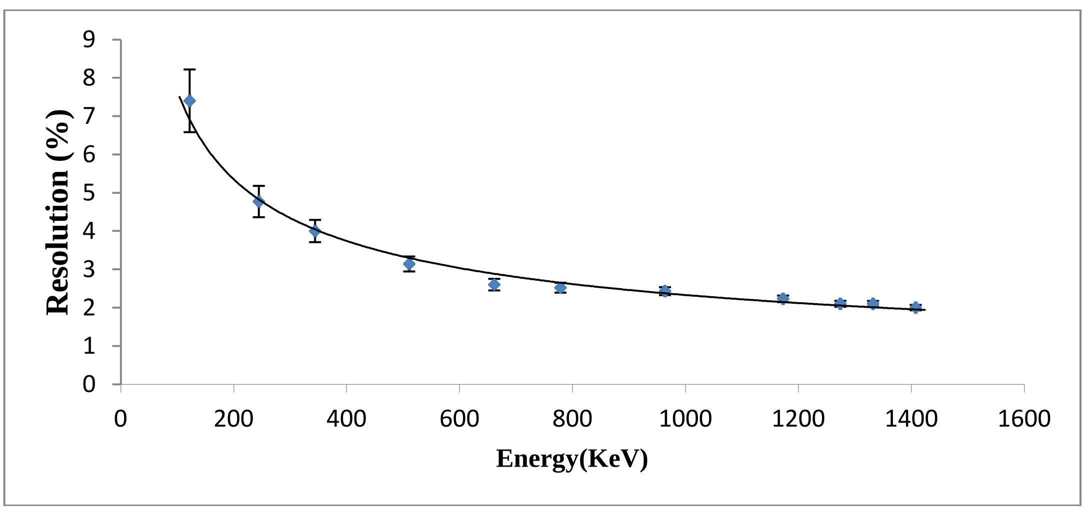


FIGURE 3. Graph of energy resolution versus energy for 122-1408KeV energy window.

The energy resolution of 2.6% at 662keV makes the resolution of this detector two times better than NaI(Tl) ~6% (Knoll, 2000).This detector is expected to distinguish two gamma peaks that lie close to each other. However, the resolution of $LaBr_3(Ce)$ detector is not as good as that of a semiconductor detector crystal such as Hyper Pure Germanium (HpGe) whose typical value lie in the range < 0.1% [2]. This remarkable energy resolution obtained from the $LaBr_3(Ce)$ detector is due to the high light output and good homogeneity of the crystals.

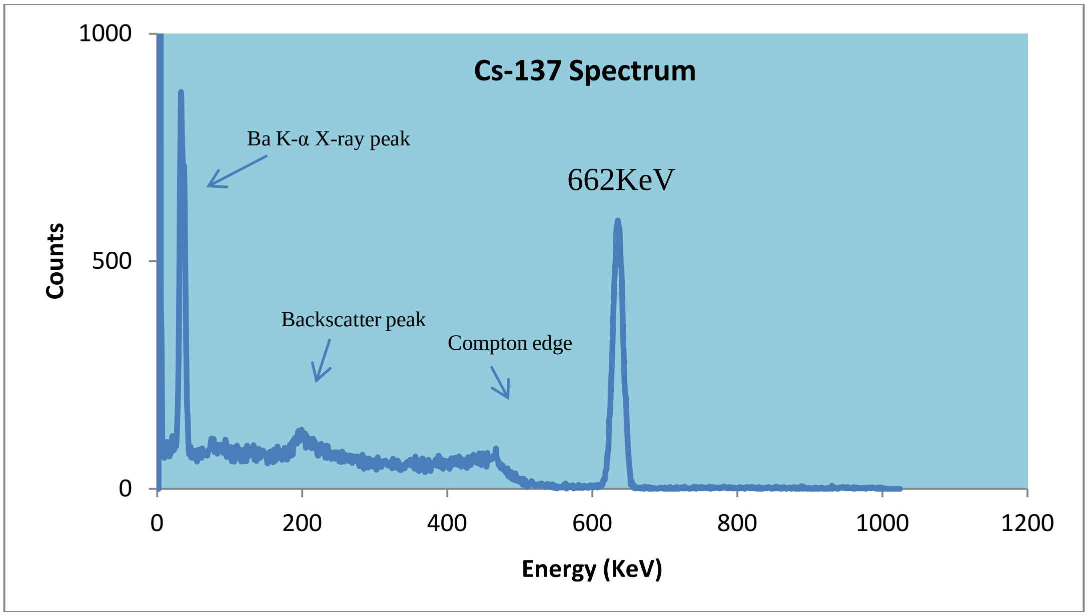


FIGURE 4. ^{137}Cs Spectrum obtained for 300 seconds using $LaBr_3(Ce)$ detector.

