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A phoswich detector for simultaneous alpha–gamma spectroscopy

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ABSTRACT

Phoswich detectors are of value for radiation spectroscopy, especially in cases where a low-cost solution for a mixed radiation field is desired. Meanwhile, simultaneous spectroscopy of alpha particles and gamma-rays has many applications in quantification and distinguishing the alpha-emitting radionuclides which usually occur in the analysis of environmental solid samples. Here, we have developed a system for detection of radioactive actinides (e.g., ^{241}Am) based on the alpha–gamma coincidence technique. The underlying concept, is to assemble two appropriately selected scintillators (i.e., a fast and a slow one) together with a discriminating unit for analysis of their data. Detailed Monte Carlo simulation procedure has been developed using the GEANT4 toolkit to design and find enough knowledge about the response of the system in the studied radiation field. Various comparisons were made between experimental and simulation data which showed appropriate agreement between them. The calibration was performed and the MDA was estimated as 60 mBq for the phoswich system.

1. Introduction

Analysis of most solid environmental samples demands precise spectroscopy of their emitted radiation which in many cases is indeed the alpha particles as well as gamma-rays. Simultaneous spectroscopy of these radiations directly enhances the identification and distinguishing possibly unknown components, especially in high-background conditions. Routinely, Liquid Scintillation Counting (LSC) detectors are useful tools for this task, although with certain limitations and drawbacks which complicates their usage and moreover increases their costs. For example, the liquid sample preparation and solving into the LSC is not only time-consuming, but also is costly and irreversible. Since the phoswich detectors can be made of solid scintillators, so it can suppress drawbacks of the liquid detectors and can be considered as an alternative for these purposes.

There are great numbers of works on the development of phoswich detectors for spectroscopy of radionuclides regarding their mixed emitted radiation. Most of these studies have considered the coincidence spectroscopy of beta–gamma-rays [1–5]. However, to best of our knowledge, a few reports are available about the development of phoswich detectors for alpha–gamma-rays coincidence spectroscopy. Usuda et al. have presented a phoswich detector for simultaneous counting of alpha, beta, gamma, and thermal neutrons by assembling a phoswich detector consisting of ZnS, Li-Glass, and NE102A scintillators [6–10]. White and Miller have investigated on a triple-layer phoswich detector for alpha, beta, and gamma-rays spectroscopy by means of ZnS, CaF_2(Eu), and NaI (Tl) scintillators [11]. Later, a simulation study about these phoswich systems has been presented in [12] which reports on beta and gamma-rays only.

This paper is devoted to development of a phoswich detector for alpha–gamma coincidence analysis of radionuclides which is able to overcome the limitations of LSCs. This system consists of a fast BC400 plastic and a CsI(Tl) scintillators; the first one is supposed to result in the fast scintillation components and the latter one would provide the slow scintillation components. To this aim, a detailed simulation procedure has been implemented in the GEANT4 framework including the radiation and particle interaction physics, light generation, and transport process and also the light detection in the photomultiplier tube (PMT). As an experimental verification of the design, several comparisons have been made between simulations and measurements via irradiating the detector by an ^{241}Am alpha–gamma source. Generally, the results are in an acceptable agreement. Furthermore, measurements related to energy–time and energy–energy correlations indicate a strong dependency between the two signals coming from BC400 and CsI(Tl) scintillators which allows one to identify the irradiation source.

The paper is arranged as follows. In the next section, the detector and its design will be described which elaborates the design procedure and method of implementation into the GEANT4 toolkit. Also in this section, the experiments will be detailed. Section 3 is

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devoted to the presentation of results. Finally, in Section 4 some concluding remarks will be presented.

2. Materials and methods

2.1. Detector design implementation

Phoswich detectors are based on optically coupled scintillators with a apparently different decay times from BC400 plastic scintillator and the CsI(Tl) was chosen as the two scintillators because of their quite different decay times (2.4 ns versus 0.68 μs) [13]. Fig. 1 shows the response time of the scintillators based on GEANT4 database, which are used in the simulation. According to Fig. 1, due to the different time behavior of signals resulting from the mentioned scintillators, pulses arising from only one decay are easily distinguished from those obtained with both decay components, using the pulse shape analysis method. Besides its appropriate decay time, CsI(Tl) has much less sensitivity to humidity which makes it appropriate for "unsealed" application, the case for our studies.

GEANT4 is an object-oriented toolkit which is well-suited for radiation detector design purposes, at both of the high-energy and low-energy ranges [14,15]. It allows accurate modeling of radiation sources and detector devices, with great accuracy in the simulation of physical processes. Monte Carlo simulation of the selected scintillators of our phoswich design was performed by GEANT4 toolkit (version 4.9.5). To do so, we have implemented the full set of physical processes contributing in the final response of the system, as follows:

- Radiation (i.e., gamma-rays and alpha particles) transport and energy deposition into the scintillator elements.
- Light generation in the scintillation process considering its time-dependency based on the principal scintillation decay time.
- Scintillation light transport in the optical components.

