




Geant4 investigation of the alpha-beta-gamma detector system used in medical imaging, environmental and nuclear site monitoring

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Abstract

No commercially available detector system can measure alpha, beta and gamma-rays at the same time and separately with good efficiency, while being cost-effective, portable and offering real-time monitoring. The main purpose of an alpha-beta-gamma detector would be for safety management and nuclear decommissioning in the nuclear industry. This idea for a detector system became more valuable, after Fukushima in Japan, because nuclear waste can contain fission products and transactinide materials which not only emit gamma-rays but also emit alpha and beta particles and in some cases, neutrons. In this research, we investigated the best available alpha-beta-gamma radiation detector materials and their optimum thickness by using Geant4 based GATE simulation. The work revealed a better efficiency result for each radiation type than in previous work. In the simulation, 0.05 mm ZnS(Ag), 3.2 mm plastic scintillator and 1.75 mm BGO were found to be best for the detection and identification of alpha, beta and gamma-rays respectively. In nuclear medicine, this type of detector system could also modify to become a miniaturized radio-guided surgery beta-gamma probe beside of the modification into the robotic surgery. This research result will influence three different areas in imaging technology, homeland security and nuclear industry.

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1. Introduction

Scintillator-based detector systems are widely used in nuclear, particle and medical physics for radiation detection applications such as spectroscopy, creating cross-sectional images and for quality control of medical radioisotopes [1]. Nowadays research labs, nuclear medicine, and nuclear power plants use standard detector systems such as Geiger-Mueller counter, HPGe, NaI(Tl) scintillation, etc. No commercially available detector system can measure alpha, beta and gamma-rays at the same time separately with good efficiency and performance. For some scintillator-based detector applications, two or more scintillators are optically coupled together to form a “phoswich” a name derived from “phos(phor) (sand)wich”. Phoswich detectors have been shown to function effectively in measuring different classes of radiation separately or simultaneously and can discriminate from high ambient background radiation. The scintillators comprising a phoswich need to have different pulse shape characteristics such as rise time and decay times. In particular, decay-time analysis of

signals from a phoswich detector gives a great opportunity to distinguish incident radiation, which

scan be a mixture of charged particles (alpha, beta) and neutrons or gamma rays. It also allows for the separation and identification of which events occurred in which scintillator such as the PARIS calorimeter [1]. In nuclear power plants, nuclear waste can contain fission products and transactinide materials which not only emit gamma-rays but also emit alpha and beta particles and in some cases, neutrons. Moreover, the environmental monitoring of radionuclides following accidents in nuclear power plants not only needs to detect gamma-rays but also necessary to detect alpha and beta particles [2]. That became more important after Fukushima in Japan. A three-layer phoswich alpha-beta-gamma imaging detector was developed by Yamamoto and Ishibashi for simultaneously monitoring alpha-beta-gamma rays following a nuclear accident or for applications in molecular imaging [2]. These considerations show that it is important to simultaneously and separately monitor alpha-beta-

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gamma radiation via the one detector system. The main aim of this study is the investigation of the measurement alpha-beta-gamma radiation with a single novel detection system by using Monte Carlo simulation. We have completed the work with the Geant4 based GATE simulation to determine which materials and what thicknesses are the most suitable depending on the radiation type. Our result is comparable and better than that obtained with a similar detector system in Japan [2].

Understanding the behaviour of actinides in the nuclear fuel cycle process is important for process and safety management in nuclear power stations [3]. Boiling water reactors, pressurized water reactors and heavy-water reactors are highly contaminated by the fission products [4]. Therefore, the radiation level of the cleaning and cooling systems and reactors have to be monitored regularly and carefully. It is also necessary to carry out continuous monitoring of the levels of radioactivity in the environment around nuclear power plants to ensure compliance with the basic safety standards. There are two methods to monitor the territory; global (outside the zone) and close monitoring to watch and check on the radiological impact on the environment. The radiological monitoring for artificial and natural radioactivity is conducted using two methods; automatic measuring networks and taking a periodic sample to analyse. Automatic measuring networks can play an important role as a warning before the accident and inform the county's citizens the event of the nuclear accident. There are several major radionuclides present in the liquid released from a nuclear power plant to the environment. Their primary decay modes are only beta-emitting (Tritium, Nickel-63, and Strontium-90) and beta-gamma emission related to some radionuclide such as Cobalt-60, Iodine-129, Cesium-134, and Cesium-137, while some of them are alpha-emitting such as Plutonium-239. Tritium contaminated water in a nuclear power plant is regularly diluted, resulting, normally the radiation level in groundwater must be less than drinking water tritium level [5]. Monitoring tritium level in groundwater helps to detect the leaks of underground piping as reported the more tritium level in one of the wells around the Vermont Yankee nuclear power station, where buried piping leaks were found [5]. Similar problems were reported in the detection of radioactive tritium in the groundwater of the more than 48 commercial US nuclear power stations [5]. Obviously, one of the safety requirements at a nuclear power plant is the close monitoring of the tritium level in underground water. Similar to tritium, the other important disposable radioisotope produced during nuclear fission is the long-lived beta-emitting