From Fig. 3, a power law relationship is obtained from where a linear relationship is deduced by considering the resolution relationship with the reciprocal of energy. This illustrated in Fig. 5.

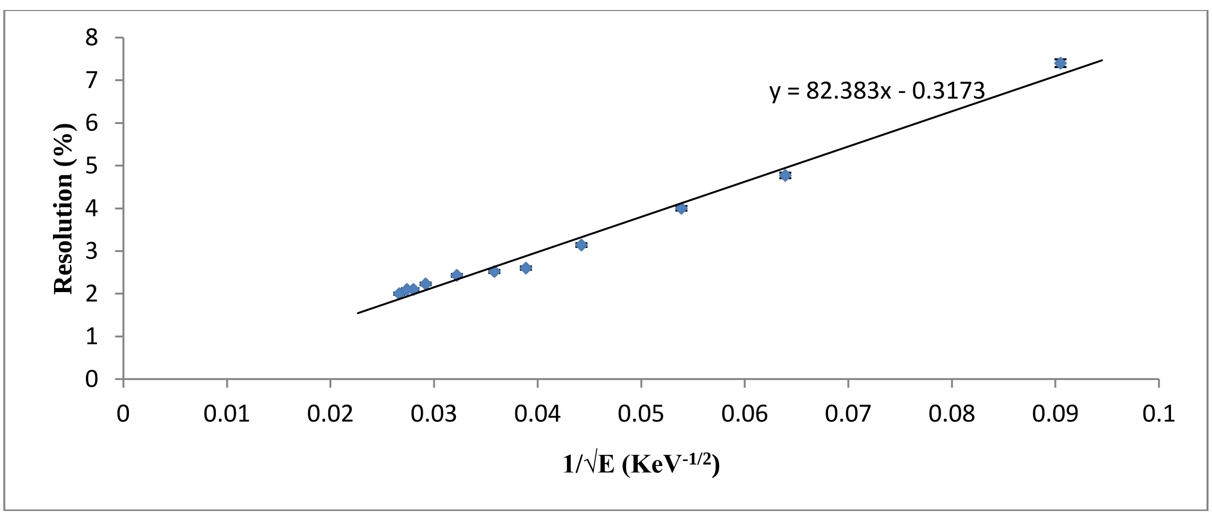


FIGURE 5. Graph showing the linearity of resolution with $1/\sqrt{E}$.

