Development of a Compton Suppressed Gamma Spectrometer using Monte Carlo Techniques

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Abstract

Gamma ray spectroscopy is routinely used to measure $\gamma$ radiation in a number of situations. These include security applications, nuclear forensics studies, characterisation of radioactive sources, and environmental monitoring. For routine studies of environmental materials, the amount of radioactivity present is often very low, requiring $\gamma$ spectroscopy systems which have to monitor the source for up to 7 days to achieve the required sensitivity. Recent developments in detector technology and data processing techniques have opened up the possibility of developing a highly efficient Compton Suppressed system, that was previously the preserve of large experimental collaborations. The accessibility of Monte-Carlo toolkits such as GEANT4 also provide the opportunity to optimise these systems using computer simulations, greatly reducing the need for expensive (and inefficient) testing in the laboratory. This thesis details the development of such a Compton Suppressed, planar HPGe detector system. Using the GEANT4 toolkit in combination with the experimental facilities at AWE, Aldermaston (which include HPGe detection systems, scintillator based detector systems, advanced shielding materials and $\gamma-\gamma$ coincidence systems), simulations were built and validated to reproduce the detector response seen in the ‘real–life’ systems. This resulted in several improvements to the current system; for the shielding materials used, terrestrial and cosmic radiation were minimised, while reducing the X–ray fluorescence seen in the primary HPGe detector by an order of magnitude. With respect to the HPGe detector itself, an optimum thickness was identified for low energy ($<300$ keV) $\gamma$ radiation, which maximised the efficiency for the energy range of interest while minimising the interaction probability for higher energy radionuclides (which are the primary cause of the Compton
continuum that obscures lower energy decays). A combination of secondary detectors were then optimised to design a Compton Suppression system for the primary detector, which could improve the performance of the current Compton Suppression system by an order of magnitude. This equates to a reduction of the continuum by up to a factor of 240 for a nuclide such as $^{60}$Co, which is crucial for the detection of low-energy, low-activity emitters typically swamped by such a continuum. Finally, thoroughly optimised acquisition and analysis software has also been written to process data created by future high sensitivity $\gamma$ coincidence systems. This includes modules for the creation of histograms, coincidence matrices, and an ASCII to binary converter (for historical data) that has resulted in an analysis speed increase of up to $\sim 20000$ times when compared to the software originally used for the extraction of coincidence information. Modules for low-energy time-walk correction and the removal of accidental coincidences are also included, which represent a capability that was not previously available.
I remember writing the acknowledgements for my Masters thesis, and how difficult it was to include everyone who helped me both professionally and personally that year. Soon after it was bound I was told by a good friend that the resulting prose had quickly deteriorated into a ‘gushing’ rhetoric of emotion. Given the vastly increased scope of this project, I will here attempt to express my thanks with some degree of concision and restraint, as to account for everyone would require a small book in itself.

Firstly I would like to thank my supervisors. They did not all contribute equally, and were not always available for advice/assistance. Quite often they were away in some foreign land, or simply busy with their own work (of which my project is a small but hopefully significant part). This is not, however, a criticism. These aspects are the direct result of working with highly respected researchers, and when I genuinely needed their help all were incredibly generous with their time. The opportunity and encouragement to work independently, and think/wonder about/understand problems (before turning to them for ‘the answer’) greatly improved my abilities as a researcher, and I am very grateful for all of their efforts. This also extends to certain life lessons; Jon – thank you for teaching me how to sleep on a bar stool, and how to deal with overly large plants/vegetables in the office. Your eternal optimism and faith in people will stay with me for a long time. Ash – my gratitude for showing us that after a hike up a mountain (still a debatable definition) it is utterly appropriate to drink a stalls worth of mulled wine, and then ‘improve’ Jon’s view of the river below by opening the doors on a glass bottomed cable lift. I’m pretty sure the swaying helped too. Paddy – thank you for your hospitality in the airport lounges, and for ‘giving me something
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mornings), drinks, meals, stories, trips, and experiences, thank you.

P.S. Simon – we still hold the record. I fear it may be unassailable – partly
due to the unrelenting march of inflation, but mostly because no other taxi
driver is that crazy.
“Success is not final, failure is not fatal, it’s the courage to continue that counts.”

Winston Churchill
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Chapter 1

Introduction & Background

1.1 Introduction

Radiation detection has been a field of active research since the discovery of radioactivity by H. Becquerel in 1896\cite{1}. Since this time, when X-rays were detected on photographic plates, many other types of radiation have been identified, with efforts to detect these ranging from a simple handheld Geiger counter measuring ionizing radiation, to the massive neutrino detection facilities, which can cover up to a cubic kilometre of ice\cite{2}.

Studying the radiation emitted from unstable isotopes allows a plethora of information to be obtained, such as the half–life of the isotope, the branching ratios of specific decays, nuclear structure information, and if from an unidentified mixture of material, the abundances of any radionuclides present. The ability to identify and quantify radioactive material in a sample is of vital importance for environmental monitoring, both for commercial and security purposes. Depending on the isotope, different forms of radioactive emission may be measured, however the method that has the widest range of application is gamma (\(\gamma\)) spectroscopy, due to the uncharged and penetrating nature of the emitted photons. The research detailed in this thesis focuses on the detection of \(\gamma\) radiation, and specifically, on dramatically improving the sensitivity of current detection systems to low energy (<300 keV) \(\gamma\) emitters.


1.2 Gamma (γ) Spectroscopy

γ spectroscopy has become an invaluable tool for the non-destructive identification of radioactive nuclides. Typically, High Purity Germanium (HPGe) detectors (figure 1.1) are employed across a wide range of energies, allowing both the identification of radionuclides and an estimate of their abundances [3]. There are many applications for γ spectroscopy, including food testing [4], neutron activation analysis [5, 6], fundamental physics research [7, 8, 9, 10] and environmental analysis [11, 12]. It is important to maximise the efficiency of such systems, as this subsequently increases the sensitivity of the detectors, allowing either smaller amounts of material to be reliably identified, or count times to be decreased, increasing measurement throughput.