Strontium-90, resulting from accidents at nuclear power plants and leaking from nuclear waste storage or nuclear weapon testing. If it is ingested into the body, the risk of damage to bone marrow, leukaemia and other bone cancer increases [6]. Therefore, it is also greatly important to monitor its activity in the environment, particularly, in groundwater around the nuclear facilities [6]. Beta-emitting radioisotopes counting system is currently a long and arduous process. Samples must be collected from groundwater wells, transported to a laboratory, processed with hazardous chemicals before the activity can be measured. This procedure also presents logistical and financial challenges for the nuclear industry. After the disaster at the Fukushima nuclear power plant is highlighted the need for Sr-90 detection be able to move quickly and easily to repeat the procedure many times. The existing methods, such as liquid-liquid extraction and chromatography, for beta monitoring, is mainly the separation of the target radionuclide from the sample to remove interface other sources of radiation because of overlapping of their energies. Traditional methods produce a large volume of secondary waste and use very hazardous concentration. Each year, thousands of samples need to be prepared in the nuclear-decommissioning industry, and it needs a new approach to reduce secondary waste production, which is more rapid and safe as well as cost-effective. After the radionuclide is isolated, the beta-emitting radioisotope from the sample can be measured using counting devices (gas ionization chamber and liquid scintillation counters). The other important beta counting device is the Triple to Double Coincidence Ratio (TDCR) which includes the liquid scintillation detector with three PMTs uniformly around a sample. However, these detector systems are large, immobile and not suitable as an in-situ detection system. Gallium-arsenide (GaAs) photodiode for detection of Strontium is a novel detection method and being a potential for direct, in situ beta detection for nuclear decommissioning application [6]. However, this new sensor technology using solid-state detectors, may not be applicable to detect low energy of the beta particles emitted by tritium [5]. These solid-state detectors have several disadvantages like being expensive and radiation damage results in noise in the detector and can negatively affect the counting statistics [6]. Current techniques are lab-based, time-consuming, and produce secondary waste. The existing novel detectors are expensive, poor for the detection of a low energy beta, and negatively affected by the radiation. Therefore, it is necessary to develop a new detector with potential for in situ, cost-effective, real-time monitoring in groundwater and more mobile detector which have not been well developed [5]. In this

research, an alpha-beta-gamma detector system will be investigated using Monte Carlo simulation to fill the needs of the nuclear industry.

Besides, simultaneous detection of three different radiations by one detector is also crucial for some medical cases such as RGS (radio-guided surgery), medical and molecular imaging [2, 7-8]. The effective operation has to remove as many tumoral tissues as possible while preserves the surrounding healthy tissues. Imaging technologies allow to localize the surgeon in the tumour and give information about its size, shape, and stages [9]. Nowadays, pre-operative imaging tools and intra-operative imagine techniques are getting higher importance because they provide real-time information of the picture of tumour boundaries [9]. Aside from these techniques, radio-guided surgery is a promising field for accurate, sensitive tumour detection, and the most available system based on gamma radiation detection. Beta detection provides higher sensitivity and signal-to-noise ratio for shallow tumours, but beta particles inside the tissues have a short range. It requires development of extremely compact devices that can be directly introduced inside the surgical cavity in contact with the surveyed tissues. In the last decade, semiconductor light sensors such as silicon photomultipliers (SiPMs) have gained a favourable position relative to photomultiplier tubes (PMTs). They offer significant advantages in being compact, low voltage, low power and immune to magnetic fields. New generation photosensors, SiPMs, emerge as the most promising for the development of intra-operative beta probes. The performance of SiPMs is also comparable to standard photomultiplier tubes (PMTs). According to the research called the development and evaluation of an intra-operative beta imaging probe studied by Sara Spadola, the performance of the probe designed with two scintillator detection and gamma subtraction method is better than the single detection system [9]. Besides, the depth of interaction method [10], which is recently the most significant improvement in the new generation of high-resolution PET detectors, may be applied to this kind of detection as a PET module to obtain better spatial resolution. In nuclear medicine, an alpha-beta-gamma detector has the potential to become a miniaturized beta-gamma imaging probe and modified to use in robotic surgery. A study conducted using a radio-guided surgery beta probe combined with a robot by an Italian research group, and they concluded that the robotic surgery performance and time consumption is better than man-hand operation [11]. There is also the opportunity to use the small size of the detector system inside a drone, that will be advantageous after

