EVALUATION STUDY OF LANTHANUM HALIDE DETECTORS (LABR3:CE , LACL3:CE, NAI(TL), (HPGE)

KarimaAbdulla[1], Abtisam A. Alakrout[2*], Manal Omer[3], and Samah Aghtisa[4]

[1,3,4] General Section, Faculty of Natural Resources Al-Ajelat, Al-Zawia University, Libya
[2] Department of Physics, Faculty of Sciences, Sabratha University, Libya
[*] ebtisamalkroot55@gmail.com

Abstract

Through the use of thesources spectrums ^{22}Na (S311.PH), ^{60}Co (S297.PH), ^{133}Ba (S295.PH), ^{137}Cs (S296.PH) and ^{152}Eu (S285.PH) the properties of Lanthanum Tri-Bromide ($LaBr_3$:Ce) and Lanthanum Tri-Chloride ($LaCl_3$:Ce) scintillation detectors were compared against Sodium Iodide (NaI(Tl)) scintillation and Hyper Pure Germanium (HPGe) semiconductor detectors. In all instances the HPGe detector was found to be superior, So, Hyper Pure Germanium detectors (HPGe) are outstanding devices for radioactivity spectroscopy. In addition the $LaBr_3$:Ce and $LaCl_3$:Ce detectors were always found to be superior to the NaI(Tl) detector.

Keywords: Lanthanum Halide; The scintillation detectors; photopeaks; mapping; peak.

Introduction

Significant research has been conducted in Lanthanum halide crystals recently (A.Iltis et al. (2006)). Saint-Gobain Crystals in conjunction with Deft University have recently (2006) developed, and have been able to routinely grow large BrilLanCe, Lanthanum Tri-Bromide doped with Cerium ($LaBr_3$:Ce), and Lanthanum Tri-Chloride doped with Cerium ($LaCl_3$:Ce), crystals.They are of comparable size to the standard Sodium Iodide doped with Thallium (NaI:Tl) crystals (4 by 4 inch) and sufficiently large to be effective in hand-held Radio-Isotope Identification Devices (RIID). Saint-Gobain Crystals have published a presentation claiming the superiority of the BrilLanCe detectors over current scintillating crystals in the major aspects: Light Yield (photons/keV), Decay Time (Tau, ns), Energy Resolution, Energy Linearity, and resolution (P.R. Menge, Saint-Gobain Crystals 2006).

The new $LaBr_3$:Ce and $LaCl_3$:Ce crystals are expected to maintainall requirements for an RIID crystal namely the fast decay times of scintillating crystals to cope with high activity sources, operating at room temperatures, and having sufficiently good resolution to distinguish complicated spectra, such as Naturally Occurring Radioactive Materials (NORM). (G.Mishra, A.Mitra, et al. 2017).

This project aims to compare the performance of the $LaBr_3$:Ce and $LaCl_3$:Ce detectors against the standard scintillation detector NaI:Tl, and the semiconductor detector Hyper Pure

Germanium (HPGe). The resolution and energy efficiency of Lanthanum halide detectors will be checked against the HPGe and NaI:Tl detectors. This comparison is to measure the characteristics of Lanthanum halide detectors and todecide whether $LaBr_3$:Ce can be a replacement for NaI:Tl. Additional investigation will be conducted to find the effect, and optimum, shaping time for each of the detectors. (M. Lowdon, P. G. Martin, et al, (2019)).

Background Theory

Scintillation Mechanism

Scintillates are one of the oldest types of radiation detector because measurements could be made with photographic film. Images could be collected and intensity measurements could be made. Measurements were also made byobserving the brightness of frequency of flashes in the scintillator with the naked eye. Nowadays the light output is converted into voltage pulses that are processed in the same way as pulses from proportional counters, semiconductor detectors (G.F. Knoll, (1999)).

