Calculation of scintillation properties of Ø1”×1” of the lanthanum bromide scintillation detector using MCNP simulation and experiment

Hamad Haidar, Fang Liu\textsuperscript{1} and Hang Yuan

Beijing Key Laboratory of Passive Safety Technology for Nuclear Energy, North China Electric Power University, No.2, Beinong Road, Huilongguan, Changping District, Beijing 102206, China

\textsuperscript{1}Email: liuf@ncepu.edu.cn

Abstract. The purpose of this study is to develop a model for gamma spectroscopy for calculation of full width at half maximum (FWHM), energy resolution and full-energy peak absolute detection efficiency of Ø1”×1” of the lanthanum bromide LaBr$_3$(Ce) detector crystal. For this purpose, a complete detector model was developed for Monte Carlo N-Particle (MCNP) transport code. In MCNP simulation, F8 tally was used as it gives the energy distribution of pulses created in the detector. Whenever incident gamma ray enters into detector volume, due to interactions short pulses are produced inside the detector volume. The pulse height spectra of various gamma sources were obtained, which were used to calculate different scintillator properties of our interest. In the simulation, the distance from the source to the detector face was varied to see the effects on scintillation properties of the detector crystal. The detector crystal offers the energy resolution of 3.455\% and the FWHM of 22.81 KeV when gamma source Cs$^{137}$ (662 KeV) was used. As we increased the incident gamma ray energy, the FWHM of the detector crystal increased, while the energy resolution and absolute detection efficiency of the detector crystal decreased. The scintillation properties of Ø1”×1” of the LaBr$_3$(Ce) detector crystal were studied experimentally. In our case, it was observed that experimental and simulation results coincided. This proves that the detector model we built for gamma spectroscopy in MC transport code is accurate.

1. Introduction

In nuclear physics, gamma detection techniques are widely used in gamma spectroscopy. Scintillator detectors are widely used in health physics, industry, energy and environmental applications. [1] NaI(Tl) scintillator detectors have been broadly used in many fields over the last 50 years [2]. Recently, the new lanthanum based scintillators have become commercially available [3]. When Ø19×19 mm$^3$ of LaBr$_3$(Ce) detector crystal was studied experimentally, a light yield of 61000 photons per MeV of absorbed gamma ray energy (ph/MeV) and the energy resolution of 3.4\% was observed at 662 KeV. [4] Similarly, when Ø2”×2” of a LaBr$_3$(Ce) detector crystal was studied, the energy resolution of 3.4\% was observed at 662 KeV. [2] In literature, scintillation properties of the LaBr$_3$(Ce) detector crystal have been studied experimentally but these values were not well justified by simulation results. In this study, scintillation properties of Ø1”×1” of the LaBr$_3$(Ce) detector crystal were studied experimentally and a complete detector model was developed for Monte Carlo N-Particle...
For gamma ray spectroscopy, NaI(Tl) and HPGe scintillator detectors have been used in last several years. But in recent years, lanthanum bromide LaBr₃ (Ce) scintillator detector was found which have good scintillation properties as compared to other scintillator detectors. [4] When Ø3"×3" of the LaBr₃ (Ce) and NaI(Tl) detector crystals were exposed to gamma rays of energy 662 KeV, the LaBr₃ (Ce) detector crystal showed the energy resolution of 2.9% and the light yield of 61000 photons per MeV of absorbed gamma ray energy (ph/MeV) while the NaI(Tl) detector crystal showed the energy resolution of 7% and the light yield of 40000 photons per MeV of absorbed gamma ray energy. [5]

The Monte Carlo calculation technique can be applied to a wide variety of applications in the radiation field, such as radiological protection, nuclear installations, shielding and detectors modeling among several others purposes. [6] MCNP simulation technique was used to validate the NaI(Tl) detector model. [6] Using this technique, full-energy peak detection efficiency and energy resolution of Ø1.5"×1" of a NaI(Tl) detector was calculated in energy range of 20 to 662 KeV. [6] By using this idea, a complete detector model was developed for Monte Carlo transport code to see the effects on scintillation properties of the LaBr₃ (Ce) detector crystal by changing distance from the source to the detector face and to verify experimental results.