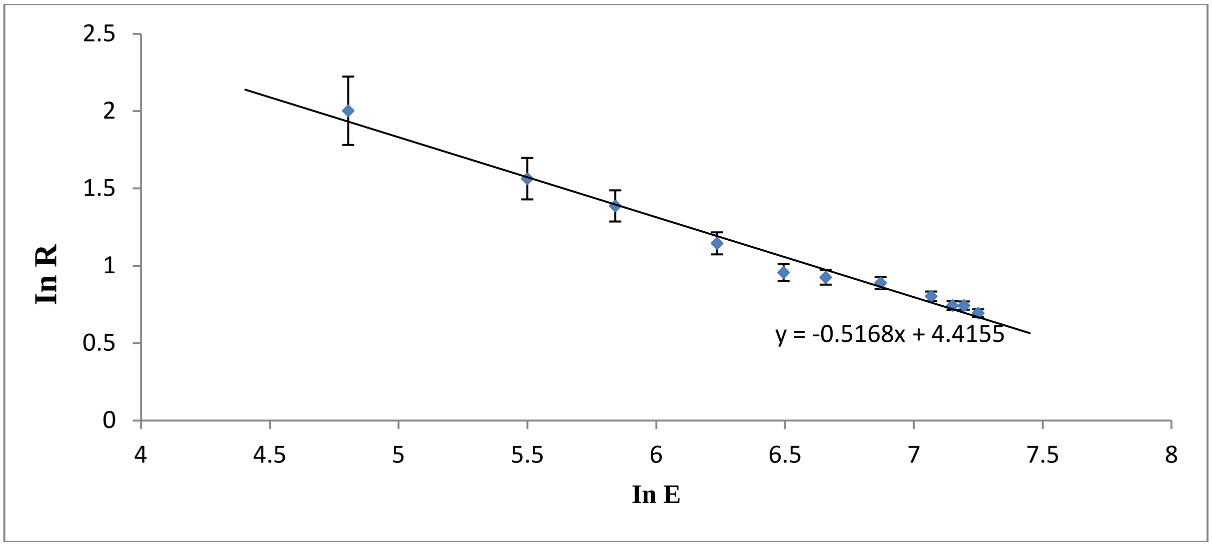


FIGURE 6. Graph of log (resolution) against log (energy).

A plot of log (resolution) against log (energy) as depicted in Fig. 6 yields a theoretical slope of -0.5. Based on the graph, an experimental value of 1.52± 0.06 was obtained for the $LaBr_3(Ce)$ detector and this is in the limit of the theoretical value.

The variation of the intrinsic photo-peak efficiency with energy from 122-1408keV at a source-detector distance of 15cm is shown in Fig. 7, and from it, it seen that the detector efficiency decreases as the gamma ray energy increases. This is because at lower energies, photoelectric absorption is the dominant mode of interaction that contributes to full energy deposition with a large number of pulses [2]. As the photon energy increases other interaction modes such as multi-Compton scattering and pair production sets in and so not all the photon energies contribute to the full- energy peak. As a result, there is a decrease in the detector efficiency. Furthermore, at higher energies the photons spend less time in the vicinity of the detector material so that the probability of interaction is lower.

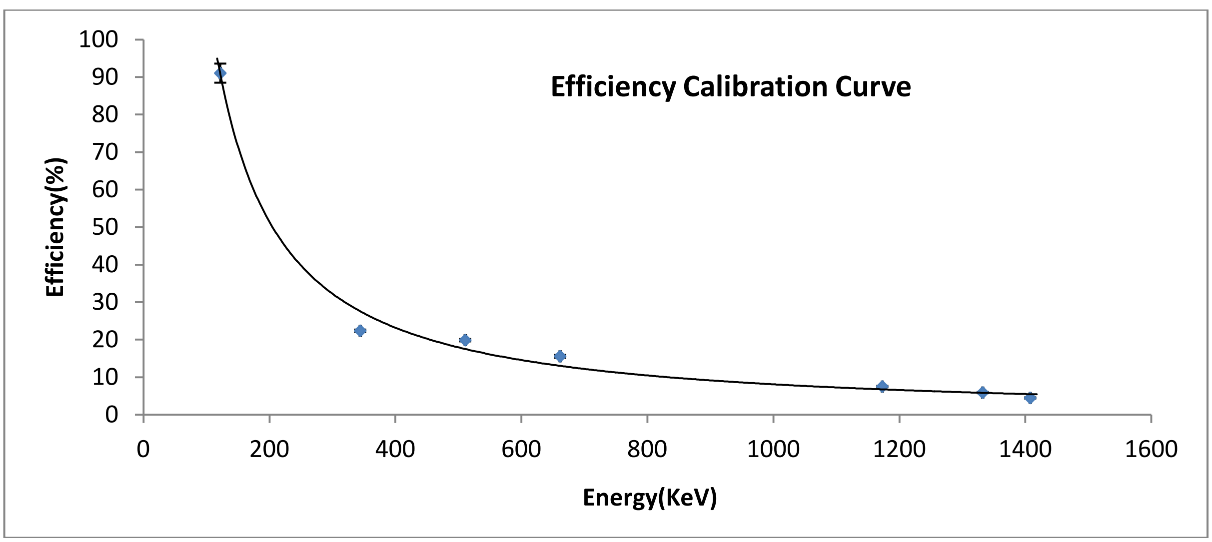


FIGURE 7. Graph of intrinsic photo-peak efficiency against energy.

