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RESEARCH ARTICLE

COMPARISON OF $LaBr_3$(CE) AND NaI(TL) SCINTILLATION CRYSTALS RESPONSES TO VARIOUS GAMMA ENERGIES USING MONTE CARLO SIMULATIONS.

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Abstract

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In this study, MCNP and FLUKA codes were applied for simulating and comparing of crystal response of $LaBr_3$(Ce) and NaI(Tl) scintillation detectors to gamma radiation of various radionuclides. Simulation results were compared with experimental data obtained from common real detectors containing these crystals. The results showed considerable differences between response of crystals and real detectors for low energy gamma rays which is mainly due to difference in simulated and experimental geometries. Also it is represented that $LaBr_3$(Ce) crystal is able to find energy peaks faster than NaI(Tl).

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Introduction:-

Using scintillation light produced in certain materials for the detection of ionizing radiations, is one of the oldest methods on record. The scintillation process is one of the most beneficial techniques available for the detection and spectroscopy of wide types of radiations (Knoll, 2000). Scintillation spectrometers are widely utilized in detection and spectroscopy of the high energy photons (γ-rays) (Kleinknecht,1998; Regis et al., 2010). These detectors are commonly applied in nuclear and high-energy physics researches, medical imaging, diffraction, non-destructive testing, and geological exploration (Moses,et al.,2005; Kurosawa,et al., 2010). There are two types of scintillator detectors, organic, and inorganic. The scintillation process in inorganic materials depends on the energy bands determined by the crystal lattice of the material. One of the most applicable inorganic scintillator, is crystalline sodium iodide, in which a trace of thallium is added as an activator [NaI(Tl)] (Knoll, 2000).

In the past few years, a new cerium doped halide scintillator, $LaBr_3$(Ce) has been developed which has attractive properties such as temperature stability, very high light yield, low decay constant (Van,et al., 2001; Zhu,et al., 1982), excellent energy resolution, high count rate capability (Rosson,et al.,2011; Löher,et al., 2012), high gamma-ray detection efficiency, and optimistic technology for manufacturing crystal at larger sizes (Iltis,et al., 2006).This scintillation detector has recently become commercially available. Although growing the crystals has been proven somewhat difficult, but they are now available commercially in the sizes up to 3×3 inches from Saint-Gobain (Saint-Gobain Crystals and Detectors, Nemours, France) (Milbrath,et al., 2007). The characteristics of mentioned inorganic scintillators are listed in Table 1 for comparison (Knoll, 2000;Iltis,et al., 2006;Shah,et al., 2013).

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Table 1:- Properties of two inorganic scintillators studied in this research.

Scintillator	Density (g/cm^3)	scintillation light decay constant (ns)	Light yield (Photons/MeV)	FWHM in 0.662 MeV (%)	Wavelength of max. emission (nm)
$LaBr_3$(Ce)	5.29	26	63000	~ 3-4	357
NaI(Tl)	3.67	250	39000	6-7	415

The detector efficiency is easily determined via computational approach. Monte Carlo simulations have been widely applied for detector modelling and efficiency calculations (Garcia-Talavera,et al., 2000;Van, Ngoc,et al., 2007) Although, the accuracy of such simulation techniques must be validated before combining them to the laboratory routine, several studies have been published about the response of high-resolution gamma-ray spectrometers with Monte Carlo calculations. Most of them reported in agreement with experimental results (Garcia-Talavera,et al., 2000; Van,et al., 2007;Debertin,et al., 1982; Sánchez,et al., 1982; Decombaz,et al., 1992;Overwater,et al., 1993), except for the low-energy ranges. In the real case, it is not commonly possible to know the response of the detector crystal itself (without holder and exit window of the detector). Monte Carlo simulation has capability to calculate the crystal response to different gamma energies.

In this study, MCNP and FLUKA Monte Carlo codes were applied for modelling and comparing of the $LaBr_3$(Ce) and NaI(Tl) Scintillation crystals responses to gamma radiation originated from different radionuclides. The calculated results were compared with experimental data obtained from frequent real detectors for these crystals.

Materials and Methodes:-

Monte carlo simulations:-

Monte Carlo simulations were performed using MCNP (version 4C) and FLUKA (version 2011.2-17) codes. They are general purpose tools for calculations of particles transport and interactions with materials (Briesmeister,et al., 2000; Ferrari,et al., 2005).

Monte carlo codes:-

The MCNP4C code:-

MCNP4C has capability to transport neutron, photon and electron or coupled neutron/photon/electron. This code utilized continuous-energy, nuclear and atomic data libraries. Photon interaction tables apply for all elements from $Z = 1$ to $Z = 94$. The data in these tables allow MCNP to consider coherent and incoherent scattering, photoelectric absorption with the possibility of fluorescent emission in the photon transport. The detailed physics treatment is the default and contains form factors for electron binding effects, coherent (Thomson) scatter, and fluorescence from photoelectric capture (Briesmeister,et al., 2000; Shultis,et al., 2004).