Performance of gamma spectrometry is greatly dependent upon the full-energy peak absolute detection efficiency of the detector crystal [7]. In this study, a complete detector model was developed for Monte Carlo transport code to calculate scintillation properties of Ø1"×1" of the LaBr₃ (Ce) detector crystal and to see the effects on scintillation properties by changing distance from the source to the detector face. The scintillation properties of the same size detector crystal were studied experimentally.

2. Material and methods

2.1. Monte Carlo simulation

MCNP is a general purpose code of Monte Carlo that can be used for the electron, neutron and photon or coupled neutron-photon-electron transport. With the great database of cross section, MCNP can simulate these particles from 1 KeV to 100 MeV. [8] The detector model that was fed as input to the Monte Carlo code was explained, followed by a description of the physical quantities calculated. The geometrical model of the LaBr₃ (Ce) detector crystal for the experimental measurements was simulated as three concentric cylinders. The first cylinder represents the sealed photomultiplier chamber’s aluminum housing (1.67-cm radius and 3.34-cm height). The second cylinder simulates the reflecting material (1.4-cm radius and 2.94-cm height) and the third one represents the LaBr₃ (Ce) detector crystal (2.54-cm radius and 2.54-cm width). While hollow cylinder with one end closed (outer radius 6.73 and inner radius 1.73) simulates the lead shield. The user code must contain all of the information about the radiation source and the detector geometry. F8 tally in MCNP provides the energy distribution of pulses created in the detector volume without any variance reduction. [8]

![Figure 1. Monte Carlo simulation model.](image)
Figure 1 shows Monte Carlo simulation model of the LaBr₃ (Ce) scintillator detector, which shows the package structure and materials in detail. The detector crystal geometry was modeled in accordance with its package structure available for experiments. If modeled parts were not the same as in package structure available for experiments, then there might be an error in results. To validate a simulation model that represents the interaction between detectors and particles measured, the pulse height simulation in MCNP was given a more realistic Gaussian energy distribution. MCNP has a special tally option in which generated parameters from experimental data can be input to the simulation in order to give the spectra the required Gaussian shape. [9] The GEB tally gives the detector simulated data a Gaussian shape by using the unbroadened energy input with the calculated spectral data and user-specified tally inputs to solve equation 1

$$f(E) = C e^{\left( \frac{E - E_0}{A} \right)^2}$$  \hspace{1cm} (1)

Where $E$ = broadened energy, $E_0$ = unbroadened energy of the tally, $C$ = normalization constant, and $A$ = Gaussian width, related to the FWHM by

$$A = \frac{FWHM}{2\sqrt{2\ln 2}}$$  \hspace{1cm} (2)

The (FWHM) of real experimental data was indirectly provided by the user by specifying the three parameters (a, b, and c) in equation 3 required by the GEB tally

$$FWHM = a + b\sqrt{E + cE^2}$$  \hspace{1cm} (3)

Where $E$ is the energy of incident gamma ray [9]. The GEB parameters values for this particular experimental detection system setup were calculated using the FWHM data acquired from experimental measurements of two gamma sources and by performing a nonlinear least square analysis using origin pro 9 software. The GEB parameters values could be better if we could get the measured FWHM data of at least five gamma sources. These parameters changed when distance was varied between the source and face of scintillator detector.

In simulation, $\gamma$-radiation sources (Co⁶⁰, Mn⁵⁴, Eu¹⁵², and Cs¹³⁷) were used. These sources produce gamma photons of energy 344, 662, 834, 1173 and 1332 KeV. The distance from the source to detector face was varied to see the effects on scintillation properties of the detector crystal. When less number of particles were run, statistical error in counts was above 15%. In order to limit error within 5%, each simulation was done using $1 \times 10^9$ particles.