![Figure 1.1: An example HPGe detector in a copper and tin lined lead cave. The lead attenuates terrestrial radiation, the tin liner attenuates fluorescence X–rays from lead, and the copper liner attenuates fluorescence X–rays from the tin. The HPGe detector has a carbon fibre window to minimise the attenuation of low energy γ radiation, and the detector signal is read out using electrodes attached to the crystal. The electronics/crystal package is contained in an aluminium canister, which also encloses the cold finger (a copper heat sink that maintains the cryogenic temperatures required for the detector to operate), and the copper crystal holder.](image)

The sensitivity of γ spectroscopy is limited by many practical factors, including the size, efficiency and resolution of the detector crystals. The Compton continuum (which arises due to the incomplete energy deposition of a scattered photon) can also obscure spectral detail, and is a particularly acute problem for the analysis of transuranic (TRU) nuclides [13] as the low energy
1.2. Gamma (γ) Spectroscopy

Photons that originate from these isotopes are swamped by the continuum from higher energy decays. Temporally coincident (cascade) γ radiation may be summed out as these are seen as a single decay, and background radiation will also obscure finer spectral detail. These factors all add noise and uncertainty to an energy spectrum (see figure 1.2), and must therefore be minimised to achieve the highest sensitivities.

Figure 1.2: An example energy spectrum from a HPGe detector. Radionuclides are identified using the energy of the peaks, which are characteristic of different γ decays. By combining the number of counts in each peak with information about the radionuclide and the efficiency of the detector system, the abundances of each radioisotope can be estimated.

Important sources of background include cosmic events, the materials used to construct the detector/laboratory and radon gas. These can be minimised with a cosmic veto and appropriate shielding, however Compton scattered events originate in the detector itself, requiring a more subtle approach. There are many methods to (partially) eliminate or recover these events, known as Compton Suppression systems, and each requires a detailed understanding of the detector system and physical processes involved. Such a detailed understanding may be achieved via experimentation with multiple detector systems, however this would prove extremely expensive and inefficient. An alternative is to use computer based Monte Carlo simulations, which allow the user to evaluate a variety of detector sizes, materials and setups before actually testing a physical detector system.
1.3 Project Scope

The majority of the experimental work presented in this thesis was performed at the Atomic Weapons Establishment (AWE), Aldermaston, and the University of Surrey. The research project aimed to determine the optimum design for a novel, Compton suppressed $\gamma$ spectrometer that substantially increases the sensitivity of existing detector systems to low energy $\gamma$ radiation.

This involved combining background reduction techniques\cite{14,15}, suppression of Compton Scattered radiation, and maximising the sensitivity of the detector system for the energy range typically seen in environmental samples (30 keV - 3 MeV). Within this range, the identification of many transuranic radionuclides (which are the focus of this study) are obscured by the large background and Compton continuum present in the spectrum, which typically arises due to other NORM (Naturally Occurring Radioactive Materials) sources present in the sample. The identification of such nuclides is further complicated by the small emission probability of the $\gamma$ decays present ($\sim 1-2$ in 10000). These aims were achieved by the following:

- Identifying an appropriate data acquisition system for multiple $\gamma$ spectrometers, and developing acquisition & analysis software to allow the processing of complex coincidence data
- Understanding and correcting for complex effects that plague coincidence systems, including cascade summing and the low–energy time–walk of coincident events
- Developing expertise in the GEANT4 Monte-Carlo toolkit, allowing the creation of detector system simulations which could be utilised to optimise the geometry and materials of the proposed system
- Validating GEANT4 detector models against existing detector systems, including NaI(Tl) scintillation detectors, HPGe detectors, and Compton Suppression systems
- Validating GEANT4 detector models against laboratory sources, including NIST (National Institute of Standards and Technology) traceable
1.3. Project Scope

complex $\gamma$ sources and environmental samples

- Optimising and improving the shielding of current detector systems to further reduce the terrestrial, cosmic, and source induced background

- Using GEANT4 models to optimise the HPGe primary crystal in a Compton Suppression system for the required nuclides

- Extending GEANT4 detector system models to identify areas where current systems can be improved, and evaluating new Compton Suppression detector designs that utilise the optimised primary HPGe crystal

This thesis is structured as follows; Chapter 2 details the relevant theory for radiation detection and measurement, including radioactive materials & the typical radiation seen when analysing environmental samples, interaction processes salient to $\gamma$ spectroscopy, and the processes involved when detecting radiation both with scintillator and semi-conductor type materials. Chapter 3 presents an overview of Compton Suppression systems (which also includes the literature review into the subject), and Chapter 4 details the Monte-Carlo toolkit GEANT4, along with the associated simulations. Chapter 5 discusses experimental details, including an overview of a typical experimental setup and a description of some of the required characterisations for each system (such as energy calibrations and efficiency calculations). Chapter 6 outlines the acquisition and processing of the detector data, for which custom routines were developed using c++ and ROOT. Chapters 7, 8 & 9 present the results from all stages of the project, separated into the logical sections in which the work was performed. Finally, the results are summarised and concluding remarks made in Chapter 10. A full list of published works and presentations pertaining to this project are included in Appendices A & B.
Chapter 2

Theory

2.1 Relevant Modes of Radioactive Decay

Radioactive decay is a statistical process, whereby unstable nuclei emit radiation that changes the state of the nucleus. Unstable nuclei are typically defined as those that have an excess of energy, and therefore decay to a lower energy state via a variety of energy-loss mechanisms. Each isotope may decay using one or many of these modes, and each mode of decay for every nucleus has a characteristic decay half-life ($t_{1/2}$). These half-lives depend on the amount of excess energy, the mode of disintegration and the underlying structure of the nucleus. The energy loss mechanisms relevant to this thesis are summarised below.

2.1.1 Alpha ($\alpha$) Decay

$\alpha$ decay most often occurs in heavy nuclei ($Z>82$)\cite{16,17}, and involves the emission of a Helium nucleus ($^4\text{He}$, consisting of 2 protons and 2 neutrons). This is because the $^4\text{He}$ nucleus is a tightly bound system, allowing the maximum release of kinetic energy for the mass of its constituent nucleons (particle emissions other than $\alpha$ decay have negative Q-values, and are therefore unable to spontaneously decay\cite{18}). Both proton and neutron numbers must be conserved in this process, and so $\alpha$ decay changes the isotope of the parent nucleus.
2.1. Relevant Modes of Radioactive Decay

\[ A^4X_N \rightarrow A^{-4}X'_{N-2} + {}^4\text{He}_2 \]

2.1.2 Beta (β) Decay

β decay is similar to α decay in that the parent nucleus changes isotope, however this occurs by converting a proton into a neutron or vice-versa. There are three sub-species of β decay, and all involve an electron or positron to achieve the conversion. Each decay also emits a neutrino or anti-neutrino, giving β decays a characteristic endpoint energy, and a large continuum of decay energies as the kinetic energy is split between the electron and neutrino.

\[ n \rightarrow p + e^- + \bar{\nu} \quad \text{β}^- \text{ decay} \]
\[ p \rightarrow n + e^+ + \nu \quad \text{β}^+ \text{ decay} \]
\[ p + e^- \rightarrow n + \nu \quad \text{electron capture} \]

2.1.3 Gamma (γ) Decay

Nuclei in an excited state can decay to the ground state through the emission of one or more γ-rays. Each γ decay is mono-energetic, and consists of a photon with the energy (ΔE) of the difference between the parent \( E_i \) and daughter \( E_f \) states (minus an often negligible correction for the recoil energy of the emitting nucleus).