At 122keV gamma line of ^{57}Co, the $LaBr_3(Ce)$ detector shows a 23.1% higher efficiency than NaI(Tl) detector and at 1332keV gamma line of ^{60}Co, a 6.14% better efficiency is obtained in $LaBr_3(Ce)$ detector compared to NaI(Tl) detector. However the efficiency of $LaBr_3(Ce)$ detector is poorer at higher energies compared to that of BGO detector based on the studies carried out by Evans and Orndoff (Knoll, 2000). This could be due to the thickness of the crystal and the internal gamma line of the $LaBr_3(Ce)$ detector.

VII. CONCLUSION

The characterisation of a 25mm X 25mm Brillance 380 $LaBr_3$(Ce) detector has been carried out and the result compared with other recent measurement reported by other authors for other detectors. Our result reveals that the resolution of $LaBr_3$(Ce) detector is two times better than that of NaI(Tl) detector, and that at an energy of 662keV, a resolution of 2.6% FWHM was obtained at room temperature. It was also determined that, the efficiency of the detector in the energy range 122keV-1408keV is also higher than that of NaI(Tl) detector. Specifically, at an energy of 122keV, the efficiency is found to be 23.1% better than NaI(Tl) detector while at 1332keV a 6.14% better efficiency is obtained compared to NaI(Tl) detector. The detector shows good efficiency at the low energy range 122-344keV (91%-22.3%) and this decreases from 662keV (15.5%) due to the photons spending less time in the detector vicinity at higher energies. We conclude that the reason for this outstanding efficiency is the high density of the detector.

Based on the high potentials of the $LaBr_3$(Ce) detector as revealed from this investigation, this detector should not only be considered as the possible replacement for NaI(Tl) detector but could be outstanding in diversified applications that require fast timing, good energy resolution and efficiency such as in medical imaging (Positron Emission Tomography and Time of Flight measurement), nuclear research and geological exploration.

REFERENCE

[1] Rainer N., *Inorganic Scintillators-a never ending story*, Nuclear Physics News **20**, 27 (2010).

[2] Knoll, G., *Radiation Detection and Measurement*, 3rd Ed. (John Wiley & sons Inc., New edition, 2000).

[3] Regan, P., *REP 2-Gamma ray Spectroscopy*, (University of Surrey Lab Script, Guildford, UK, 2008).

[4] Sellin, P., *Spectroscopy Systems*, (Lecture Notes), (University of Surrey, Guildford, 2009).

[5] Shah, S. K., Glodo, J., Klugerman, K. L., Moses, M., Derenzo, S. and Weber, M., *$LaBr_3$(C)e Scintillatars for gamma ray spectroscopy*, (Lawrence Berkeley National Laboratory, USA, 2002).

[6] Szymanska, K., Achenbach, P. and Agnello, M., *Resolution*, *Efficiency and stability of HpGe detector operating in a magnetic field at various gamma-ray energies*, Nuclear Instruments and Methods in Physics Research **A592**, 486-492 (2008).

[7] Van Loef E. V. D., Dorenbos, P. and Kramer, K. W., *Scintilllation properties of $LaBr_3$: Ce crystals: fast, efficient and high energy-resolution scintillator*, Nuclear Instruments and Methods in Physics Research **A486**, 254-258 (2002).

APPENDIX

A Table of Resolution at various Voltages for ^{60}Co and ^{137}Cs source

Voltage (V)	Resolution		
	^{133}Cs-662KeV	^{60}Co-1173KeV	^{60}C0-1332KeV
480	9.09	8.70	5.30
500	6.67	6.30	4.20
520	5.00	4.40	3.60
540	4.200	3.60	3.10
560	3.47	3.40	2.96
580	3.20	2.51	2.45
600	3.10	2.41	2.28
650	2.90	2.01	1.63
700	2.84	1.76	1.49
750	2.55	1.74	1.19
800	1.83	1.14	0.99
850	1.56	1.12	1.15
900	1.41	1.03	1.00

B Table of Resolution at different Shaping Time

Shaping Time(μs)	FWHM	Channel number, H_o	Resolution $\left(\frac{FWHM}{H_o} \times 100\%\%\right)$	Error
0.5	23.31905	890.04	2.62	1.618641
1	16.29843	634.18	2.57	1.603122
2	10.85198	388.96	2.79	1.670329
4	6.3874	218	2.93	1.711724
8	5.282172	110.97	4.76	2.181742
12	2.77838	76.12	3.65	1.910497