The scintillation mechanism is the changing of incident radiation energy into UV or visible photon energy (J.T. Bushberg et al. (2001)). The wavelength of the output light can be determined by the crystal composition of the scintillation crystal, and it is chosen to match the optimum wavelength sensitivity of the photo-cathode at the front window of the photo-multiplier tube. The peak emission output wavelength of $LaBr_3$:Ce is 380 nm, which closely matches the optimum response wavelength of a Bialkali photo-multiplier tube (PMT) front window, 390 nm. Light emitted by $LaBr_3$:Ce will have a very high probability of being absorbed by the photo-cathode (G. F. Knoll (2000)).

Efficiency

The efficiency of a spectrum can be classified in two ways, absolute or intrinsic, and sub-categorised as either total or peak. The Absolute Total Efficiency (ε_t) is the ratio of the number of radiation quanta incident on the detector against the total number of radiation quanta emitted by the radiation source:

$$\boldsymbol{\varepsilon_t = \left(\frac{C_t}{N_\gamma}\right) \times 100\%} \qquad \rightarrow \qquad (\mathbf{1})$$

Where C_t isthe ratio of the number of radiation quanta incidentby the radiation source.

Where N_γ is the total number of counts integrated over the whole recorded spectrum and is the number of quanta emitted by the source per unit time:

$$\boldsymbol{N_\gamma = D_s\, I_\gamma(E_\gamma)} \qquad \rightarrow (\mathbf{2})$$

Where D_s is the disintegration rate of the source and $I_\gamma(E_\gamma)$ is the fractional number of gammas emitted per disintegration (Lab script, (2009)).

The Intrinsic Total Efficiency (ε_t) takes into account the solid angle of the surface area of the detector crystal in relation to the spherical surface area of radiation emitted by the radioactive source:

$$\boldsymbol{\varepsilon_t = \left(\frac{C_t}{N'_\gamma}\right) \times 100\%} \qquad \rightarrow (\mathbf{3})$$

Where N'_γ is the total number of gamma-rays which are incident on the detector:

$$\boldsymbol{N_\gamma = \frac{\Omega}{4\pi} D_s\, I_\gamma(E_\gamma)} \qquad \rightarrow (\mathbf{4})$$

Where Ω is the solid angle. (Lab script, (2009)).

The Intrinsic Photopeak Efficiency (ε_p) is the same as the intrinsic total efficiency except only a single photopeak will be taken into consideration:

$$\boldsymbol{\varepsilon_p = \left(\frac{C_p}{N''_\gamma}\right) \times 100\%} \qquad \rightarrow (\mathbf{5})$$

Where C_p is the net number of counts in a photopeak corresponding to E_γ and $N''_\gamma$is the total number of gamma quanta of energy E_γ which fall on the detector crystal:

$$\boldsymbol{N_\gamma = \frac{\Omega}{4\pi} D_s\, I_\gamma(E_\gamma)} \qquad \rightarrow (\mathbf{6})$$

Where $I_\gamma(E_\gamma)$ is the fractional number of photons of energy E_γ emitted perdisintegration (Lab script, (2009)).

Resolution

The resolution (R) is a measure of the width of a photopeak as a function of the value of the centroid of the same peak:

$$\boldsymbol{R = \left(\frac{FWHM}{H_0}\right) \times 100\%} \qquad \rightarrow (\mathbf{7})$$

Where *FWHM* is the Full Width Half Maximum of a specific photo-peak and H_0 is the centroid of the same photo-peak (Lab script, (2009)).

Methodology

The five sources spectrums ^{137}Cs, ^{60}Cs, ^{22}Na, ^{133}Ba and ^{152}Eu were used to study the interaction of gamma rays with the four detectors $LaBr_3$, $LaCl_3$, HPGe and NaI:Tl.

Calibration

Each of the four detectors were calibrated using the source ^{152}Eu (S285.PH), focussing on the full-energy photo-peaks 121.78, 244.7, 344.27, 778.9, 964, 1112.05 and 1407.92 keV.