The angular momentum of the emitted photon (L) will be constrained by the initial and final angular momentum of the nucleus (\( I_i \) & \( I_f \) respectively)

\[ | I_i - I_f | \leq L \leq I_i + I_f \] (2.1)

while the parity (π) change of the transition is given by the following selection rules (for Electric (E) and Magnetic (M) transitions):

\[ \Delta \pi(EL) = (-1)^L \] (2.2)
\[ \Delta \pi(ML) = (-1)^{L+1} \] (2.3)
2.1. Relevant Modes of Radioactive Decay

Photons have an intrinsic spin of $1\hbar$, and therefore transitions between two $0^+$ or $0^-$ states (where $\Delta L = 0$) are forbidden. These transitions instead occur via internal pair formation or internal conversion\cite{19}.

$\gamma$ decay typically occurs after $\alpha$ or $\beta$ decay in unstable nuclei, where the initial decay leaves the daughter nucleus in an excited state. Subsequent $\gamma$ emission allows the nucleus to reach the ground state, via a single emission or multiple decays. These emissions are often prompt ($<10^{-10}\text{s}$) in comparison to the half-lives of the parent nuclei, and so are observed as a cascade of radiation.

2.1.3.1 Isomers

Half-lives for $\gamma$ decays are generally short, however excited, metastable states can exist with half-lives substantially longer than that of a normal decay. These are known as isomers, and are generally accepted to be $\gamma$ decays with a $t_{1/2} > 10^{-9} \text{seconds}$, although there is no strict limit on this. There are four main types of isomer; Seniority, Shape (or fission), Spin and K Isomers. Each is characterised by a nuclear effect that inhibits the decay to the ground state, elongating the half-life of the isomer.

2.1.3.2 Internal Conversion

Internal conversion is a process whereby the nucleus deexcites by transferring its energy to an atomic electron, which is then ejected. This competes with $\gamma$ emission, and is notably different to $\beta$ emission as no change of proton or neutron number occurs, and the atom becomes ionised in the process. The subsequent relaxation or rearrangement of atomic electrons causes the emission of characteristic X-rays or Auger electrons\cite{19}.

2.1.3.3 Fluorescence X–rays

This is a similar process to internal conversion, however the impinging radiation originates from an external source. The radiation (which may be X–rays, $\alpha$ particles, $\beta$ particles, photons, etc.) excites or ejects an atomic electron, creating a vacancy in one of the electron shells (note that the impinging radiation
may also excite the nucleus, which will subsequently decay via the processes described above). Depending on the shell in which the vacancy is created (K, L, M, etc.), and the isotope that is affected (the composition of the nucleus defines the spacing of the electron shells), the subsequent relaxation of atomic electrons or capture of an electron into the vacancy will cause the emission of an X–ray of characteristic energy. The energy of the X–ray increases with the electron shell binding energy and the Z–number of the nucleus. Typically, only K–shell X–rays are seen during environmental studies as their energy is greatest; for example Radium (Z = 88) emits a K–shell X–ray of <104 keV, while the maximum L–shell X–ray is <20 keV\[20\]). These cause a particular problem in the low energy region where fluorescence X–rays may obscure peaks of interest.

\subsection{Annihilation Radiation}

Annihilation radiation is seen as a result of two processes in environmental samples. If a parent nucleus undergoes $\beta^+$ decay (such as $^{22}\text{Na}$), the emitted positron may only travel a few mm (dependent on the material) before losing its kinetic energy and encountering an electron. At this point the $\beta^+ - \beta^-$ pair will annihilate, producing two 511 keV photons emitted at 180° to each other (to conserve momentum). There is also a finite probability (increasing with energy) that the interaction of a photon of energy $>1.022$ MeV in a material will produce an electron–positron pair (see section 2.4.3), again resulting in annihilation radiation.

\subsection{Nuclear Decay Rates}

Unstable isotopes will decay with a characteristic decay constant, $\lambda$. From an initial population $N_0$, the number of the radioactive isotopes present ($N$) after time $t$ can be described by the following relation:

$$N = N_0 e^{-\lambda t} \quad (2.4)$$

When extended to a system where the daughter is also radioactive, both
2.2. Nuclear Decay Rates

the parent ($\lambda_P$) and daughter ($\lambda_D$) decay constants must be considered to evaluate the daughter population. This is because the daughter population will grow according to the decay rate of the parent, and also experience loss that will be proportional to the daughter population at any point in time. In the following equations, an additional subscript ‘0’ is used to denote parent ($N_P$) or daughter ($N_D$) populations at $t = 0$:

$$N_D = N_{P0} \frac{\lambda_P}{\lambda_D - \lambda_P} (e^{-\lambda_P t} - e^{-\lambda_D t}) + N_{D0}e^{-\lambda_D t} \quad (2.5)$$

The final term in equation [2.5] accounts for an initial daughter population at $t = 0$. This equation also assumes that the parent nuclide decays exclusively to the daughter isotope. In reality, this may not be the case, and the relevant branching ratios will need to be applied.

### 2.2.1 Reaching Equilibrium

There are two limiting cases for equation [2.5], allowing it to be simplified substantially. Where $\lambda_P >> \lambda_D$ (the half-life of the parent is much shorter than that of the daughter), then at large values of $t$ (compared with the parent half life), the parent nuclide will have mostly decayed away, leaving a daughter population to decay with its own half life.

The second (and far more interesting) limiting case is where $\lambda_P << \lambda_D$, and the half-life of the parent is much greater than that of the daughter. For values of $t$ far greater than the half life of the daughter, equation [2.5] reduces to:

$$N_D = N_{P0} \frac{\lambda_P}{\lambda_D} e^{-\lambda_P t} \quad (2.6)$$

$$\therefore A_D = \lambda_P N_P \quad (2.7)$$

The activity of the daughter ($A_D$) can therefore be expressed as the product of the parent’s decay constant and the parent population, i.e. its activity matches that of the parent. This is because the parent and daughter are in secular equilibrium, and the daughter is decaying at the same rate at which
it is formed.