C Table Energy Resolution Parameters

Source	*E(KeV)*	*FWHM (KeV)*	*R (%)* $\left(\frac{FWHM}{E} \times 100\right)$	*In E*	*In R*	$1/\sqrt{E}$	*Error in R*	*Error in In R*
^{57}Co	122.1	9.04	7.40	4.8048	2.00148	0.09049	0.81858	0.22140
^{152}Eu	244.7	11.67	4.77	5.5000	1.562346	0.06392	0.40874	0.13387
^{152}Eu	344.3	13.77	4.00	5.8415	1.386294	0.05389	0.29048	0.10067
^{22}Na	511	16.05	3.14	6.2363	1.144223	0.04423	0.19564	0.07129
^{137}Cs	661.6	17.2	2.60	6.4946	0.955511	0.03887	0.15116	0.05555
^{152}Eu	778.9	19.63	2.52	6.6578	0.924259	0.03583	0.12837	0.04708
^{152}Eu	964	23.43	2.43	6.8710	0.887891	0.03220	0.10371	0.03789
^{60}Co	1173.2	26.16	2.23	7.0674	0.802002	0.02919	0.08524	0.03065
^{22}Na	1274.5	26.77	2.10	7.1503	0.741937	0.02801	0.07844	0.02771
^{60}Co	1332.5	27.98	2.10	7.1948	0.741937	0.02739	0.07505	0.02651
^{152}Eu	1408	28.16	2.00	7.2499	0.693147	0.026650	0.07102	0.02461

D Table of Corrected Source Activity

Sources	Serial No.	Initial Activity, $A_0(KBq)$	Calculated Activity(KBq) $A(t) = A_0 / 2^{t/T_{1/2}}$
^{57}Co	S302.PH	3.499	3.133
^{137}Cs	S172.RP	22.662	22.601
^{60}Co	S317.PH	40.231	39.613
^{22}Na	S311.PH	20.045	19.426
^{152}Eu	S259.PH	152.911	151.977

E Table of Detector Efficiency Parameters

Source	Energy(KeV)	Count Rate, C_P(Sec^{-1})	Activity, D(Bq)	$I_\gamma(E_\gamma)$	$N_\gamma(D \times I_\gamma)$	N'_γ $(N_\gamma \times \frac{\Omega}{4\pi})$	ε_p $(\frac{C_P}{N'_\gamma} \times 100\%)$	Error$(\varepsilon_p \frac{\Delta C_P}{C_P})$
^{57}Co	122.1	4.26	3133.30	0.86	2694.64	4.677892	91.066670	2.547382
^{152}Eu	344.3	15.91	151977.10	0.27	41033.83	71.23473	22.334610	0.323283
^{22}Na	511.0	12.42	20045.00	1.80	36081.00	62.63662	19.828660	0.324842
^{137}Cs	661.6	5.18	22601.01	0.85	19210.86	33.35005	15.532210	0.394010
^{60}Co	1173.2	5.14	39612.54	1.00	39612.54	68.76737	7.474475	0.190344
^{60}Co	1332.5	4.03	39612.54	1.00	39612.54	68.76737	5.860338	0.168543
^{152}Eu	1408.0	2.41	151977.10	0.21	31915.20	55.40479	4.349805	0.161771

Correction in Activity

$$A(t) = \frac{A_o}{2^{t/t_{1/2}}}$$

Where A_0 is the original activity and $A(t)$ is the activity at time, t.

Solid angle of the detector

$$\Omega = \frac{A}{d^2} = \frac{\pi a^2}{d^2}$$

$$Source - detector\ distance, d = 15cm$$

$$radius\ of\ the\ detector, a = 1.25cm$$

$$Solid\ angle = \frac{\Omega}{4\pi} = \frac{1.25^2}{4 \times 15^2} = 1.736 \times 10^{-3}.$$

