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Abstract

Researchers at the University of Missouri-Columbia have designed a triple crystal phoswich detector that allows for simultaneous detection of alpha, beta, and gamma radiation. A ZnS:Ag layer detects alpha particles, a CaF₂:Eu scintillator preferentially interacts with beta particles, and a NaI:Tl cell is used for gamma detection. The detector output is digitally collected, processed, and analyzed by a personal computer using custom software. MCNP simulations of this detector found that the phoswich design has inherent minimum energy limits of 250 keV E_{\max} for beta particles and 50 keV for gamma-rays. For a 2.54 cm thick NaI:Tl crystal, intrinsic gamma efficiency for photons ranges from a maximum of 80% at 100 keV to 26% for 2 MeV photons. Mischaracterized gamma events in the CaF₂:Eu crystal above 175 keV can be corrected by subtracting 26 +/- 4% of the total number of counts in the NaI:Tl crystal from the CaF₂:Eu response. Beta induced events in the NaI:Tl crystal primarily result from Bremsstrahlung interactions, and can be estimated by multiplying the CaF₂:Eu energy spectrum by a fourth order polynomial.

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1. Introduction

A preliminary design for a triple crystal phosphor sandwich (phoswich) detector was developed and constructed through prior research at the University of Missouri-Columbia. This detector can measure alpha, beta, and gamma radiation simultaneously and can be beneficial whenever all three radiations need to be detected in a single system [1]. The research reported here uses Monte Carlo N-Particle (MCNP) version 4C software analysis to determine optimum operating parameters for this detector and to set a basis for determining the optimum design of future detectors based on the original design.

Phoswich detectors are based on the use of different scintillators interacting preferentially with different types of radiation. Multiple scintillators are placed on top of each other and viewed with a single PMT to create a detector that is able to measure multiple types of radiation with high efficiencies [2]. Each scintillator used has different light output characteristics [3, 4]. A post-processing unit interprets the pulse information and assigns its origination by discriminating between characteristic light emission spectra from the three crystals. It is common for phoswich units to be created specifically for one characteristic mixed radiation field as they are mainly used for research purposes.

The first layer with which radiation interacts in an $\alpha/\beta/\gamma$ phoswich detector consists of a thin phosphor that is highly sensitive to alpha radiation and much less sensitive to beta or gamma radiation. Only radiations with much longer ranges are able to penetrate to the second layer, which is optimized for general charged particle interactions. The third layer is much thicker than the first two layers and is designed to interact with the remaining gamma rays. This design allows preferential, but not exclusive, interaction of various radiations with specific layers. Without correction, it is assumed that events in the first layer originate from

alpha particles, second layer events are due to beta radiation, and third layer pulses result from gamma rays.

Figure 1 shows a simplified schematic of the current phoswich detector design. The ZnS:Ag layer is 10 mg/cm² thick (0.002445 cm), the CaF₂:Eu layer is 0.254 cm thick and the NaI:Tl scintillator is 2.54 cm thick. The diameter of all crystals is 5.08 cm. These dimensions were not precisely planned; instead they are estimated thicknesses that stop common high energy alpha and beta particles, while attenuating a fair amount of gamma rays. A more systematic method was needed to optimize the design and operation of the detector.

Monte Carlo radiation simulation computer codes produce interaction data by generating random interactions in a given environment. Source particles are individually generated and undergo a series of reactions probabilistically determined by combining a user-generated input file containing environmental specifications and experimental interaction data. These source particles and their interactions are tallied until a user-defined condition, either length of simulation or a specific error value, is met. MCNP is a Monte Carlo simulation program developed by Los Alamos National Laboratory that is capable of simulating electrons, photons, and neutrons. MCNP has a simple geometry system (surfaces comprised of spheres, cylinders, planes, cones, ellipsoids and transformations) that can be used to quickly build models.

The use of Monte Carlo computer simulations to estimate interactions in detectors has been shown to yield errors less than 5% when compared to collected data [5-7]. As both the physical interactions of radiation and Monte Carlo simulations are based on random occurrences, the accuracy of computer simulations is only limited by the accuracy of experimental data and proper geometry input. In this report, both the type of interactions and

the system geometry are simple and well defined. Thus, it was decided that Monte Carlo simulations should be used to quickly provide accurate interaction data for several hundred detector configurations in order to study its behavior.