Another case where equilibrium is achieved occurs when the half-life of the parent is longer than that of the daughter, but not significantly so ($\lambda_P < \lambda_D$). For a time $t$ that is significantly greater than the half life of the daughter isotope, equation 2.5 can be simplified to:

\[
N_D = N_{P0} \frac{\lambda_P}{\lambda_D - \lambda_P} (e^{-\lambda_P t}) 
\] (2.8)

\[
\therefore \frac{N_D}{N_P} = \frac{\lambda_P}{\lambda_D - \lambda_P} 
\] (2.9)

The ratio of the number of parent to daughter nuclei therefore tends to a constant value, as defined by the isotopes relative decay constants. The closer the decay constants, the longer it will take to achieve transient equilibrium.

It is possible to have many radioactive generations for a radioactive isotope, where each decay transmutes the isotope via various decay methods. These are known as ‘decay chains’ or ‘radioactive series’.

2.3 $^{232}$Th, $^{235}$U, $^{238}$U, & $^{237}$Np Decay Chains

There are four major naturally occurring decay chains, and each is known by the most stable isotope within it. Three of these ($^{232}$Th, $^{235}$U & $^{238}$U) are long lived enough that they are still present within the earth in significant quantities, allowing us to measure them today. Small amounts of the fourth series ($^{237}$Np) may also be present in the environment due to the artificial production of $^{241}$Pu (the head of the $^{237}$Np decay chain) since the 1940’s.

Naturally occurring ores of these materials are generally assumed to be in secular equilibrium, however geological activity or isotope extraction/ore processing can significantly alter this. Each decay chain is detailed below (note that where additional decay branches are identified in the captions, these are restricted to branches that decay via $\alpha$ and $\beta$ modes).
Figure 2.1: The $^{232}\text{Th}$ decay chain. All nuclides in the decay chain will be present in samples that contain thorium, and if left undisturbed for a sufficiently long time, be in equilibrium with $^{232}\text{Th}$ (with the possible exception of $^{220}\text{Rn}$, which may escape the sample as it is a noble gas). $^{232}\text{Th}$ makes up almost all of the natural thorium found in the earth. Image from reference\textsuperscript{[21]}. 
Figure 2.2: The $^{235}$U decay chain. In addition to the nuclides seen, there is a weak decay branch from $^{231}$Th, which may $\alpha$ decay into $^{227}$Ra before $\beta$ decaying back into the main branch. This decay chain actually starts at $^{239}$Pu, however as $^{235}$U has a much longer half-life only this is found in (natural) terrestrial sources. Image from reference [21].
Figure 2.3: The $^{238}\text{U}$ decay chain. Weak decay branches include $^{218}\text{Po}$ $\beta$ decaying through $^{218}\text{At}$ to $^{218}\text{Rn}$ (with both daughter nuclides also $\alpha$ decaying back into the main chain). Image from reference [21].
2.3. $^{232}$Th, $^{235}$U, $^{238}$U, & $^{237}$Np Decay Chains

Figure 2.4: The $^{237}$Np decay chain. The head of this chain is $^{241}$Pu, which $\beta$ decays into $^{241}$Am. This subsequently $\alpha$ decays into $^{237}$Np, which is the longest lived nuclide in the chain. This decay chain has gained importance since the production of Plutonium for early nuclear weapons. Image from reference [21].
2.4 Nuclear Interactions

Ideally, a $\gamma$-spectrometer would have a response function consisting of only the full photopeak energies ($E_\gamma$), with no continuum or background. In the real world however, spectra often have very prominent continua, often masking important spectral information\cite{18}.

![Figure 2.5: Monte Carlo calculation of the contribution to the full energy photopeak for different energy loss mechanisms in a 6cm x 6cm HPGe detector. From Roth\cite{22}](image)

Photons can interact via the photoelectric effect, Compton scattering or pair production, and the probability of each interaction type is energy dependent, (see Figure 2.5). For environmental analysis, the energy range of interest is between 50 keV-3 MeV, and so Compton scattering is the dominant mechanism. The cross-section for Compton scattering ($\sigma_{\text{compton}}$) varies linearly with the $Z$ of the detector material, while the cross-section for the photoelectric effect varies as $\sigma_{\text{photo}} \sim Z^{4-5}$, and for pair production as $\sigma_{\text{pair}} \sim Z^2$\cite{23}. Choice of a high $Z$ material will therefore improve both the full photopeak efficiency and the proportion of photons stopped in the detector.

2.4.1 Photoelectric effect

The incident photon interacts with an atomic electron, ejecting it with a kinetic energy equal to the difference between the photons energy and the
binding energy of the electron. The resulting electron shell vacancy can be filled by either capturing a free electron or via rearrangement of the electrons in other shells, resulting in the emission of a characteristic X-ray or Auger electron. This is the dominant mechanism at low energies ($\lesssim 100$ keV), and the probability for full absorption decreases rapidly with increasing photon energy ($\propto E^{-3}_{\gamma}$)\cite{19}.

### 2.4.2 Compton Scattering

Compton Scattering is the process whereby an incident photon scatters off of an atomic electron, resulting in a photon with reduced energy, and an electron carrying the energy lost from the photon\cite{19}. The energy of the photon and electron depend upon the angle at which the interaction occurred, and is described by the following equation (assuming the electron is free and at rest):

$$E'_{\gamma} = \frac{E_{\gamma}}{1 + \frac{E_{\gamma}}{m_0c^2}(1 - \cos \theta)}$$

(2.10)

where $E_{\gamma}$ is the energy of the incident photon, $m_0c^2$ the rest mass energy of the electron and $\theta$ the angle through which the photon is scattered. When $\theta = 0^\circ$, the photon is forward scattered (or not scattered at all), and equation 2.10 reduces to $E_{\gamma} = E'_{\gamma}$ (as expected). For high energy photons that are completely back scattered ($\theta = 180^\circ$), equation 2.10 reduces to $E_{\gamma} \approx mc^2/2$.