Table of Absolute Total and Intrinsic Total Efficiency

Source	**Activity, D(KBq)**	**I(E)**	**Total Count Rate, $C_t(sec^{-1})$**	N_γ $(D \times I)$	N'_γ $(\frac{\Omega}{4\pi} \times N_\gamma)$	**Absolute Total Efficiency , ε_a(%)**	**Error in ε_a**	**Intrinsic Total Efficiency,ε_i(%)**	**Error in ε_i**
^{22}Na	19.426	2.8	92.50	54392.8	94.4259	0.170059	0.00102	97.96041	0.58805
^{137}Cs	22.601	0.85	20.46	19210.8	33.35004	0.106502	0.00135	61.34926	0.78306
^{60}Co	39.613	2	72.84	79226	137.5363	0.09194	0.00062	52.96055	0.35826
^{152}Eu	151.977	1.24	240.82	188451.	327.1518	0.127789	0.00047	73.6111	0.27386

Error Propagation

(1) Error in Resolution

$$\left(\frac{\Delta R}{R}\right)^2 = \left(\frac{\Delta FWHM}{FWHM}\right)^2 + \left(\frac{\Delta H_0}{H_O}\right)^2$$

$$\left(\frac{\Delta R}{R}\right)^2 = \left(\frac{1}{FWHM}\right)^2 + \left(\frac{FWHM}{(2.35\sqrt{N})H_0}\right)^2$$

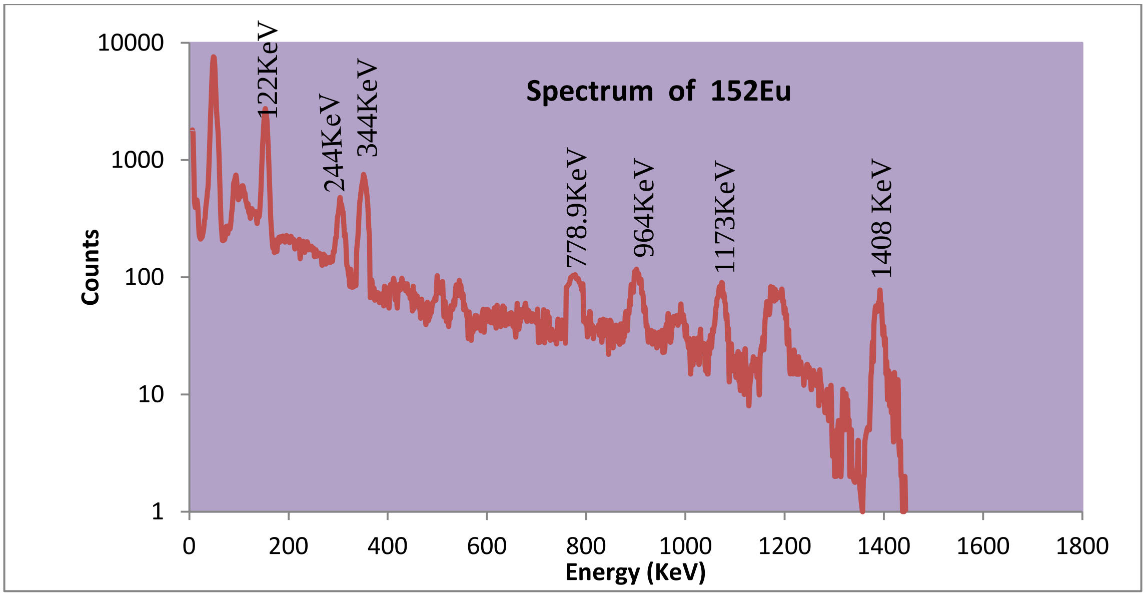
$$\Delta InR = \left(\frac{\Delta R}{R}\right) InR$$