The FLUKA code:-

FLUKA can simulate the interaction and propagation in matters of about 60 various particles with high accuracy. For the photon transport, photoelectric effect with real photoelectron angular distribution according to the fully relativistic theory of Sauter was taken into account. Interactions sampled separately for each component element and for each K edge as well. The edge fine structure is considered. Parameterizations/tabulations for photoelectric cross sections containing all known edges from a few eV up to $Z = 100$. Optional emission of fluorescence photons and approximate treatment of Auger electrons for all K and most L lines are existed. Compton effect with Doppler broadening are utilizing the appropriate form of the Compton profiles, and account for the atomic bonds by means of inelastic Hartree-Fock form factors. Rayleigh scattering was considered, and the Photon polarization was taken into account for Compton, Rayleigh, and photoelectric interactions. The lowest transport limit for the photons is 1 keV. Due to the lack of Coster-Kronig effect, fluorescence emission may be neglected at the energies lower than the K-edge in high-Z materials. The minimum recommended energy range for primary photons is about 5 to 10 keV. In this code, optical photons include Cherenkov, scintillation, and transition radiation can be generated and transported. It can also transport the light of given wavelength in the materials with user defined optical properties (Ferrari,et al., 2005).

Simulation Procedure:-

Crystals geometries:-

In both codes, cylindrical geometries were used for the crystals simulations corresponding to the crystals used in experimental measurements (Milbrath,et al., 2007). $LaBr_3$(Ce) crystal was simulated 1.5 inch in height and 1.5 inch in diameter and with 5.3 g/cm^3 density, while NaI(Tl) crystal, 2.2 inches in height and 1.5 inch in diameter, and 3.67 g/cm^3 density. The crystals were aligned with y-axis and set from y=0 to y= -1.5 and y=0 to y= - 2.2 inch, respectively. Figure 1 shows NaI(Tl) crystal dimensions and source position relative to it.

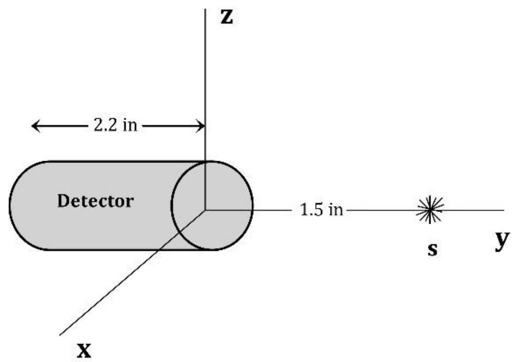

Fig 1:- NaI(Tl) detector dimensions and source position relative to it.

Source Specification:-

Definition of the sources in MCNP:-

Button sources such as experimental condition (Milbrath,et al., 2007)were specified as planar sources with uniform distribution of radioactive material upon them that emit gamma rays isotropically. These sources were assumed as a disc with 5 cm in diameter and parallel to x-y plane in (0, 0, 100) or (0 , 0, 10) origin coordinates. Sources energies, depending on radioactive materials, spam from 59.54 keV for ^{241}Am in 10 cm distance from crystals to 1332.52 keV for ^{60}Co radioisotope in 100 cm. The activities of various sources were chosen different. Except for ^{241}Am (9.8µCi) and ^{109}Cd (6.7µCi) sources (because of their low gamma rays energies), the distant between the sources and the crystals was selected 100 cm.

Definition of the sources in FLUKA:-

Because of source definition complexity (especially distributed radioactive sources on a plane or in a volume) in FLUKA, for the purpose of convenience, a beam with characteristics mentioned in Table 2 was simulated. The features of this beam are equal to the button sources used in experimental measurements and MCNP simulations. The position that the beam starts is the same as MCNP and measurement setups.

Table 2:- The characteristics of the beam equal to button sources simulated by the FLUKA code.

Momentom spread type	Flat
Momentum spread (Gev/c)	0
Beam shape in x axis	Rectangular
Beam shape in y axis	Rectangular
Beam width in x direction (cm)	2.5
Beam width in y direction (cm)	2.5
Divergence type	Isotropic

For investigating of the simulated data accuracy, the button sources accompanied by shielding were studied. The shielding was 0.5 inch steel plate which was placed between the sources and the crystals at a distance of 20 cm from the crystals (Fig 2).

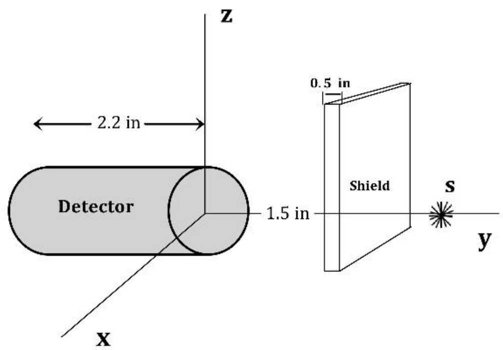

Fig 2:- The position of steel plate shield relative to simulated NaI(Tl) detector and button source.