The probability of a photon Compton Scattering at an angle $\theta$ is given by the *Klein-Nishina* formula:

$$\frac{d\sigma_c}{d\Omega} = r_0^2 \left[ \frac{1}{1 + \alpha(1 - \cos \theta)} \right]^3 \left[ \frac{1 + \cos \theta}{2} \right] \left[ 1 + \frac{\alpha^2(1 - \cos \theta)^2}{(1 + \cos^2 \theta)[1 + \alpha(1 - \cos \theta)]} \right]$$

(2.11)

Here $\alpha$ is the photon energy in units of electron rest mass energy ($\alpha = E_{\gamma}/mc^2$), and $r_0$ is the classical electron radius ($r_0 = e^2/4\pi\epsilon_0mc^2 \approx 2.818$ fm).
2.5. Environmental Radiation

2.4.3 Pair Production

During pair-production the impinging photon creates an electron-positron pair with kinetic energies $T_-$ & $T_+$ respectively. An atom must be present nearby for momentum conservation, and the energy of the incident photon must be greater than the rest masses of the electron and positron combined:

$$E_\gamma = 2mc^2 + T_- + T_+$$  \hspace{1cm} (2.12)

Pair-production only contributes significantly at higher energies, and becomes the dominant interaction mechanism at $E_\gamma > 5$ MeV.

2.5 Environmental Radiation

2.5.1 Background radiation

Environmental $\gamma$-spectroscopy often requires measurements on weak ($\leq 1$ kBq), distributed sources. Such samples may be counted for days to achieve the re-
2.5. Environmental Radiation

Required sensitivity, and background radiation greatly affects the measurement times required. A full understanding of the background present in a laboratory is therefore critical for highly sensitive $\gamma$-spectroscopy systems.

**Terrestrial radiation** - This primarily consists of radioisotopes from the $^{238}$U, $^{235}$U and $^{232}$Th decay chains, $^{40}$K, and $^{222}$Rn. Apart from $^{222}$Rn, these may all be present in the detector materials, cryostat, shielding and the building materials that house the laboratory. $^{222}$Rn is usually present in the air, and as such is difficult to minimise. Standard lead shielding is also contaminated with $^{210}$Pb (typically up to 500 Bq/kg), although low background (aged) lead can reduce this to 25 Bq/kg or less.

**Fluorescence & Compton scattering** - Atoms in the shielding can become ionised or excited by impinging radiations, and then deexcite emitting a characteristic x-ray. For lead (which makes up the majority of the shielding) these are around 74–85 keV, and are typically shielded with a liner of low Z material such as tin and cadmium. These also emit characteristic x-rays from 23–29 keV, which can be shielded with an even lower–Z material (often copper). Radiation from the source will also interact with these liners, where it is far more likely to Compton scatter than it would be in the lead due to their lower Z value. Excessive liner thicknesses will therefore increase the background seen in a detector, despite suppressing any fluorescence.

**Cosmic radiation** - The main components seen in laboratory detectors are secondary radiation from cosmic ray interactions within the upper atmosphere. These include high energy muons and fast neutrons, both of which may require several hundred meters of overburden (such as rock in the case of underground laboratories) to shield. Specialist plastics can be effective for thermalising and absorbing the neutron flux, such as polyethylene (PE) and borated polyethylene (PE:B) respectively, however neutrons cannot be completely removed from the system as they are also produced by the interaction of muons with high
Z materials (such as lead shielding). The background caused by the cosmic muon flux itself can be reduced by using active shielding, with reductions of up to 75% possible\cite{26}, however not eliminated due to the highly penetrating nature of the radiation. Some of the pertinent radionuclides produced by interactions with cosmic radiation (and any shielding materials, detector materials and the atmosphere itself) are detailed in table 2.1.

### 2.5.2 Isotopes of Interest

The focus of this thesis is the detection of low energy (<300 keV) radionuclides that are indicative of material releases from nuclear reactors, and the detonation of nuclear weapons. The most important isotopes that are monitored include $^{140}$Ba, $^{95}$Zr, $^{147}$Nd, $^{131}$I, $^{134}$Cs, and $^{137}$Cs. All these are relatively easy to detect and quantify as they $\gamma$ decay with a large branching ratios and high multiplicity. There are many more isotopes, however, that are more difficult to detect, both due to the characteristics of each nuclides decay path, and the fact that they are often obscured by the Compton continuum from the aforementioned, higher energy decays. These include $^{241}$Am, $^{144}$Ce, $^{99}$Mo, $^{141}$Ce, $^{235}$U, $^{95\text{m}}$Nb and $^{99\text{m}}$Tc. All of these nuclides are important from both a nuclear security viewpoint, and for environmental studies, as they are reliable indicators of material that has undergone nuclear fission.

### 2.6 Radiation Detection

The basis of all radiation detectors is a material that stops an impinging radiation. This creates a signal that can then be amplified and measured to determine the properties of the incoming radiation. The two main types of radiation detector used during this project are outlined below.

#### 2.6.1 Inorganic Scintillators

Scintillators use the process of prompt fluorescence to convert the kinetic energy of an incoming radiation into fluorescence photons. These are collected
Table 2.1: Common $\gamma$–rays observed in background spectra as a result of Cosmic radiation, compiled from references [27] (marked $^1$) & [28] (marked $^2$). Note that it is not only interactions with the detector crystal itself, but with all materials surrounding the detector (including the atmosphere) that produces this background.