There were three main objectives of this research. Self-correction parameters were developed as a function of energy so that raw data from the three scintillators could be accurately separated into numbers of total events resulting from alpha, beta, and gamma radiation. Predictions of system intrinsic efficiency were made for beta and gamma radiations. Finally, the thicknesses of the scintillators were optimized for uses in various mixed radiation fields.

2. MCNP Methods

Simple MCNP 4C input files were constructed that modeled the three scintillation layers housed by an aluminum casing and surrounded by air. Unless otherwise noted, crystal thicknesses were 10 mg/cm^2 for ZnS:Ag, 0.254 cm for CaF₂:Eu, and 2.54 cm for NaI:Tl. A 5.08 cm diameter disc source 0.5 cm underneath the detector was used in the analysis. The doping materials were not modeled in the MCNP environment, as their concentrations are low and can vary from different manufacturers. Sample runs were completed that included typical levels of doping materials, but these showed a difference of only 0.2% compared to simulations that did not include dopants.

While MCNP has several types of tallies to record the various interactions of radiation in system components, this report utilizes the F8, or pulse height, tally. This tally simulates gamma spectrometers for photons and neutrons by returning probability distributions of energy deposition (or energy spectra) for a given set of energy bins. Separate F8 tallies were applied to each scintillator to model energy pulses that would be created in detectors by

radiation interactions [8]. Mode p, or transport of only photons, was used for all analysis other than cross-talk characteristics and electron sources. In this mode, MCNP uses a full transport model for photon interactions but assumes that secondary electrons deposit all of their energy at the point of interaction. Mode p proved to be an average of 250 times faster and nearly as accurate as Mode p e, which performs full transport calculations for both photons and their secondary electrons. Mode p e was used with electron source particles. Simulations typically modeled 10 million source photons or 500,000 source electrons, which yielded relative errors in significant energy bins of 1% or less.

3. Results

Since alpha particles are not simulated by MCNP, do not create many secondary particles, and follow a relatively straight path with a definite range, they are not analyzed in this report. The ZnS:Ag crystal is assumed to have a 100% intrinsic efficiency for alpha detection. Also, ZnS:Ag will only produce scintillation photons for alpha particles and electrons less than 6 keV [4], which would become low energy noise that is filtered out by the software. Thus, there will be no mischaracterized events in ZnS:Ag. The term “electron” will be used to indicate data for monoenergetic electron emissions, and “beta particle” will be used when referring to beta particle efficiency data. It was only necessary for MCNP to simulate electrons at various energies, and mathematical manipulation of the product of detector efficiency as a function of energy with typical beta emission spectra allowed beta particle efficiencies to be generated.

MCNP results determined that the ZnS:Ag layer attenuated a significant amount of electron energy. Figure 2 shows the ZnS:Ag energy spectrum for electrons with a variety of

energies. The most probable electron energy loss in ZnS:Ag is around 20 keV due to the short pathlength that electrons can traverse in ZnS:Ag. Similar calculations were performed for gamma events in the ZnS:Ag scintillator, but MCNP showed that the ZnS:Ag attenuation of gamma rays is mainly restricted to gamma particles below 50 keV. This energy is considered below the likely energy range of the detector, so the ZnS:Ag layer can be considered inconsequential for photon blockage.

Movement of secondary electrons from one scintillator to other locations was analyzed by comparing the differences generated by Mode p and Mode p e. Figure 3 compares Mode p and Mode p e results for 1 MeV photons in CaF₂:Eu. The data series “Mode p e – Mode p” is the difference of these two simulations. As expected, the electrons that were created with higher energies were transported out of the CaF₂:Eu, while a few low energy electrons entered the CaF₂:Eu crystal from the aluminum housing and the other two scintillators. The total number of events was 3.5% higher using Mode p e than the Mode p estimation. When concerned about determining mischaracterized event parameters for the CaF₂:Eu crystal, the statistic of utmost importance is the ratio of CaF₂:Eu events to NaI:Tl events. This parameter did not change appreciably, and thus it was determined that use of Mode p was sufficient for photon analyses.