Energy Efficiency and Resolution

Additional sources ^{60}Co, ^{22}Na, ^{133}Ba and ^{137}Cs spectra were taken for 10 minutes using the energy calibration already calculated using the ^{152}Eu source. The total number of counts for the whole spectra and underneath the main photo-peaks was counted and the energyefficiency was calculated using the equations (1), (3) & (5). The resolution was calculated using the equation (7) and the centroid channel and FWHM of each of the main photo-peaks. All significant values are measured using the intrinsic line fitting algorithms of Maestro.

Shaping time

Parts 3.1 to 3.3 were repeated for a range of shaping time values from 0.5μs to 10μs using an analogue electronics set-up.

Digital vs. Analogue

The digital set-up replaced the analogue set-up for both the $LaBr_3$ and $LaCl_3$ detectors, and the range of shaping times 0.75μs to 2μs, to investigate the possible differences caused.

Results and Discussion

Resolution

Figure (1) shows that the $LaBr_3$:Ce and $LaCl_3$:Ce were found to be of similar values across the entire energy range from 121.78keV to 1407.92keV and substantially better than NaI(Tl) at lower energies, but not at higher energies. Across the entire range of energies, the HPGe detector has better resolution than the other three detectors.

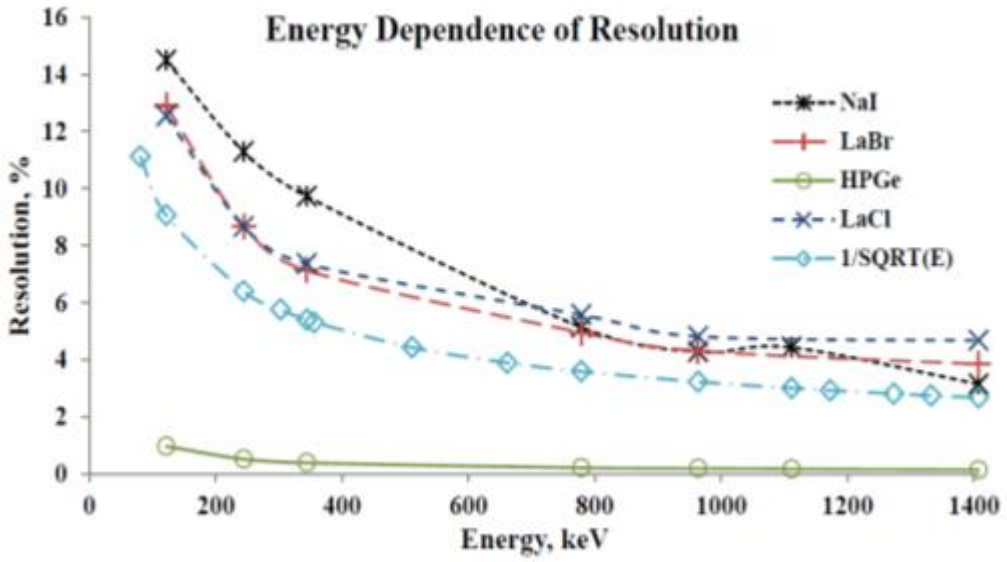

Figure (1): The Dependence of Resolution on Energy for HPGe, NaI(Tl), $LaBr_3$(Ce) and $LaCl_3$(Ce) Using the Photopeaks Found in only the ^{152}Eu.

The specific resolutions obtained from the 662keV photopeak in the ^{137}Cs spectrum are 6.54% for NaI(Tl), 6.2% for $LaCl_3$:Ce, 5.3% for $LaBr_3$:Ce and 0.23% for HPGe.

Linearity of Energy response

The ^{152}Eu photopeaks used during calibration were also used to determine any non-linearity in any of the detectors. Figure (2) shows that the HPGe has the smallest variation of recorded energy against the actual peak energies, of around 0.1%. NaI(Tl) recorded below the actual energy at lower energies and as the energy increased above 500keV it moved to read higher and at 1,408keV it was again reading below the actual energy. At the lower energies $LaBr_3$ (122kev and 245keV) read above the actual energy by almost 1% but from energies above this it was always within 0.2%.