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>Isotope &amp; reaction</th>
<th>Source of interaction/comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>53.5</td>
<td>$^{73m}$Ge</td>
<td>$^{72}$Ge (n,$\gamma$) in detector crystal$^1$</td>
</tr>
<tr>
<td>66.6</td>
<td>$^{73m}$Ge</td>
<td>$^{72}$Ge (n,$\gamma$) sum 13.1 + 53.5 keV, in crystal$^1$</td>
</tr>
<tr>
<td>139.7</td>
<td>$^{75m}$Ge</td>
<td>$^{74}$Ge (n,$\gamma$) in detector crystal$^1$</td>
</tr>
<tr>
<td>198.4</td>
<td>$^{71m}$Ge</td>
<td>$^{70}$Ge (n,$\gamma$) sum 175.0 + 23.4 keV, in crystal$^1$</td>
</tr>
<tr>
<td>477.6</td>
<td>$^{10}$B (n,\alpha)</td>
<td>Reactions with trace amounts of boron$^1$</td>
</tr>
<tr>
<td>511.0</td>
<td>Annihilation</td>
<td>$\beta^+$ emitters, &gt; 1.022 MeV $\gamma$–rays$^1$</td>
</tr>
<tr>
<td>537.4</td>
<td>$^{206}$Pb (n,n')</td>
<td>Reactions in lead$^1$</td>
</tr>
<tr>
<td>558.4</td>
<td>$^{114}$Cd (n,n')</td>
<td>Reactions in cadmium$^1$</td>
</tr>
<tr>
<td>569.7</td>
<td>$^{207}$Pb (n,n')</td>
<td>Reactions in lead$^1$</td>
</tr>
<tr>
<td>595.9</td>
<td>$^{74}$Ge (n,n')</td>
<td>Broad and asymmetric due to reactions in crystal$^1$</td>
</tr>
<tr>
<td>669.6</td>
<td>$^{63}$Cu (n,n')</td>
<td>Reactions in copper$^1$</td>
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<tr>
<td>691.5</td>
<td>$^{72}$Ge (n,n')</td>
<td>Broad and asymmetric due to recoil summation$^1$</td>
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<tr>
<td>718.3</td>
<td>$^{10}$B (n,n')</td>
<td>Reactions with trace amounts of boron$^1$</td>
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<td>803.1</td>
<td>$^{206}$Pb (n,n')</td>
<td>Reactions in lead$^1$</td>
</tr>
<tr>
<td>834.0</td>
<td>$^{72}$Ge (n,n')</td>
<td>Broad and asymmetric due to recoil summation$^1$</td>
</tr>
<tr>
<td>843.8</td>
<td>$^{27}$Al (n,n')</td>
<td>Reactions in aluminium$^1$</td>
</tr>
<tr>
<td>846.8</td>
<td>$^{56}$Fe (n,n')</td>
<td>Reactions in iron$^1$</td>
</tr>
<tr>
<td>962.1</td>
<td>$^{63}$Cu (n,n')</td>
<td>Reactions in copper$^1$</td>
</tr>
<tr>
<td>1014</td>
<td>$^{27}$Al (n,n')</td>
<td>Reactions in aluminium$^1$</td>
</tr>
<tr>
<td>1039</td>
<td>$^{70}$Ge (n,n')</td>
<td>Broad and asymmetric due to recoil summation$^1$</td>
</tr>
<tr>
<td>1369</td>
<td>$^{27}$Al (n,\alpha)</td>
<td>Reactions in aluminium$^1$</td>
</tr>
<tr>
<td>1779</td>
<td>$^{27}$Al (n,\gamma)</td>
<td>Reactions in aluminium$^1$</td>
</tr>
<tr>
<td>2223</td>
<td>$^{1}$H(n,\gamma)</td>
<td>Reactions in hydrogenous material$^1$</td>
</tr>
<tr>
<td>2312</td>
<td>$^{14}$N(n,n')</td>
<td>Reactions in nitrogen gas near the detector$^2$</td>
</tr>
<tr>
<td>4439</td>
<td>$^{12}$C(n,n')</td>
<td>Reactions in carbon near the detector$^2$</td>
</tr>
<tr>
<td>5106</td>
<td>$^{14}$N(n,n')</td>
<td>Reactions in nitrogen gas near the detector$^2$</td>
</tr>
<tr>
<td>6129</td>
<td>$^{16}$O(n,n')</td>
<td>Reactions in oxygen gas near the detector$^2$</td>
</tr>
<tr>
<td>6506</td>
<td>$^{74}$Ge(n,n')</td>
<td>Q values of neutron capture in Ge crystal$^1$</td>
</tr>
<tr>
<td>6783</td>
<td>$^{72}$Ge(n,n')</td>
<td>Q values of neutron capture in Ge crystal$^1$</td>
</tr>
<tr>
<td>7115</td>
<td>$^{16}$O(n,p)</td>
<td>Reactions in oxygen gas near the detector$^2$</td>
</tr>
<tr>
<td>7416</td>
<td>$^{70}$Ge(n,n')</td>
<td>Q values of neutron capture in Ge crystal$^1$</td>
</tr>
<tr>
<td>10199</td>
<td>$^{73}$Ge(n,n')</td>
<td>Q values of neutron capture in Ge crystal$^1$</td>
</tr>
</tbody>
</table>

by a photomultiplier that acts to amplify this signal, allowing it to be measured using suitable electronics. Scintillators can be made from both organic and inorganic materials, with the former generally used for $\beta$-spectroscopy and neutron detection (due to their high Hydrogen content), and the latter
2.6. Radiation Detection

for $\gamma$-spectroscopy. Knoll defines six key criteria for an ideal scintillation material:

1. The energy of the impinging radiation should be converted into detectable light with a high efficiency
2. The energy conversion should be linear, i.e. the light yield should be proportional to the deposited energy
3. The medium should be transparent to the wavelength of its own emission
4. The decay time of the induced luminescence should be short so that fast signal pulses can be generated
5. The material should be of good optical quality and suitable for manufacture in sizes large enough to be of interest
6. The index of refraction should be close to that of glass to enable an efficient coupling to a photomultiplier

2.6.1.1 Band Structure

Electrons within a crystalline, inorganic scintillation material can either be found in the conduction or valence bands. The valence band is formed of bound electrons, while the conduction band electrons are free to move throughout the material. When excited, an electron can 'jump' from the valence to the conduction band, and then de-excite back into the valence band by emitting a photon of energy equal to that of the band gap. This de-excitation however is an inefficient process, and the band gap is often too great to emit a visible photon. Impurities (activators) are therefore added that modify the band structure around the lattice sites where the activators sit. If the right activators are chosen, this can reduce the band-gap enough to create visible photons, and substantially increase the efficiency of electron de-excitation. No scintillator material is perfect however, and losses still occur through quenching (where the electron de-excites via radiationless transitions) and phosphorescence (where a transition to the ground state is forbidden, and the electron requires thermal energy to achieve a state that can decay).
2.6.2 Semiconductors

Semi-conductor based materials typically have far better energy resolution than scintillators due to the relatively small amounts of energy required to create an information carrier. In scintillators, this can be of the order $\sim 100$ eV\textsuperscript{[18]}, and therefore the inherent statistical uncertainty in the number of information carriers limits the resolution of the detector. In semiconductors, the conduction and valence bands are only separated by $\sim 1$ eV, and therefore thermal energy from the environment will occasionally excite an electron into the conduction band. HPGe detectors are cryogenically cooled to minimise the thermal noise from this process, which results in a detector with $\sim 0.2$ \% resolution instead of the 5-20 \% typically seen in inorganic scintillators. An electrical bias is applied so that when an electron-hole pair is created by the incoming radiation, the electron and hole move in opposite directions. These charge-carriers are collected at the electrodes, producing a measurable signal.