Figure 4 shows energy deposition spectra for electrons interacting in CaF₂:Eu. The spread of the deposited energies is due to ZnS:Ag attenuation and electrons passing through CaF₂:Eu and depositing the remainder of their energy elsewhere. Figure 5 shows an intrinsic efficiency curve for electrons in CaF₂:Eu, while Figure 6 is a generated intrinsic efficiency curve for beta particles using Figure 5’s data combined with beta emission spectra. Figure 5 shows that ZnS:Ag absorbs nearly all electrons up to 70 keV, although its average attenuation

for electrons that pass through it is only 25 keV. This discrepancy is likely due to the increased specific ionization at the end of an electron's range and the minute size of the ZnS:Ag crystal. It can be seen that the ZnS:Ag layer greatly reduces beta efficiency, as the curve would be nearly 100% throughout the range if the CaF₂:Eu layer were bare. This reduction can be mainly attributed to the absorption of low energy electrons by ZnS:Ag. The efficiency reduction becomes much greater when measuring sources that exhibit self-shielding, as this further drops the energy of electrons that arrive at the detector. Since low energy electrons lose much of their energy in the ZnS:Ag layer, the CaF₂:Eu crystal may have events that are near the energy noise level of the detector system that would be removed by the detector's collection and processing software. Thus, the results in Figure 6 are also given assuming a low energy bias of 0.1 MeV and 0.15 MeV to account for this effect.

Gamma interactions are significant in both the CaF₂:Eu and NaI:Tl crystals. Figure 7 shows intrinsic efficiencies as a function of source photon energy. It demonstrates that NaI:Tl is most efficient for photons around 100 keV, but Figure 8 shows that the ratios of CaF₂:Eu to NaI:Tl events at this energy are very high. At energies above 175 keV, the ratio of CaF₂:Eu events to NaI:Tl events begins to stabilize between 20% and 25%. The efficiency of NaI:Tl is reduced by a factor of 2 between 0.1 MeV events and 0.8 MeV events. As expected, the ZnS:Ag events and energy deposition are negligible for photons above 50 keV. However, ZnS:Ag absorbs almost all gammas below 30 keV, CaF₂:Eu absorbs most gammas between 30 and 70 keV, and NaI:Tl begins to become efficient only at energies above 70 keV.

Correcting for gamma events in CaF₂:Eu can be done by subtracting a fraction of the NaI:Tl events from CaF₂:Eu events. Figure 8 shows that at gamma energies beyond 175 keV, CaF₂:Eu will interact with 22% +/- 2% of NaI:Tl events above 175 keV. The fraction of

NaI:Tl events occurring past 175 keV to the total number of NaI:Tl events was found to be 0.83 +/- 0.10. Combining these two corrections, the self-correction for gamma events in CaF₂:Eu is calculated by subtracting 26% +/- 4% of NaI:Tl's total events above 0.175 MeV. This method is only valid for the total number of event corrections. If accurate energy spectra from each crystal are desired, more complex (and less accurate) corrections must be established.

Most NaI:Tl events that occur from electron sources result from Bremsstrahlung radiation. The ratio of NaI:Tl electron events to CaF₂:Eu electron events as a function of energy was fit to the following polynomial with high accuracy:

$$\text{NaI:Tl}_{\text{ratio}} = 0.055 E^4 - 0.17 E^3 + 0.19 E^2 - 0.059 E + 0.008 \quad (\text{Eqn. 1})$$

where E is energy in MeV and NaI:Tl_{ratio} can be multiplied by the CaF₂:Eu electron counts in an energy channel to determine the total number of NaI:Tl events resulting from electron interactions. As with the CaF₂:Eu corrections, these parameters will only correct the summed NaI:Tl events, and not individual counts in energy bins.

Implementation of these two corrections becomes more difficult in mixed β/γ fields, as both spectra will need to be corrected simultaneously. The method for correction will change based on the relative abundances of electrons and photons, and multiple corrections must be built into the software for various NaI:Tl to CaF₂:Eu count ratios. Electrons with energies less than 1.5 MeV will not produce NaI:Tl events above 175 keV, and higher energy electrons will only produce a small amount of events. This allows the total number of CaF₂:Eu events to be corrected fairly accurately before altering the NaI:Tl counts, although it is not possible to accurately adjust the CaF₂:Eu spectrum as a function of energy. It is necessary to estimate the new shape of the CaF₂:Eu spectrum to use in Equation 1 to correct NaI:Tl events. It was

found that nearly all $\text{CaF}_2:\text{Eu}$ interactions with photons above 70 keV were Compton events, which would lead to a fairly even energy distribution of mischaracterized events. This estimation and the determination of the number of electrons over 1.5 MeV will be the most challenging aspects of correcting events in the scintillators.