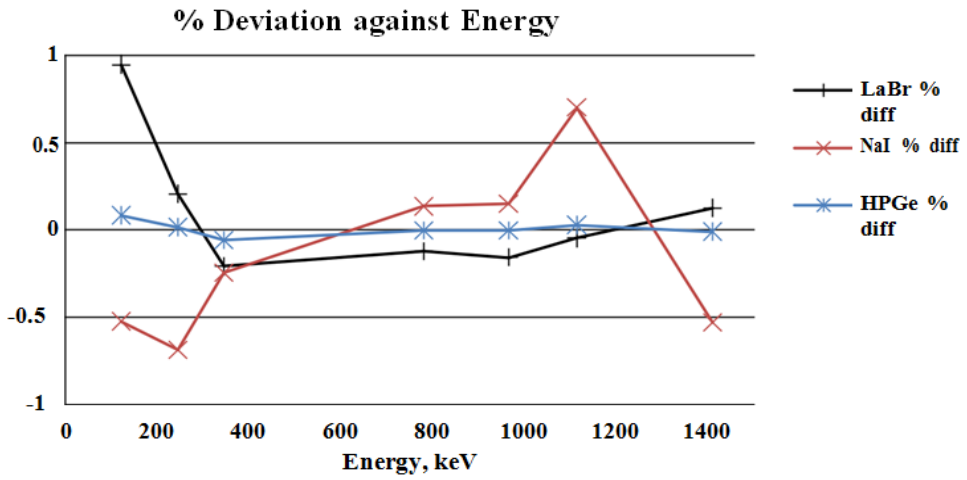

Figure (2): The% Deviation of the Calibrated Peaks Against the Expected Values for the Photopeaks.

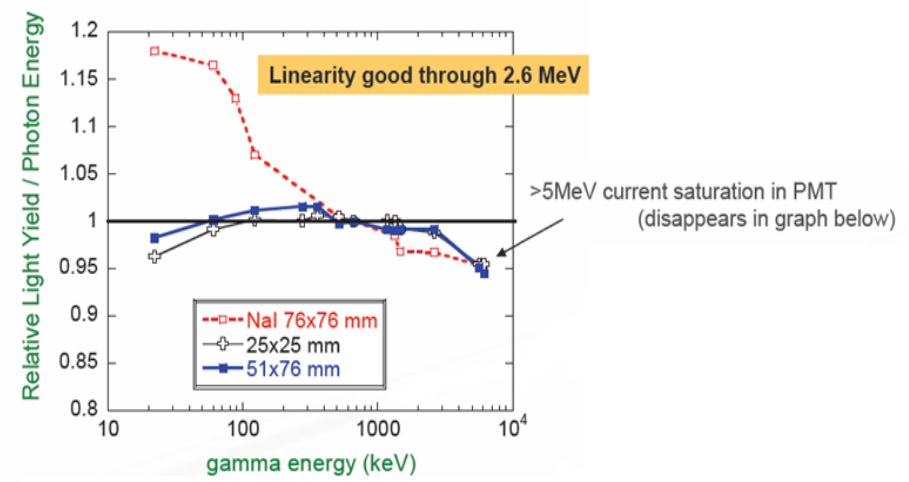

Figure (3): Saint-Gobain Crystals Calculated Variation of Linearity Against Gamma Energy for the BrilLanCe380 $LaBr_3$:Ce (Peter R. Menge, Saint-Gobain Crystals (2006)).

Figure (3) shows the relative output of the detectors against the energy of the gamma photons. A relative light yield / Photon energy of 1 means the energy recorded by the detector equals the energy of the photon. A number greater than 1 means that the detector would record a higher energy than the incident photon, and when the relative light yield / Photon energy falls below 1 the detector would record energies of less than that of the initial photon. Figure (3) compares 2 different sizes for a $LaBr_3$:Ce detector against a NaI:Tl. It can be seen that the variation in the physical size of the $LaBr_3$ makes a little difference to its linearity of response. Only at very low photon energies (less than 100keV), there is a real difference with the smaller crystal producing less relative light yield than the larger crystal. The NaI however has a much less linear response at very low photon energies (around 20keV) the relative light

output is more than 15% greater than it should be. In the experiments, we carried out the lowest energy, 122keV from the ^{152}Eu source, which had a 1% variation. At photon energies between 500keV and 1MeV the linearity of the NaIislike the $LaBr_3$linearityi.e. very good, but at energies greater than 1MeV it drops.