Crystals materials specification and amount of added activators:-

$LaBr_3$(Ce) crystal was considered about 99.5% as combination of $LaBr_3$ (one lanthanum ion La^{+3} with three bromine ion Br-1) and 0.5% (concentration of cerium Ce^{+3}) as a combination of $CeBr_3$(Dorenbos,et al., 2004) . Also for NaI(Tl) crystal, concentration of thallium activator similar to the most sodium iodide commercial crystals was regarded about 0.11% as a TlI combination against 99.89% Na as a NaI form (Saha, 2006).

Tally definition:-

F8 and DETECT pulse height tallies were utilized in MCNP4C and FLUKA simulations, respectively. In MCNP4C, F8 (pulse height tally), scores the energy distribution of pulses in a cell that simulate a physical detector. Its energy bins corresponds to the total energy deposited in a detector in the defined channels by each physical particle. This tally will not work with any variance reduction other than source biasing. Another aspect of the pulse height tally that is different from other MCNP tallies is that photon, electron and photon/electron modes of transport are all equivalent. All the energy from both photons and electrons, if present, will be deposited in the cell, no matter which tally is specified. Score value is zero if no track entered the cell during the history. Zero scores are due to particles passing through the pulse height cell without depositing energy. Therefore, it is possible to calculate the recorded number of counts in any channels corresponding to various energies per gamma photon that enter the detector.

In FLUKA, DETECT tally, scores energy deposition on an event by event basis (detector), providing coincidence and anti-coincidence capabilities such as those of a trigger. An “event” means energy deposited in the detector regions by one primary particle and its descendants. A “signal” means energy deposited in the detector by the same primary particle, and descendants. The output of DETECT is a distribution of energy deposited per event in detector. In this study, coincidence capability was selected as a trigger. For physics setting, the DEFAULTS card was set to EM–CASCAde in which electromagnetic interactions are considered. Generally, in this work, for both MCNP and FLUKA, 10^6-10^8 photons histories were tracked for reaching the error of calculations below 5%. The Monte Carlo simulations did not contain the light collecting and amplifying in the photo-cathode of the detector. Internal and external background counts did not considered in the simulations. The crystals of both detectors were just modelled in this study.

Results and Discussions:-

Non-shielded button sources calculations:-

Since Monte Carlo simulation results are calculated per particle, net area/net count of photopeaks for sources with various energies and for one gamma ray entrance in the crystals, were obtained from the calculations. For calculating of the total count caused by all the photons are emitted from the sources, as an example, the calculations for ^{241}Am are shown below. With assumption of the ^{241}Am button source has 9.8 µCi activity, for 10 second counting (by means of $LaBr_3$(Ce) crystal), occurs about:

$9.8\ \mu Ci \times 3.7 \times 10^4\ dis.s^{-1}/\ \mu Ci \times 10\ s = 3.626 \times 10^6$ disintegrations. (1)

Considering about 35.9% of ^{241}Am atoms disintegrations produce gamma ray with 59.54 keV energy, the total number of the photons emitted with this energy (in 10 sec counting) would be equal to:

$3.626 \times 106 \times (35.9/100) = 1.302 \times 10^6$ photons. (2)

As such, the net count of the crystal using MCNP simulated results is as follow:

7.63×10^{-3} counts/photon $\times 1.302 \times 10^6$ photon = 9934 counts. (3)

For all the sources, similar calculations were performed. The results were shown in Tables 3, and 4.

Table 3:-Comparison between the simulated and experimental data obtained from 10 second counting of button sources (without background) using $LaBr_3$(Ce) and NaI(Tl) detectors.

Isotope	Energy (keV)	Activity (µCi)	Distance from detector (cm)	Net area of photopeak					
				$LaBr_3$:Ce			NaI(Tl)		
				MCNP	FLUKA	Exp.*	MCNP	FLUKA	Exp.*
^{241}Am	59.54	9.8	10	9934	10447	9644	9580	9864	6261
^{133}Ba	81.01	28.8	100	318	311	213	309	297	124
^{109}Cd	88.04	6.7	10	726	735	473	704	705	374
^{57}Co	122.07	37.1	100	1046	994	749	1033	976	526
^{133}Ba	276.29	28.8	100	62	52	30	---	---	---
^{133}Ba	302.71	28.8	100	142	126	43	---	---	---
^{133}Ba	355.86	28.8	100	351	373	330	347	383	318
^{133}Ba	383.70	28.8	100	50	48	16	---	---	---
^{137}Cs	661.62	9.1	100	108	110	78	92	93	96
^{60}Co	1172.23	7.1	100	48	45	36	43	39	19
^{60}Co	1332.51	7.1	100	43	36	19	39	33	16

* Milbrath, et al., 2007.