2.2. Experimental setup

Ø1"×1" of the LaBr₃ (Ce) scintillation detector manufactured by Saint Gobain company were used for experiments. The scintillator crystal was contained in an aluminum sealed housing with a glass entrance window. [3]
We can adjust the high voltage (HV) on computer easily by using CANBERRA's basic spectroscopy software.

The multichannel analyser (MCA) is an important laboratory instrument which can measure distributions of input signals consisting of pulses. It operates in two different modes: pulse height analyser (PHA) mode and multichannel scaler (MCS) mode. In PHA mode, the input pulses are sorted into bins (channels) according to their amplitude. [10] The PMT converts an extremely weak light output of a scintillation pulse into the corresponding electrical signal. [10] Lead (Pb) shields were used to reduce the background level of the system, to reduce the interference of visible light and ensure that the measuring environment was dark. For each measurement, HV was set at 625 V and experiment was run for one thousand seconds. The pulse height spectra of two radioactive sources were obtained and analysed.

3. Results and discussion
Scintillation properties Ø1"×1" of the detector crystal are discussed below one by one.

3.1. Full width at half maximum
The Full Width at Half Maximum (FWHM) is defined as the width of the distribution at a level that is just half the maximum ordinate of the peak. As the width of the distribution becomes smaller, we can get more details about incident ionizing radiation. [10]

![Figure 3](image_url)

**Figure 3.** Measured and simulated pulse height spectrum of Cs$^{137}$.

Figure 3 is the measured and simulated pulse height spectrum of Cs$^{137}$ (662 KeV). From figure 3 it is clear that the simulated pulse height spectrum aligns with the experimental pulse height spectrum especially in peak area. The gaps between the experimental and simulation results in low energy region are due to reflection and scattering of gamma rays in real experiment setup which cannot be simulated in simulation. To get desirable results, single peak analysis was done by using origin pro 9 software.

| Nuclide | Energy of photo peak (KeV) | Experimental FWHM (KeV) | Simulated FWHM (KeV) |
|---------|---------------------------|-------------------------|----------------------|
| Eu$^{152}$ | 344 | 15.18 |
| Cs$^{137}$ | 662 | 22.85 | 22.09 |
| Mn$^{54}$ | 834 | 26.60 |
| Co$^{60}$ | 1170 | 32.9424 | 31.6 |
| Co$^{60}$ | 1330 | 33.3347 | 32.7 |

In table 1, experimental and simulated FWHM for Ø1"×1" of the detector crystal are presented. Due to lack of gamma sources, experimental values of the detector crystal for Cs$^{137}$ and Co$^{60}$ were
presented. For gamma source $\text{Cs}^{137}$ (662 KeV), detector crystal showed the FWHM of 22.85 KeV. In literature, reported experimental value for the FWHM of Ø1”×1” of the detector was 22.51 KeV. [11] In our case, experimental value is 22.85 KeV, which is quite close to the reported one. The same is true for other gamma sources.

Figure 4. FWHM as a function of incident gamma ray energy.

From figure 4, it is seen that as incident gamma ray energy increases, the FWHM of the detector crystal increases. The reason is that at low energy gamma rays dominant mode of interaction is photoelectric absorption, which contributes to full energy deposition with a large number of pulses. [12] As gamma ray energy increases Compton scattering and pair production modes of interaction become dominant, so not all gamma rays add to the full-energy photo peak. Due to which full-energy photo peak decreases and widens. It is also seen that the FWHM of the detector crystal is not dependent on distance since its value did not vary by changing the distance from the source to the detector face.