the nuclear accident. This simulation research can also modify into the development and evaluation of intra-operative miniaturized imaging probes. That will be based on two or three layers' scintillators coupled to compact SiPMs as a real-time alpha-beta-gamma monitoring system in the nuclear industry.

2. Materials and Methods

In this study, we aim to investigate the detector which can carry out alpha-beta-gamma monitoring separately and simultaneously by Monte Carlo simulation. Our scintillation material should have better performance and be much more portable than what has previously made. It will apply to many different applications from environment monitoring to medical imaging. We have carried out an initial simulation of such a device using the Geant4 based GATE simulation program. To benchmark the simulation, we reproduced the performance of the Yamamoto et al. detector using the same detector shapes and dimensions illuminated by the same radioactive sources as shown the detector geometry in Figure 1.

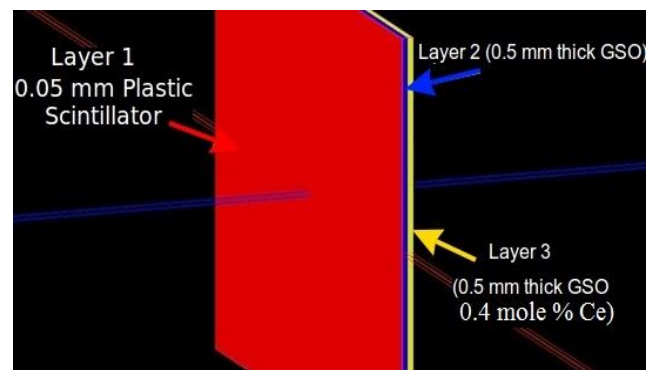


Figure 1. Three different layers described in the simulation; the red layer illustrates to 0.05 mm thick plastic scintillator for detection of the alpha particles. The second layer shows in blue color for beta particles detection and 1.5 mol % Ce doped GSO. The last layer is 0.4 mole % Ce doped GSO as shown in yellow color.

We then explored the effect of increasing each layer's efficiency depending on radiation type, through changing the thickness of layers and component material. Geant4 visualization of the essential detector and three different radioactive sources shown in Figure 2.

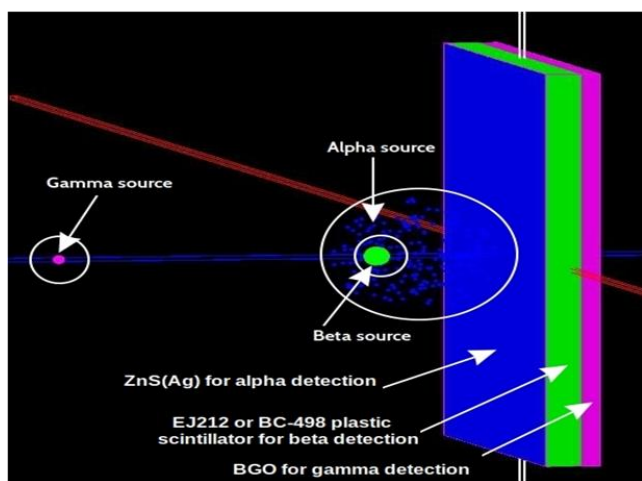


Figure 2. The first layer (blue) defined as Layer 1 is a scintillation material with $50 \times 50 \times 0.05 \text{ mm}^3$ dimension. The blue points represent the alpha source (Am-241) with 15 mm diameter and two kBq activity located in front of the detector. The second layer (green) was Layer 2 scintillation material with $50 \times 50 \times 3.2 \text{ mm}^3$ dimension. Green spherical point showed for beta particle source (Sr, Y-90) with 2 mm diameter and 100 Bq activity positioned 1 cm from the detector surface. The last layer (magenta) determined Layer 3 inorganic scintillation with 1.75 mm thickness. Magenta points also represented the gamma source (Cs-137) with 1 mm diameter and 370 kBq activity located at 4 cm away from the detector surface.

