Heterogeneous scintillator geometries to maximise energy deposition for waterborne beta particle detection

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ABSTRACT

Here the geometries that maximise detection efficiency of heterogeneous scintillators used to detect beta particles in aqueous solutions by maximising energy deposition are described. The determination of the geometry was achieved with the Monte Carlo code Geant4 using CaF_2:Eu scintillator as a pertinent case study, and validated with experimental data using single crystal CaF_2:Eu and heterogeneous CaF_2:Eu scintillators. Both 2D and 3D structures composed of arrays of primitive unit cells of packed spheres were examined to find the optimal geometry to maximise detection of volumetric sources of tritium and aqueous Carbon 14 and Lead 210. The 2D structures were evaluated relative to a single crystal scintillator and results show the detection efficiency of the 2D structures is maximised when the sphere radius is c.a. 0.46x the maximum track length of the beta particle in the scintillator. Data for the 3D structures show that the efficiency is maximised when the sphere radius is minimised, but it is further shown that practical issues limit the minimum radius that can be used for transient radiological contamination monitoring.

1. Introduction

Heterogeneous scintillators are multiphasic scintillators (Birks, 2013) that may consist of a porous structure whereby the scintillator is in the form of small particles instead of a single large crystal. The advantages of moving from scintillators in single crystal form to those made of an arrangement of small volumes have been examined before. The reason for the enhanced performance of nanoparticle scintillators was explored by Dujardin (Dujardin et al., 2010), who suggested that the crystal structure was altered by the particles small size which led to, for a number of reasons, a reduction in non-radiative recombinations, thereby increasing luminescence. However, heterogeneous scintillators have the additional advantage of having a larger surface area than single crystal scintillators which helps mitigate attenuation effects of radiation monitoring in aqueous systems. For this reason, a number of heterogeneous scintillator flow cells have been devised (DeVol and Fjeld, 1995; Kawano et al., 2011; Shirahashi et al., 1984). In particular, Kawano et al. has recently published a number of papers based around a heterogeneous scintillator for tritium detection (Kawano et al., 2014, 2015). (see Table 5)

A few studies have been conducted on the effects of the geometries of heterogeneous scintillators used for waterborne beta particle monitoring. For instance, Monte-Carlo modelling of heterogeneous scintillators was undertaken for 3D packed spherical geometries (Tan and

DeVol, 2003). In that work, hexagonal close packed (HCP), face centred cubic packed (FCC), body centred cubic packed (BCC) and square packed (SP) spherical structures were analysed using the PENELOPE2000 Monte-Carlo code. While the simulations only considered initially mono-energetic low energy beta particles, they did show that porosity, material, sphere size and beta particle energy significantly affect detection efficiency. In particular, they showed that for low energies, smaller scintillator particle sizes increased the energy deposition within the scintillator and hence detection efficiency. Similarly, experimental results by Kawano et al. show that when using 25 μm, 50 μm and 150 μm radius 3D packed spheres, the smaller radius spheres results in more energy deposited in the scintillator as compared to in the water (Kawano et al., 2011).

An area currently unexplored is the use of 2D heterogeneous scintillators. It will be shown here that there are practical advantages to using 2D structures for transient environmental monitoring of radiological contamination of aqueous systems. This paper also uses Monte-Carlo combined with flow cell analysis to examine both 2D and 3D packed spherical structures with a view of maximising the energy deposition into the scintillator. The majority of this article will focus on tritium because of its detection challenges (Alton et al., 2017); however, both aqueous Carbon 14 and Lead 210 will also be analysed as they are common tracers in the environmental sciences, and the results show the conclusions derived are quite general.

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Europium activated Calcium Fluoride (CaF$_2$:Eu) is used as the scintillator material throughout this work. This is because CaF$_2$:Eu is an ideal scintillator for flow cell based detectors due to its non-hygroscopic nature, its refractive index similar to that of Silicon Photomultipliers (SiPM), an emission curve that aligns with the peak efficiency of SiPMs (Materials, 2017; Sensl, 2017) and a similar output brightness (30,000 photons per MeV) when compared with the commonly used NaI:Tl (Gobain, 2017; Materials, 2017). The inorganic scintillator can also be formed into a heterogeneous scintillator as particulate CaF$_2$:Eu with varying sphericity has been fabricated through various chemical approaches (Ritter et al., 2014). It is therefore particularly suitable to be used to detect tritium, commonly found in the form of tritiated water in the environment, whereby it replaces the ^{1}H in H$_2$O with ^{3}H and decays with a low energy beta particle (< 18.6 keV) with a half-life of 12.3 years. The current standard for detecting tritiated water is liquid scintillation counting due to its high efficiency (Elmer, 2017), while single crystal designs suffer from very low efficiency (Alton et al., 2017). However, liquid scintillation is inconvenient for transient monitoring of radiological contamination. Heterogeneous flow cell scintillator detectors have been developed as a compromise between liquid scintillation and single crystal scintillation. This work shows that the efficiency of these heterogeneous detectors can be maximised with appropriate scintillator geometry choice.