Table 4:- Comparison of experimental and simulated data for 180 and 200 second counting of shielded button sources using $LaBr_3$(Ce) and NaI(Tl) detectors respectively.

Isotope	Energy (keV)	Activity (µCi)	Distance from detector (cm)	Net area of photopeak					
				$LaBr_3$:Ce			NaI(Tl)		
				MCNP	FLUKA	Exp.*	MCNP	FLUKA	Exp.*
^{57}Co	122.07	37.1	100	1808	1647	1499	2012	1959	1622
^{133}Ba	302.71	28.8	100	927	844	620	1015	910	631
^{133}Ba	355.86	28.8	100	2399	2583	2245	2591	3081	2393
^{137}Cs	661.62	9.1	100	867	854	622	918	840	601
^{60}Co	1172.23	7.1	100	485	461	495	499	460	372
^{60}Co	1332.51	7.1	100	451	416	397	444	415	317

*Milbrath, et al., 2007.

In Table 3, simulated and experimental data obtained from 10 second button source counting (without background) using $LaBr_3$(Ce) and NaI(Tl) crystals were compared.

Simulated results calculated by means of MCNP4C and FLUKA codes represent that for $LaBr_3$(Ce) crystal, net area of photopeak for each source (corresponding to various energies) is greater than NaI(Tl). In other words, the simulated data states that $LaBr_3$(Ce) crystal can measure peaks with different energies faster than the NaI(Tl) crystal. Also, it can be said that for counting the same number of photons (with certain energy), NaI(Tl) crystal needs more counting time than $LaBr_3$(Ce) crystal. For $LaBr_3$(Ce), maximum difference between codes prediction is 19.44%. For both crystals, simulated results are close to the experimental data in 355.86 keV (maximum difference 9.11% and 20.44% for MCNP and FLUKA, respectively). For NaI(Tl), there is a good agreement between the simulation and experiment in ^{137}Cs energy (96.87% agreement). As such, in short counting time (non-shielded condition), there is not good accommodation between simulated results and experimental data in most cases due to difference in simulated geometries and the real detectors (containing holder and exit window of the detectors).

Shielded button sources calculations:-

For better comparison of the two crystals, in Table 4, the simulated data of shielded button sources was compared with the experimental measurements obtained from Ref. (Milbrath,et al., 2007).In Table 4, experimental and simulated data indicate that although NaI(Tl) crystal had counted more time (200 second) than $LaBr_3$(Ce) crystal (180 second), but due to the high intrinsic efficiency of $LaBr_3$(Ce) in relative to NaI(Tl) (Iltis,et al., 2006) , the recorded number of the photons for each of the sources using both crystals are almost equal. This also state that

$LaBr_3(Ce)$ crystal is able to find energy peaks faster than NaI(Tl). For $LaBr_3(Ce)$, unlike NaI(Tl), the simulation are close to experiment in most cases. For both crystals, maximum difference between the codes was 15.9%. For $LaBr_3(Ce)$ and NaI(Tl), maximum differences between the simulated results and experimental data were 49.52%, 37.3%, 60.86% and 44.22% for MCNP4C and FLUKA, respectively. The best agreement between the simulated and experimental data was for ^{60}Co counting by $LaBr_3(Ce)$ (more than 86.4%). Some factors which limited the accuracy of the Monte Carlo simulated efficiency are the following (Garcia-Talavera,et al., 2000):

The uncertainties in the interaction parameters, such as cross sections and ranges, the simplifying supposition applied to simulate the interaction processes and the errors in the detector and source definition due to the lack of a precise knowledge of their characteristics.

In this study, the difference between the simulated results and experimental data may cause as follows; Monte Carlo method error, difference between simulated geometry and the real case which applied by Ref. (Milbrath,et al., 2007), not considering of the entrance window of the detectors, not considering of the crystal holder of the detectors, not considering the self-absorption of the sources.

Conclution:-

In this research, MCNP and FLUKA codes were applied for simulating and comparing of the $LaBr_3(Ce)$ and NaI(Tl) scintillation crystals response to gamma radiation originated from different radionuclides. The simulated results were compared with the experimental data from the literature. There was a considerable difference between simulated results and experimental data in most energy (especially in low energies) related to the radionuclides. As such, in all cases, there is not good accommodation between simulated results and experimental data in most cases due to difference in simulated geometries and the real detectors (containing holder and exit window of the detectors). From the results of this study, It can be concluded that $LaBr_3(Ce)$ crystal is capable of finding energy peaks faster than NaI(Tl). As such, it would be a good selection for the portable radioisotope identification devices.

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