3.2. Energy resolution
The energy resolution ($R$) is defined as the FWHM divided by the energy central of photo peak.

$$R = \frac{\text{FWHM}}{E} \times 100$$ (4)

Where $R$ is energy resolution, FWHM is the full width at half maximum of photo peak and $E$ is the energy central of photo peak. [10] The FWHM, we calculated for Ø1”×1” of the LaBr$_3$ (Ce) detector crystal, was used for calculation of energy resolution.

Table 2. Experimental and simulated energy resolution values of the detector crystal.

| Nuclide | Energy of photo peak (KeV) | Experimental Energy R (%) | Simulated Energy R (%) |
|---------|---------------------------|---------------------------|-----------------------|
| Eu$^{152}$ | 344                      | 3.451662                  | 4.53488               |
| Cs$^{137}$ | 662                      | 3.336858                  |                       |
| Mn$^{54}$ | 834                      | 3.18944                   |                       |
| Co$^{60}$ | 1170                     | 2.6939471                 | 2.4586466             |
| Co$^{60}$ | 1330                     | 2.506368                  |                       |

In table 2, experimental and simulated energy resolution values for Ø1”×1” of the LaBr$_3$ (Ce) detector crystal are presented. For gamma source $\text{Cs}^{137}$ (662 KeV), detector crystal showed the energy resolution of 3.4516%. In literature, reported experimental energy resolution for Ø1”×1” of the LaBr$_3$ (Ce) detector crystal was 3.2%. [11] Only for gamma source $\text{Cs}^{137}$ (662 KeV), our value differs from the reported one. It may be because of different packing of crystal. In our case, we used basic package scintillator but for other gamma sources, our values are comparable with the reported values from the literature. An Error in experimental results may occur due to counting statistics, energy linearity of PMT and interference of light. Because of high light yield of the LaBr$_3$ (Ce) detector crystal combined
with a very fast decay, the peak currents flowing through the photomultiplier tube are high. [4] PMT available in laboratory was not suitable for the LaBr$_3$ (Ce) detector crystal. Which is a major cause of an error in experimental results.

![Energy resolution as a function of incident gamma ray energy.](image)

Figure 5. Energy resolution as a function of incident gamma ray energy.

From figure 5 it is seen that as incident gamma ray energy increases, the energy resolution of the detector crystal decreases. It is also seen that energy resolution of detector crystal is not dependent on distance since its value did not vary by changing the distance from the detector face to the source. The PMT and other electronic devices did not take into consideration in MCNP simulation, which was the minor cause of an error in simulation results. Also, values of GEB parameters were determined by using only two gamma sources which also produced an error in simulation results. But, a good agreement was found between simulation and experimental results.

### 3.3. Full-energy peak absolute detection efficiency

Full-energy peak absolute detection efficiency is defined as

$$\varepsilon_{MC} = \frac{N_C}{N_S}$$  \hspace{0.5cm} (5)

Where $\varepsilon_{MC}$ is the Monte Carlo full-energy absolute detection efficiency, $N_C$ is the number of counts under full-energy photo peak and $N_S$ is the number of photons emitted by source per batch. [2]

$$\varepsilon_{exp} = \frac{N}{A \cdot t \cdot P}$$  \hspace{0.5cm} (6)

Where $\varepsilon_{exp}$ is the experimental full-energy absolute detection efficiency, $N$ is the number of counts detected by detector crystal, $A$ is the radionuclide activity, $t$ is the counting time of experiment and $P$ is the emission probability of gamma ray being measured. [13] Number of counts detected by the detector crystal are equal to number of counts under full-energy photo peak. Absolute detection efficiency of the detector crystal depends upon many factors such as volume, the shape of the detector crystal, material and attenuation layers in front of the detector crystal and distance between the detector face and source. [7] In this study, distance from the source to the detector face was varied to see the effects on absolute detection efficiency. In table 3, activity and emission probability of gamma rays for various radioactive sources are given.

| Nuclide | Energy of photo peak (KeV) | Activity (nano Ci) | Emission Probability (%) |
|---------|---------------------------|--------------------|--------------------------|
| Cs$^{137}$ | 662                       | 970                | 85.30                    |
| Co$^{60}$ | 1173                      | 843                | 99.85                    |
| Co$^{60}$ | 1332                      | 843                | 99.98                    |
Figure 6. Absolute detection efficiency of the detector crystal as a function of gamma ray energy.