An alternative way to remove low energy gamma data from the $\text{CaF}_2:\text{Eu}$ events is removal during pulse collection. Since the PMT obtains all data from a single photon's interactions simultaneously, this can result in a Compton or photoelectric NaI:Tl pulse superimposed on a Compton pulse from $\text{CaF}_2:\text{Eu}$. The resulting combined pulse rise time could be recognized by the pulse shape software as abnormal and the $\text{CaF}_2:\text{Eu}$ pulse shape would then be subtracted from the signal before being tallied as a $\text{CaF}_2:\text{Eu}$ event. The ability to assess and manipulate pulses on an individual level is a benefit of the digital collection system, but further software development is necessary to add these more complex features. It would be advantageous to apply this individual pulse rejection technique to low energy $\text{CaF}_2:\text{Eu}$ events so that data would not have to be discarded, and efficiency reduced, to improve accuracy. Due to the complexity of the corrections and the low energy resolution of the actual detectors, it will be necessary to experimentally determine the best solution for event corrections.

The recommended ZnS:Ag thickness is its lower manufacturing limit, 10 mg/cm^2 . This will stop any alpha particle with an energy of 6.2 MeV or less, and most other alpha particles up to 9 MeV in energy due to non-perpendicular entry angles. Though the electron efficiency remains unchanged for a thinner $\text{CaF}_2:\text{Eu}$ scintillator, total energy deposition will decrease. Figure 9 shows this change in deposition as the thickness of the $\text{CaF}_2:\text{Eu}$ crystal is decreased from 0.254 cm to 0.2 cm and 0.15 cm. If there is a possibility of detecting

significant activity of electrons over 1.2 MeV, CaF₂:Eu thicknesses of less than 0.254 cm should not be used without programming the detector's software to compensate for the extra energy deposited in NaI:Tl.

There are positive aspects to decreasing the CaF₂:Eu layer. Figure 10 demonstrates the decrease in gamma events in CaF₂:Eu as its thickness is decreased. This effect is almost linear as reducing thickness from 0.254 cm to 0.15 cm, or 59% of 0.254 cm, reduces the number of events to 63% of the 0.254 cm data for both 0.4 and 0.8 MeV photons. Ideally, the CaF₂:Eu layer should be only thick enough to stop the highest energy electron, as extra thickness only adds to its photon interactions. Due to requirements for full electron energy deposition in the CaF₂:Eu crystal, it is recommended that a CaF₂:Eu thickness of 0.2 cm be used if electrons with maximum energies of less than 1.5 MeV are anticipated and a thickness of 0.254 cm otherwise. Smaller thicknesses could be used in conjunction with a quartz window to stop the remainder of the electrons, but this would not eliminate the Bremsstrahlung radiation detected by NaI:Tl.

Changing the thickness of the NaI:Tl layer only alters the phoswich detector's gamma efficiency and does not have an effect on the amount of mischaracterized events. Figure 11 shows photon efficiencies for several NaI:Tl thicknesses. The extra efficiency gained by increasing the thickness deteriorates due to the added portion being farther from the source and the exponential nature of attenuation. NaI:Tl's thickness should be designed to meet photon efficiency requirements for the system in which it is to be used; thus no global recommendations can be made.

4. Conclusions

This research has established that the use of a triple crystal phoswich detector trades the advantage of simultaneous detection of alpha, beta, and gamma radiation for a loss in the detection of low energy beta and gamma particles. An MCNP-based model has been developed to determine these trade-offs in efficiency, energy threshold, and cross-talk probabilities for different scintillator thicknesses and source energies. Evaluating the use of a triple crystal phoswich detector instead of three separate detectors must be done with considerations to the detector's potential uses. Further research is required to determine what method of mischaracterized event correction will yield the best results in real-world applications. This research has provided a basis for further study in phoswich design, as phoswich detectors with an annular configuration of scintillators are already being investigated at the University of Missouri-Columbia using the same techniques. In general, it has been determined that phoswich detectors of the design evaluated here will be effective and reasonably accurate in the detection of a mixture of mid to high energy particles.