(II) Error in Efficiency

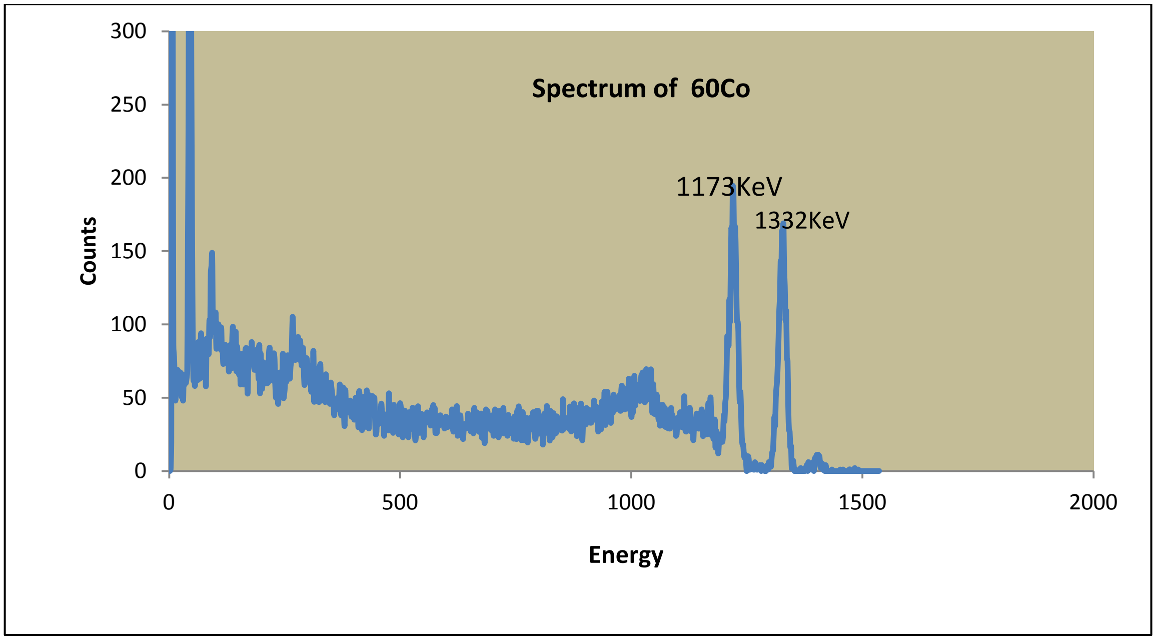
$$\Delta\varepsilon = \left(\frac{\Delta C}{C}\right)\varepsilon.$$

Response of Radioactive Sources in $LaBr_3$(Ce) detector

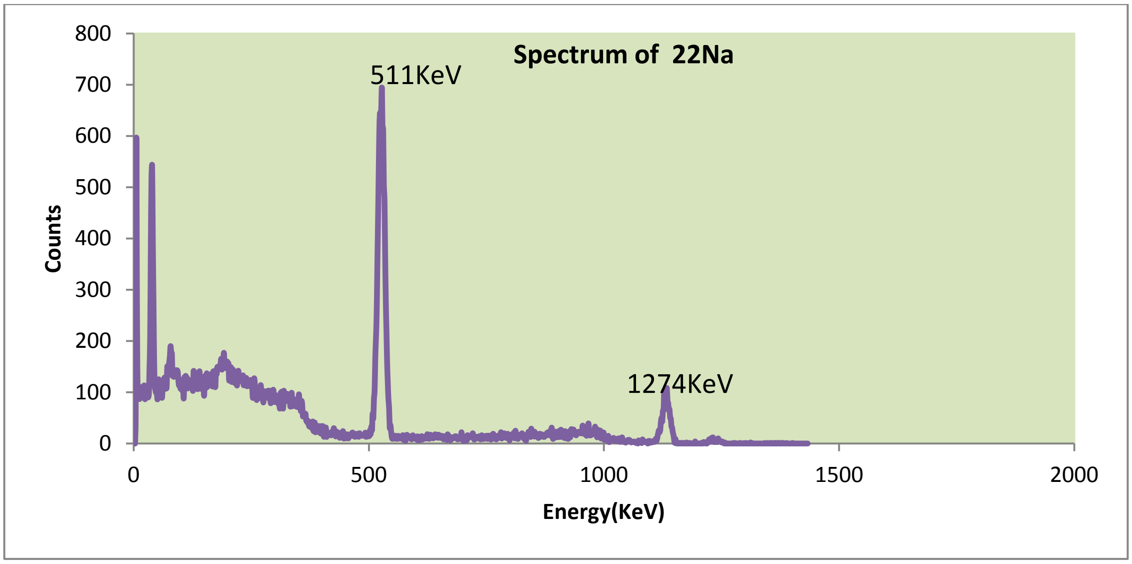
(I) Spectrum of Europium-152 Source



(II) Spectrum of Cobalt-60 Source



(III) Spectrum of Sodium-22 Source



(IV) Spectrum of Cobalt-57