2. Method

2.1. Geant4

The simulations for this article were carried out using the Geant4 10.3 (Incerti et al., 2010; J Allison et al., 2006) Monte Carlo software. Geant4 is a library of tools for the simulation of nuclear and particle physics and is freely available, its applications range from nuclear reactor physics and high energy particle physics to radiotherapy and shielding. Some advantages of the Geant4 software is the flexibility of the code allowing the user to tailor each simulation to the application, improving the efficiency of the simulation and only analyzing the relevant parts. The Geant4 physics packages used in the following simulations included the standard electromagnetic (EM) processes, with the eIonization and eBremsstrahlung functions, electron ionisation and bremsstrahlung respectively. These two used the Livermore models for improved accuracy at low energies.

2D heterogeneous scintillators were modelled as planar arrays of face-centred-cubic (FCC) and square packed (SP) spheres (see Fig. 1). As is experimentally relevant, the arrays of spheres were modelled as being supported beneath by a thick layer of polydimethylsiloxane (PDMS) and exposed to a thick layer of radiologically contaminated water on top. The water acted as a source of tritium, aqueous carbon 14 and lead 210 in turn, modelled as cubic volumes of water with homogeneously distributed isotropic beta sources. The energies of the beta particles were described through a 1000 bin histogram, with 10^8 beta particles and using radiological data for each isotope from the Radiological Toolbox (Center for Radiation Protection Knowledge, 2015). In order to create accurate models in the simulations any beta particles generated in the scintillator material in the simulations were immediately killed as the initial position of the beta particle should be within the water. In addition generated photons were not tracked, as photon detection is outside the scope of this work and is assumed ideal. These geometries were compared to an equivalent sized single crystal CaF$_2$:Eu scintillator. The thicknesses of the PDMS and water layers were set to exceed the maximum beta particle track length and can be assumed to be effectively infinite in depth; the values are shown in Table 1. The actual simulations were formed from arrays of the unit cells described in Fig. 1 tiled together. For the square packed array, the lateral dimensions of the unit cell were 2x the scintillator particle radius, i.e. 2R. The lateral dimensions of the face centred cubic unit cell were 4Rsin(45°). Convergence studies showed that arrays of 1000 × 1000 tiled unit cells
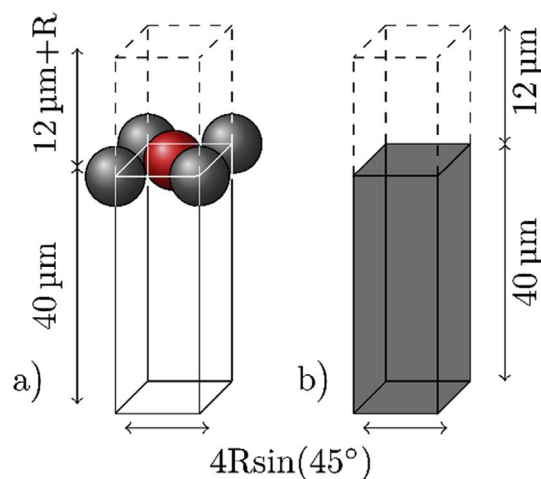

Fig. 1. Diagram of the 2D structures for the Geant4 simulation where red and grey colours indicate the scintillator material. Only the Face Centred Cubic arrangement is shown, the dashed volume denotes the water and source volume of radionuclides. The dimensions shown are for tritium, see Table 2 for all the radionuclides dimensions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1
Thickness of the various layers for the 2D unit cells for the Geant4 simulations. Where R is the radius. See Fig.1 for the schematic of these geometries.

Isotope	Single Crystal Water (Dashed Volume) Height	Single Crystal Scintillator Height	Microparticle Water (Dashed Volume) Height	Microparticle Substrate Height
Tritium	12 μm	40 μm	12 μm + R	40 μm
Carbon 14	0.6 mm	1.2 mm	0.6 mm + R	1.2 mm
Lead 210	150 μm	300 μm	150 μm + R	300 μm

Table 2
Table showing the Maximum Track Lengths and Maximum Geometric Track Lengths for tritium, carbon 14 & lead 210. Here the Maximum Track Length is the maximum distance the beta particle travelled calculated by summing the distances between each scattering event and the Maximum Geometric Track Length is the maximum straight-line distance travelled from point of origin to final particle location. These results are from Geant4 simulations.

Isotope	Maximum Track Length (μm)	Maximum Geometric Track Length (μm)
Tritium	7.541	4.088
Carbon 14	340.004	201.724
Lead 210	63.544	35.553

were equivalent to infinite 2D surfaces, i.e. edge effects were negligible. These structures are named *2D* as the position of each sphere can be described in $\hat{x}$ & $\hat{y}$ coordinates. A development of this utilises a dual 2D layer design whereby two 2D spherical packed planes with an optimal 3.5 μm radius SP arrangement are placed facing each other to act as a channel. Here, the water source is placed between the two layers and simulations were ran to determine the efficiency of the flow channel as a function of width, i.e. the gap between the two layers. The intent is that this structure can be layered to create a quasi-3D structure with practical advantages over a homogeneous 3D packed spherical structure.

3D heterogeneous scintillators were modelled as face centred cubic, body centred cubic and square packed arrays of spheres (see Fig. 2). As before, these spheres were comprised of CaF$_2$:Eu scintillating material and interspersed with radiologically contaminated water as described