Physics processes settings must be included in the Physics List class that tells GEANT4 kernel identify what physics processes are registered to each particle. Physics processes are classes that specify how particles decay or interact with materials at different energies [16]. Notably, the following physics processes and settings have been adopted throughout the simulations:

- G4PhotoElectricEffect, G4ComptonScattering, G4GammaConversion, G4EMStandardardPhysics related to the gamma-rays transport physics which controls the transport and interactions of gamma-rays.
- G4eMultipleScattering, G4eIonisation, G4eBremsstrahlung related to the electron transport physics which controls the electrons interactions.
- G4IonPhysics related to the alpha-particles physics.
- G4OpticalPhysics, G4OpticalProcessIndex related to the scintillation and optical physics.

There were also many other classes in our simulations which mostly due to the scintillation process and optical boundary effect. Importantly, the UNIFIED model for treating the boundary conditions was selected [17]. It applies to dielectric-dielectric interfaces and tries to provide a realistic simulation, which deals with all aspects of surface finish and reflector coating. As the scintillators did not have external reflectors, the ground surface boundary condition (in the UNIFIED model nomenclature) has been set in the simulations. Based on the GEANT4 options, we set surface finish to "groundair". An optical properties database has been included in GEANT4 which contains essential data for various scintillation materials. Notably, it contains the decay time, refraction index, and optical attenuation lengths of BC400 and CsI(Tl) as a function of wavelength.

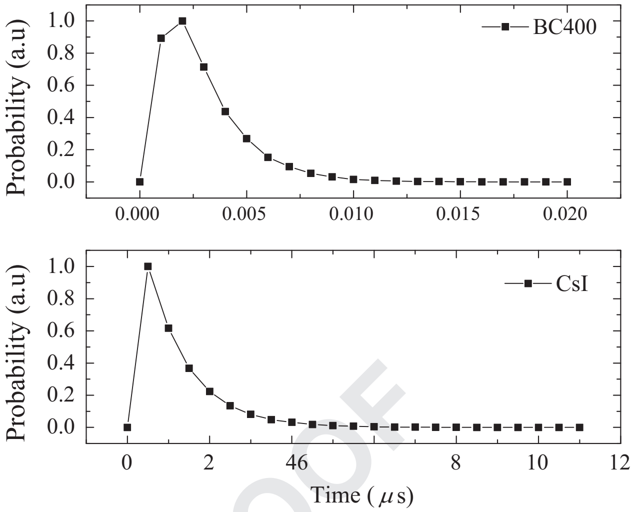

Fig. 1. Simulated time response of the fast (BC400) and slow (CsI) components of the phoswich detector.

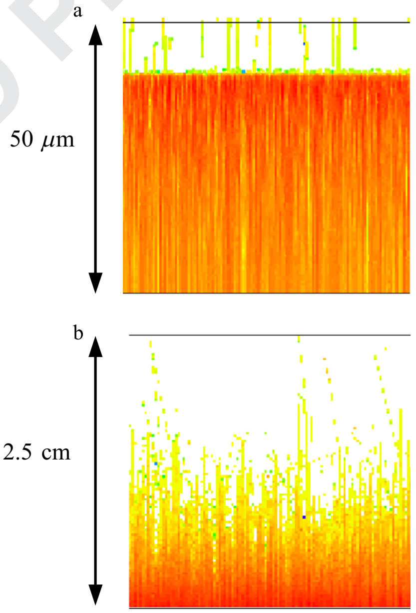

Fig. 2. Range distribution of energy deposition by (a) alpha particles inside the BC400, and (b) gamma-rays inside the CsI(Tl).

The alpha-emitting nuclides of interest (especially ^{241}Am) have usually alpha-particles with energy of about 4–6 MeV and gamma-rays of below 100 keV. Through a design based on fully stopping the alpha-particles in the BC400 (as the first layer of phoswich), the simulations indicate that it should have a thickness of at least 45 μm. Fig. 2(a) shows range distribution of the (relative) deposited energy for 5485.56 keV alpha-particles in the BC400 scintillator which demonstrates that with a thickness of 50 μm we will be assured that the entire energy of particles would be deposited. Similarly, Fig. 2(b) shows the same results for CsI(Tl) impinged by 100 keV gamma-rays which again ensures that the value of 2.5 cm is acceptable for this component. As shown, collimated alpha and gamma sources (with dimensions of 3 cm × 3 cm) were placed on the bottom surface of the BC-400 and CsI(Tl) layers, respectively. A 50 μm thickness of plastic scintillator would not result in a considerable attenuation of gamma-rays, before reaching the CsI(Tl).

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