2.7 Efficiencies and Abundances

Apparent peak efficiencies ($\varepsilon$) were calculated using equation 2.13, where $N$ is the total number of counts in the peak, $t$ is the detection time in seconds, $A$ the activity of the source in decays per second (Bq), and $\phi$ the photons branching ratio\textsuperscript{[18]}. $C_i$ is a correction factor due to dead time, radionuclide decay and coincidence summing corrections.

$$\varepsilon = \frac{N}{tAbC_i}$$ (2.13)

The isotopic abundances are calculated using equation 2.14, where $N_0$ is the total number of atoms present, and $\lambda$ is the decay constant:

$$N_0 = \frac{N}{\lambda tb \varepsilon}$$ (2.14)
2.8 Minimum Detectable Activity (MDA)

The Minimum Detectable Activity (MDA) of a detector is an energy specific measure of the activity required to identify a radiation source with an amount of statistical certainty (normally 95%).

A widely used form of MDA is the *Currie* equation\[18\]. This evaluates the level of counts needed from a source to ensure a false-negative and false-positive rate of no greater than 5%. This statistical level can be changed by modifying the number of standard deviations from the mean that the equation is based on (5% uses a value 1.64 standard deviations from the mean). The MDA can be calculated in the following way:

\[
N_R = 4.65 \sqrt{N_B} + 2.71
\]  \hspace{1cm} (2.15)

\(N_R\) is the number of counts required for statistical certainty, and \(N_B\) the number of background counts. To convert this value to a minimum detectable activity \((\alpha)\), additional factors for the branching ratios, detection efficiency, and the counting time must be considered:

\[
\alpha = \frac{N_R}{bet}
\]  \hspace{1cm} (2.16)

Note that the smaller \(\alpha\) is, the better. The MDA performance of a detector is therefore proportional to the detectors efficiency, and inversely proportional to the square root of the number of background counts across the peak\[29\]. Simply increasing the size of the detector will therefore provide a greater MDA benefit than shielding up to a point, however this is not a cost effective solution. Large detectors also suffer from increased summing effects, and Peak to Count (P/C) ratios level off around 50-66% relative efficiency\[30\]. Bigger detectors also see much more environmental radiation, reducing the MDA benefit gained.
Chapter 3

Compton Suppression

3.1 Methods

3.1.1 Anti-Coincidence mode

As crystals are a finite size, scattered photons may escape the detector before depositing their full energy. These events can be preferentially rejected if the escaped photons are detected in coincidence by a surrounding guard detector (GD).

In Figure 3.1 photons travel down a heavy metal collimator to prevent direct interaction with the GD. These are incident on the primary detector (PD) in the centre, and a substantial portion of these are scattered back out into the GD. If the GD is an active volume it can generate a signal to prevent any coincident signals from being recorded in the PD. This anti-coincidence setup is the most common for Compton Suppression systems, and has been studied extensively, with common suppression factors of 4.1-12.1 for $^{60}$Co\cite{5,31,32,33} peaks, and 3.9-12.7 for the $^{137}$Cs\cite{5,31,32,34} peak.

Reductions of the Compton continuum of up to 85 have also been reported at the Compton edge\cite{35,36}, with significant variance in each systems effective energy range. These differences are primarily due to their geometries and build materials, hence the popularity of Monte-Carlo simulations to optimise Compton Suppression systems before production.

By vetoing any event that occurs in coincidence with a signal from the
3.1. Methods

Figure 3.1: A fairly standard multiple HPGe and BGO Compton Suppression system. Two coaxial HPGe detectors are in the middle, with a thin planar HPGe in front of these. BGO shields are employed in both the forward and backscattered positions.

Figure 3.2: A Compton Suppressed HPGe detector surrounded by a 6-way segmented NaI(Tl) shield.

GD, some photopeak counts will be lost due to chance coincidences with other radiation. High multiplicity γ’s will also be partially suppressed as they can not be discriminated from normal escape events. Nuclei with such cascade suppressions may gain little or no benefit from CS systems as the continuum
3.1. Methods

Figure 3.3: A suppressed and unsuppressed spectrum for a $^{60}$Co source. From Masse.$^{35}$

Reduction is cancelled out by the reduction in photopeak.$^{35}$ High count rates can also detrimentally affect CS systems, as the increased dead time and higher flux increases the rate of chance coincidences.

3.1.2 Sum-Coincidence mode

Most Compton continuum events are caused by the single scattering of an incoming photon, which subsequently escapes the detector. Full energy events typically involve multiple scatterings (with escape from the detector significantly less likely with each further interaction). Therefore the continuum can be suppressed by requiring the event to register in two adjacent segments/detectors. This provides excellent suppression at the expense of efficiency; Palms et al.$^{37}$ achieved a 25 fold reduction in the continuum with a 5 fold reduction in photopeak, resulting in a Compton suppression factor of 4-5.

3.1.3 Pair Spectroscopy

This approach involves recording only the double escape peak (at $E_{\gamma}-1.022\text{MeV}$), therefore limiting it to energies where a significant proportion of interactions will involve pair production. For an event to be recorded in Pair Spectroscopy, the initial event must be detected in the PD, and both the 511 keV annihilation photons must be detected in the GD. This approach sacrifices efficiency
but can produce very clean spectra. A small residual continuum is generally caused by interactions near the edge of the crystal (where the likelihood of scattering out is high), which can be significantly reduced by using pulse shape analysis to discard slow rising charge pulses\[^{38}\].

### 3.1.4 Pulse Shape Analysis (PSA)

The pulse shape from HPGe detectors depends on the charge collection time, which is determined by the initial position of the $\gamma$-ray interaction. Traditional methods of Compton suppression rely on ultra-low background techniques and multiple detectors to veto or record an event. PSA uses the information in each charge pulse to infer both where the event occurred in the detector, and the likelihood that it is a full energy or escape event\[^{39, 40}\].

### 3.2 Design Considerations

#### 3.2.1 Primary Detectors

HPGe is almost exclusively used as the primary detector due to its excellent energy resolution ($\sim 0.18\%$ at 662 keV), the availability of large volume crystals, and Germanium’s high Z value (increasing the materials stopping potential). The major drawback, (apart from the cost), is the need for liquid nitrogen cooling. This can restrict the geometry, and therefore the efficiency that the guard detector can achieve. Specifically designed cooling apparatus however can minimise this disruption, and mechanically cooled HPGe detectors are now widely available.