Energy Efficiency

Table (1): The Absolute Total and Peak Efficiency Calculated Forthe Entire ^{137}Cs Spectrum and the ^{137}Cs 662 Kev Peak

Detector	Absolute Efficiency	Absolute Peak Efficiency
LaBr3:Ce	1.44	8.20
LaCl3:Ce	1.38	7.86
HPGe	0.16	1.80
NaI:Tl	1.31	5.32

Table (1) compares the measured efficiency in count per emitted gamma-ray for ^{137}Cs (S296.PH) source, which has 662 KeV photo peak energy. It is clear from the table that the **$LaCl_3$:Ce** and **$LaBr_3$:Ce** detectors provide better efficiency performance.

Furthermore, the Absolute efficiency values of the **$LaCl_3$:Ce** and **$LaBr_3$:Ce**are about 1.48 and 1.54 times that of**NaI(TI)** respectively.

Shaping Time

Figure (4) shows the relationship between the resolution for the four detectors using the 137Cs 662 keVphotopeakof the detectors and the shaping time. As expected the HpGe detector has very good resolution and the NaI is poorer but extremely stable. The LaBr detector is shown to have slightly better resolution than the sodium detector and appears to perform at its best around 1.25-1.50 μs though across the tested range it is fairly consistent. The LaCl detector appears to be much more affected by the changing of shaping time; with a much better performance occurring when the response is fast, at 0.5 μs. This would indicate that this detector would be suitable in areas where high activities are expected.

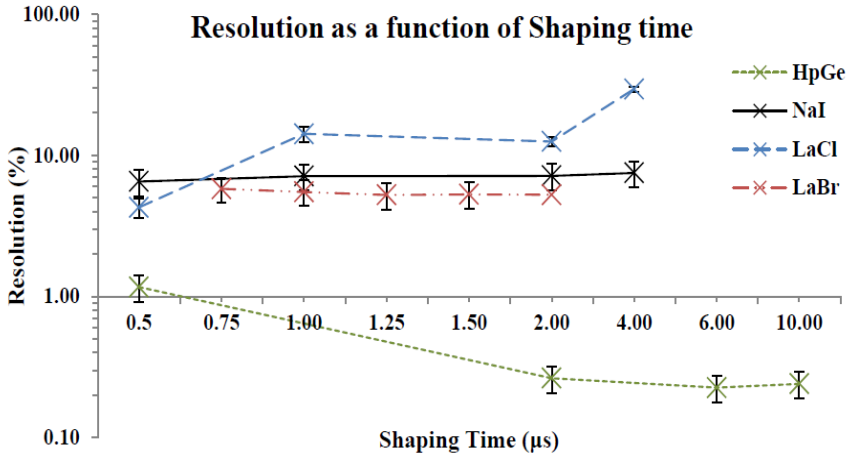

Figure (4): Shows How Resolution Varies with Shaping Time for the Four Detectors Using the ^{137}Cs 662 keVphotopeak.

As stated, it is expected that the efficiencies for the detectors are to remain stable despite changing the shaping times, however, as can be seen in Figures (4) & (5) the readings for the benchmark detectors are very consistent with the HpGe performing poorly due to the size of the crystals depletion region.

The Lanthanum detectors however appear to vary with shaping time and both have lower absolute efficiencies at longer shaping times which could indicate an increase in deadtime.