The optimum thickness of the first layer material found to be 0.05 mm and that thickness is enough to absorb almost all alpha particles. The Layer 1 material is ZnS(Ag) powder with medium decay time (200 ns). The second layer, EJ212 or BC-498 plastic scintillator, is 3.2 mm thick with the fast decay time (2.4 ns). The last layer to detect gamma-rays could be various inorganic scintillation materials: LYSO, CsI, LaBr₃, CeBr₃, SrI, GAGG, NaI(Tl), BGO and GSO etc. Some of them are highly hygroscopic materials. Since one of the possible applications for the detector will be to use it in direct contact with water or other liquids, we prefer to use non-hygroscopic materials. Several inorganic scintillations have internal beta or gamma

radiations coming from Lutetium (Lu) such as LYSO, results in the measurement would be deteriorated by the intrinsic activity. Therefore, BGO was found to be the most suitable inorganic scintillator with better efficiency results in the simulation as a Layer 3 material.

If the first layer thickness is increased in the simulation, beta particle absorption went up inside layer 1. Similar behaviour was also observed for the second and third layers. Increasing the second layer thickness seriously affects the gamma-rays count inside the layer 2. In addition, layer 3 is also affected by annihilation photons (511 keV) occurring after positrons annihilate with electrons. The detection of the gamma-ray inside the third layer could be spoiled by the annihilation photons. For this reason, the simulation work completed with annihilation photons to optimize the thickness of each layer. Depending on radiation type, we found out the best three materials and optimum thickness of the layers by using Monte Carlo simulation. In the application, these three different layers have different signal shape characters (different decay time) that will allow applying the pulse shape analysis, resulting in the separation and identification of which events occurred in which layers. Alpha, beta and gamma-rays have different range inside the medium, so these range differences help to show the first layer detects alpha, the second layer detects beta and the last layer detects gamma-rays.

3. Results and Discussion

Geant4 simulation was completed in two parts. The first part reproduced the earlier detector design of S. Yamamoto et al. In the second part, the scintillation material and the thickness were changed step by step to find out the best option and optimum thickness depending on the radiation type. The simulation results listed in Table 1 as a percentage of the detected counts for each layer.

Table1. Comparison between GATE simulation and the work of S. Yamamoto et al. In the simulation, the result obtained after investigating the best materials and thickness for each layer.

Percentage of relative detected counts for alpha particles			
	$\epsilon_{\text{exp.}}(\%)$ (Yamamoto S. et al.)	$\epsilon_{\text{GATE}}(\%)$ (present work)	$\epsilon_{\text{GATE}}(\%)^{\dagger}$ (present work with different material)
First Layer	93.5	93.44	99.87
Secand layer	6.1	5.79	0.13
Third layer	0.4	0.77	0
Percentage of relative detected counts for beta particles			
First Layer	0.4	0.32	1.85
Secand layer	87.7	89.24	90.02
Third layer	11.9	10.44	8.13
Percentage of relative detected counts for gamma-rays			
First Layer	0.6	0.95	1.65
Secand layer	60.0	54.71	19.74
Third layer	39.4	50.33	78.61

[†] Different materials with optimum thickness

The thickness of the first layer material (ZnS(Ag)) was 0.05 mm; that thickness was enough to absorb 99.87% of the alpha particles. The second layer, EJ212 or BC-498 plastic scintillator, was 3.2 mm thick and found to have 90.02% efficiency for beta particles. To detect gamma-rays, a 0.75 mm thick BGO scintillation material was found to be the best option with its 78.61% gamma-rays efficiency value. The detected counts for gamma-rays in the second layer calculated at 19.74 %. In terms of the gamma-ray interaction with the medium, there is no way to remove these counts inside the second layer because of the higher Compton scattering probability of the high energy gamma rays. However, it is highly possible to apply a correction for the gamma detection efficiency of the second and third layers [12]. According to the simulation result, detected counts for alpha, beta particles and gamma rays noticeably increase with these scintillation materials and their optimum thickness found out in the simulation. The experimental result consists of the result obtained by Geant4 simulation in terms of particle detection. Percentage of the relative detected counts for gamma-rays obtained better efficiency in the simulation. That could be related to the experimental conditions. In the literature, there are many determined technical properties for 0.5 mole % Ce doped GSO, but not much more technical information for 0.4 mole % Ce doped GSO. The other difference between

experimental and calculated value in gamma-rays might be about the lack of the material definition for layer3 inside the simulation.