First, effects on absolute detection efficiency of the detector crystal by changing gamma ray energy was studied, while distance from the source to the detector face was kept constant. Figure 6 is the graph for absolute detection efficiency of the detector crystal as a function of gamma ray energy while distance from the source to the detector face was 20cm. From figure 6, it is seen that as incident gamma ray energy increases, the absolute detection efficiency of the detector crystal decreases. The reason is being, as gamma ray energy increases Compton scattering and pair production modes of interaction become dominant, so not all gamma rays add to the full-energy peak. [12]

Now, effects on absolute detection efficiency of the detector crystal by changing the distance from the source to the detector face was studied while the same gamma source kept.

Figure 7. Variation of absolute detection efficiency as a function of distance using source (Cs\textsuperscript{137}).
Figure 8. Variation of detection efficiency as a function of distance using source Co\textsuperscript{60} (1.17MeV).

Figure 9. Variation of detection efficiency as a function of distance using source Co\textsuperscript{60} (1.13MeV).

It is seen that absolute detection efficiency of the detector crystal varies when distance from the source to the detector face was changed. The results are displayed in figure 7, 8, 9. It is seen that the absolute detection efficiency of the detector crystal decreases as distance from the source to the detector face increases.

4. Conclusions
In this paper, we developed a model for gamma spectroscopy to calculate scintillation properties of Ø1"×1" of the LaBr\textsubscript{3} (Ce) detector crystal exposed to gamma rays in the energy range from 300 KeV to 1.13 MeV by using Monte Carlo transport code. In MCNP simulation, F8 tally was used as it provides the energy distribution of pulses created in the detector volume without any variance reduction. The pulse height spectra of various gamma sources were obtained, which were used to get desirable results. The distance from the detector face to the source was varied to see the effects on scintillation properties of the detector crystal. Also, the scintillation properties of the same size detector crystal were studied experimentally. The gamma radiation sources Co\textsuperscript{60} (1.17 MeV, 1.33 MeV) and Cs\textsuperscript{137} (662 KeV) were used and experiment was run for one thousand seconds.

The detector crystal offers the energy resolution of 3.45\% and the FWHM of 22.81 KeV when gamma source Cs\textsuperscript{137} (662 KeV) was used. As we increased the incident gamma ray energy, the FWHM of the detector crystal increased, while the energy resolution and absolute detection efficiency of the detector crystal decreased. The reason is that at low energy gamma rays dominant mode of interaction is photoelectric absorption, which contributes to full energy deposition with a large number of pulses. As gamma ray energy increases Compton scattering and pair production modes of
interaction become dominant, so not all gamma rays add to the full-energy photo peak. Due to which full-energy photo peak decreases and widens. It is seen that the FWHM and the energy resolution of the detector crystal is not dependent on distance since its value did not vary by changing the distance from the source to the detector face. While the absolute detection efficiency of the detector crystal is dependent on distance since its value varied by changing the distance from the source to the detector face. Crystal with low energy resolution value is considered to be more suitable for gamma spectrometry.

An Error in experimental results may occur due to counting statistics, energy linearity of PMT and interference of light. Because of high light yield of the LaBr₃(Ce) detector crystal combined with a very fast decay, the peak currents flowing through the photomultiplier tube are high. The PMT available in laboratory was not suitable for the LaBr₃(Ce) detector crystal which is a major cause of error in experimental results. The PMT and other electronic devices did not take into consideration in MCNP simulation, which was the minor cause of an error in simulation results. Also, values of GEB parameters were determined by using only two gamma sources which also produced an error in simulation results. But in our case, a good agreement was found between simulation and experimental results.

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