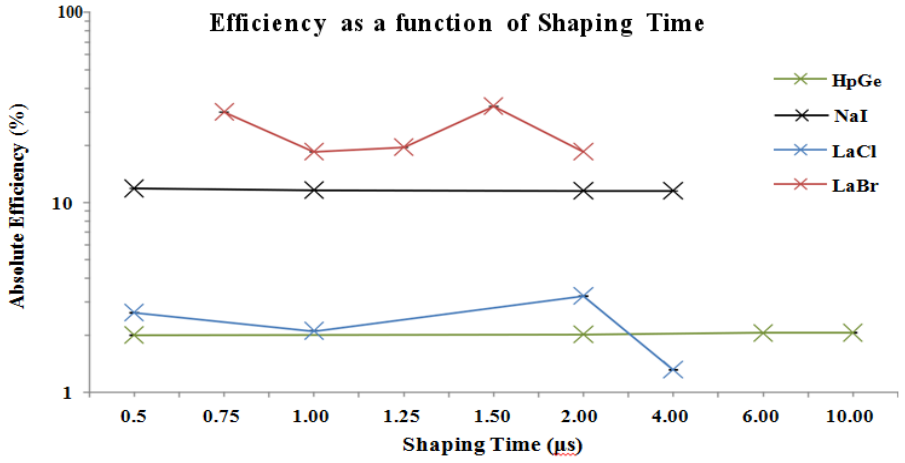

Figure (5): Shows how Absolute Efficiency Varies with Shaping Time for the Four Detectors Using ^{152}Eu.

The absolute peak efficiency shown in Figure (6) for the LaBr is particularly interesting as it shows that above a shaping time of 1μs there is a sharp decrease. The value of the LaCl detector at 4μs is probably spurious and down to a fault in the equipment or reading.

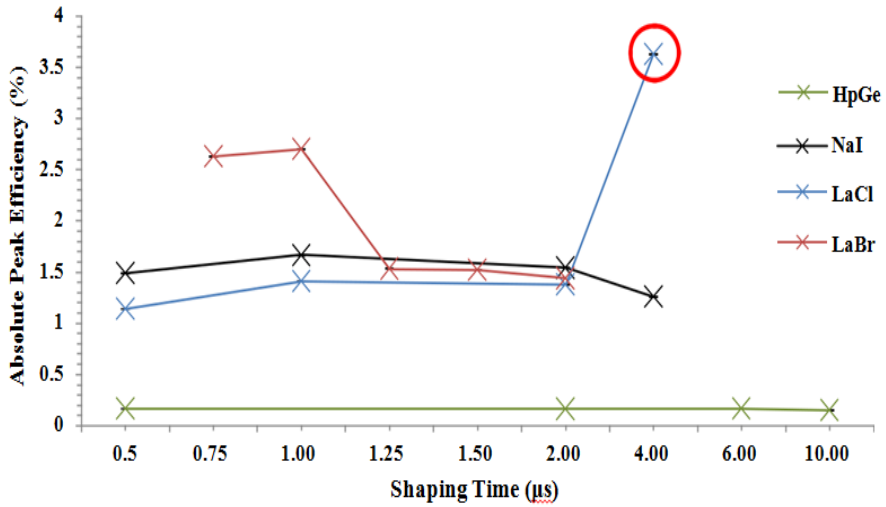

Figure (6): Shows how Absolute Peak Efficiency Varies with Shaping Time for the Four Detectors Using the ^{137}Cs 662 keVphotopeak.

Conclusions

In this paper, the scintillation detectors (NaI:Tl, $LaBr_3$:Ce and $LaCl_3$:Ce) show a similar shape in Figure (2) as the curvethat confirms the dependence of the resolution of the scintillation detector on the poisson statistics of the output light pulses from the scintillation crystal to the photocathode. The NaI detector having better resolution at higher energies but not at lower energies is the opposite of what was expected. The expected value for $LaBr_3$:Ce resolution is 2.8% (P.R. Menge, Saint-Gobain Crystals (2006)), but this could not be achieved. For Figure 1, the resolution has a higher value than expected and it can be attributed to the Compton scattering from the higher energy photopeaks leaking down and widening the photopeaks.