4. Conclusions

No commercially available detector system can measure alpha, beta and gamma-rays at the same time separately with better efficiency and performance. The main aim of this study is the investigation of the measurement alpha-beta-gamma radiation with a single novel detection system by using Monte Carlo simulation. We have completed the work with the Geant4 based GATE simulation to determine which materials and what thicknesses are the most suitable depending on the radiation type. Our result is comparable and better than that obtained with a similar detector system in Japan [2]. In the application, each layer will be optically coupled, then the other side of the last layer will couple with the photosensor (PMTs or SiPMs). For medical imaging, position-sensitive SiPMs or PMTs must be used to define the position of the events. Proper collimators could be employed to obtain a meaningful image from the three layers detector system [13]. The operating principle of this detector will be simple, easily portable and easy to use comprising a combination of novel materials with a state-of-the-art electronic system. This kind of device

can be used for medical or molecular imaging during surgery or other applications in medicine as a beta-gamma probe. In addition, this detector could be modified for different application fields from environmental measurement to monitoring at nuclear power stations. Aside from the detection of the three different radiation types with one detector, the newly designed detector system can also be in direct contact with liquid or water to monitor the radiation inside or

around of the nuclear power plant. We have already succeeded to measure directly beta particles emitted by Ga-68 and F-18, used as a PET radiotracer in Hull PET research centre [14]. This type of detection system is highly suitable to be applied to nuclear-decommissioning applications such as in situ alpha-beta-gamma detection, which will be mobile, and becoming for real-time monitoring in groundwater as shown a possible detector design in Figure 3.

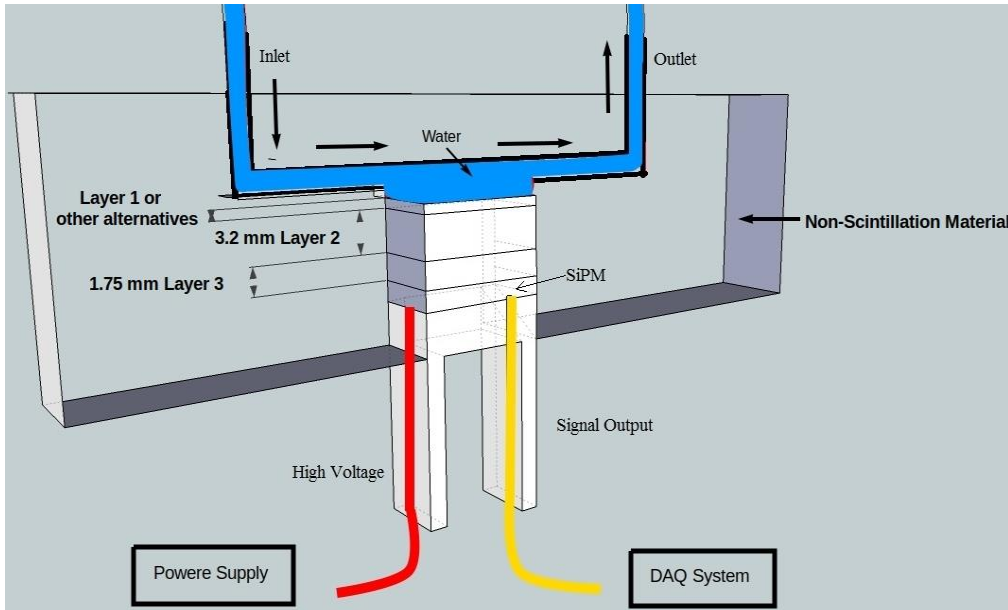


Figure 3. Detector design for monitoring radiation level in water or liquid, which is slightly similar to plastic scintillator-based microfluidic devices developed for measuring positron-emitting radionuclide and successfully patented by the research group of the University of York and University of Hull [1,14]. The first Layer material ZnS(Ag) is a thin powder, and it is commercially available within plastic scintillator. Therefore, it could not damage by liquid or groundwater.

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Conflict of Interest

No conflict of interest was declared by the author.

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