Acknowledgements

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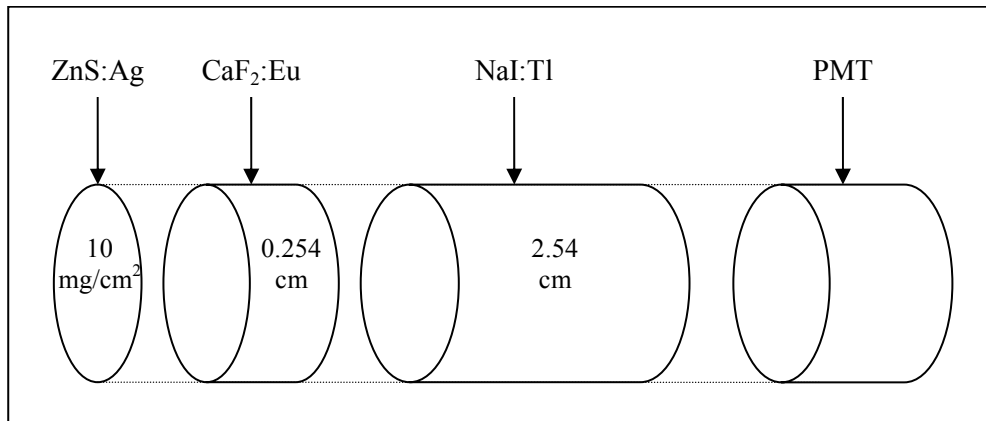


Figure 1. Schematic of detector.

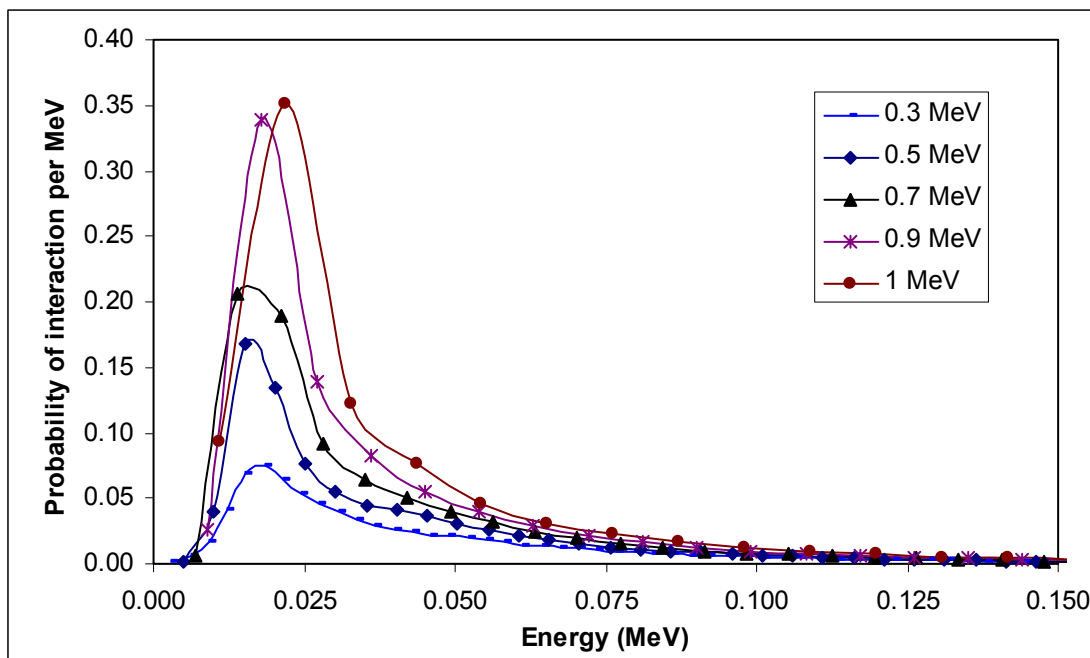


Figure 2. ZnS energy spectra for various electron energies.