HPGe detectors have the best resolution, typically ~1%, which gives well defined photopeaks and thus allows very precise calibrations. NaI:Tl has a typical resolution of ~6% and therefore has broad photo-peaks and the energy calibration will not be as precise. $LaBr_3$:Ce has an energy resolution in between these two of 2.8% at 662keV. (Peter R. Menge, Saint-Gobain Crystals (2006)) The $LaBr_3$ was overall comparable to NaI but not as precise as the HPGe detector.

Ideally, all detectors would have a linear response across the entire range of energies; however, this is not the case. In the experiments we have done, they are all within 1% of the

actual peak energy. Over the range of energies observed the HPGe has the best linearity while the $LaBr_3$ and NaI are not quite as good. When a detector material is selected, it is important that it has a linear response across the range of energies it is to be used to look at, which is one of the reasons that these materials are used in gamma ray detection, so it is of no surprise that they have such good linearity.

The energy efficiency is greater for the $LaBr_3$:Ce and $LaCl_3$:Ce than the NaI detector, which is what is expected for the density and Z number for the scintillating crystals (A.Favalli et al. (2008)).

In most detectors, there is a trade-off between the resolution and the event rate or activity of the source. This is due to the longer shaping time reducing the ballistic deficit meaning all of the slow charge carries are collected and thenoise from the amplifieris reduced. However, a long shaping time means that the detector cannot be used for a longer period whilst it recovers (dead time) and this reduces its effectiveness in high fluxes.

The efficiency of a detector should remain stable unless there is a large increase in the dead time causing less of the radiation to be detected that would indicate that the relative position of the detector to the source would have to be changed to get a more accurate result.

References

- A. Iltis et al. (2006), "Lanthanum halide scintillators: Properties and applications". Nuclear Instruments and Methods in Physics Research, pp. 359-363.
- P. R. Menge, Saint-Gobain Crystals (2006), "Performance of large BriLanCe 380(lanthanum bromide) scintillators". SORMA XI.
- J. T. Bushberg et al. (2001), "The Essential Physics of Medical Imaging" .2nd Edition. Lippincott Williams & Wilkins.
- G. F. Knoll (2000), "Radiation Detection and Measurement" .3rd Edition. John Wiley & Sons, Inc.
- Lab script, (2009), " REP2 – Gamma-Ray Spectrometry". University of Surrey.
- B.D. Milbrath et al. (2007), "Comparison of $LaBr_3$:Ce and NaI:Tl scintillators for radio-isotope identification devices".Nuclear Instruments and Methods in Physics. pp.774-784.
- D. Yuan et al. (2011), "Computational removal of lanthanum-cerium bromide self-activity" .Nuclear Instruments and Methods in Physics Research A.

- F.G.A. Quarati et al. (2010), "High energy gamma-ray spectroscopy with $LaBr_3$ scintillation detectors". Nuclear Instruments and Methods in Physics Research A. pp.157-169.

- G.Mishra, A.Mitra ,et al. (2017), "Characterization of the CeBe3 scintillator detectors" .Pro of the DAE Symp. On Nucl.phys.62 (1092).

- M. Ciemala et al. (2009), "Measurements of high-energy gamma-rays with $LaBr_3$:Ce detectors". Nuclear Instruments and Methods in Physics Research A. pp. 76-79.

- A.Owens et al. (2007), "The hard X-ray response of Ce-doped lanthanum halide scintillators".Nuclear Instruments and Methods in Physics Research A. pp. 158-162.

- A.Favalli et al. (2008), "Wide energy range efficiency calibration for a lanthanum bromide scintillation detector", Radiation Measurements, Vol. 43, pp. 506-509.

- G.F. Knoll, (1999), "Radiation Detection and Measurement".3rd Edition (Chapters 16 to 18), John Wiley & Sons.

- M. Lowdon, P. G. Martin, et al, (2019), " Evaluation of Scintillator Detection Materials for Application within Airborne Environmental Radiation Monitoring". Sensors 2019, 19, 3828.