While few materials can match the resolution of HPGe ($\sim 0.2\%$), LaBr(Ce) detectors can approach 3\%, a substantial improvement over NaI(Tl) (6-7\%) and BGO (16\%)\[^{18}\]. This is also achieved at room temperature, allowing a more effective guard detector than can be achieved with a HPGe primary detector.
3.2.2 Guard Detectors

NaI(Tl) is an effective low-cost active shield, and has excellent properties where space is not restricted. If this is not the case, BGO is often used\[^5, 31\] as it has 3 times the linear attenuation coefficient at 500keV, allowing for substantial space savings and/or performance improvements (albeit at higher cost). NaI(Tl) does have a better energy resolution and greater light yield than BGO\[^18\], and these limitations may be of importance for systems that also perform $\gamma-\gamma$ coincidence work.

3.2.3 Geometries

Geometry is perhaps the most important aspect of CS system design. Inadequate shielding or insufficient detector material can dramatically alter the performance of the system, and these issues are most commonly addressed with Monte-Carlo modelling and optimisation studies. The path of scattered photons is highly energy dependent, and therefore the geometry of the detectors should be tailored to the materials being analysed.

Guard detectors typically surround the primary crystal, with the aim of covering the maximum solid angle for scattered events. Photons that are scattered through 180° (and therefore make up the Compton edge) would be best suppressed with a veto detector placed above the primary crystal, while high energy photons that are predominantly forward scattered (and only deposit a small amount of energy) may only be captured with another veto detector below the primary crystal. Between these two extreme cases, photons are scattered in a variety of angles, and therefore an all encompassing geometry is most effective.

Achieving this in reality, however, is increasingly difficult as such a design (with minimal dead layers but more active material) presents a significant engineering challenge. This is especially true when considering efficient light collection and signal formation in the guard detector, and combining this with effective shielding from background radiation. Compton suppression systems for environmental studies therefore represent a compromise between the effec-
3.2. Design Considerations

tiveness of the primary/guard detectors and the low-background environment in which the system is placed.

3.2.4 Electronics/Timing Circuits

The electronic systems used for pulse discrimination and vetoing vary greatly in cost and complexity. Digital ‘all-in-one’ systems have also become available as an alternative to traditional analogue systems, and each will be briefly described below.

3.2.4.1 Analogue Electronics

Analogue electronics are used for the shaping, delay and discrimination of event signals from the detector. Anti-coincidence setups achieve this by blocking the Analogue to Digital Converter (ADC) from registering the event if a coincident event is detected in the GD.

A study by Canberra[41] evaluated 3 different analogue electronics setups, ranging from basic systems to those that are more complex. The most basic setup utilised logic pulses from Canberra 2002 Amplifiers which were fed into a Canberra 2040 Coincidence unit. If the two pulses were received within a set resolving time an output was generated, which was shaped in a delay and gate unit and prevented the ADC from recording. The more complex system used Timing Filter Amplifiers (TFA’s) and Constant Fraction Discriminators (CFD’s) for a greater time resolution, before delaying and shaping the pulse that would prevent the ADC from recording. Both systems performed similarly, with the two achieving suppression ratios of 2.37 and 2.45 respectively, suggesting that ultra-fast electronics may not be necessary for some common radioanalytical applications.

3.2.4.2 Digital Electronics

Traditionally, analogue electronics were used for the shaping and timing of incoming charge pulses, however, digital systems replace these with numerical analysis of the incoming charge pulse to determine its properties. Such
3.3 Quantifying Compton Suppression

Several methods are available for quantifying the levels of suppression achieved, however the main ones used in this thesis are the Peak to Count ratio (P/C), Peak to Total ratio (P/T) and the Compton Suppression Factor (CSF).

- **P/C ratio** - This is defined as the ratio of the counts in the highest photopeak channel to the counts in a typical channel of the Compton continuum. This is usually taken to be a flat, representative portion just to the left of the Compton edge.

- **P/T ratio** - The peak-to-total ratio (P/T) is expressed as the ratio of the counts in the full-energy peak to the total counts in the spectrum.

- **Peak CSF** - This is the ratio of P/C for suppressed and unsuppressed spectra, which also takes into account the reduction in photopeak efficiency as well as the suppression of the continuum. Unless otherwise
3.3. Quantifying Compton Suppression

stated, all CSF factors refer to peak CSF values;

\[
CSF_{\text{Peak}} = \frac{(P/C_{\text{suppressed}})}{(P/C_{\text{unsuppressed}})} \quad (3.1)
\]

- **Total CSF** - This is the ratio of P/T for suppressed and unsuppressed spectra;

\[
CSF_{\text{Total}} = \frac{(P/T_{\text{suppressed}})}{(P/T_{\text{unsuppressed}})} \quad (3.2)
\]

A summary of achieved suppression ratios (for a variety of methods) is shown in table 3.1.

<table>
<thead>
<tr>
<th>Author</th>
<th>CSF</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>100</td>
</tr>
<tr>
<td>Cooper (A) [36]</td>
<td>-</td>
<td>5.2</td>
</tr>
<tr>
<td>Beetz (A) [46]</td>
<td>-</td>
<td>15.0</td>
</tr>
<tr>
<td>Aarts (A) [47]</td>
<td>7.2</td>
<td>-</td>
</tr>
<tr>
<td>Moszynski (A) [32]</td>
<td>12.1</td>
<td>2.5</td>
</tr>
<tr>
<td>Masse (A) [35]</td>
<td>-</td>
<td>23.0</td>
</tr>
<tr>
<td>Mauerhofer (A) [5]</td>
<td>8.7</td>
<td>4.5</td>
</tr>
<tr>
<td>Lin (A) [48]</td>
<td>-</td>
<td>18.8</td>
</tr>
<tr>
<td>Voigt (A) [31]</td>
<td>4.0</td>
<td>23.0</td>
</tr>
<tr>
<td>Fukuda (A) [33]</td>
<td>6.0</td>
<td>13.2</td>
</tr>
<tr>
<td>Peerani (A) [49]</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Duchene (A,S) [50]</td>
<td>-</td>
<td>2.0</td>
</tr>
<tr>
<td>Schumaker (A,S) [51]</td>
<td>-</td>
<td>7.8</td>
</tr>
<tr>
<td>Pearson* (PSA) [43]</td>
<td>-</td>
<td>7.5</td>
</tr>
</tbody>
</table>

Table 3.1: A summary of suppression ratios for various experiments. CSF is defined earlier, and the additional data is the ratio of counts in the suppressed to unsuppressed spectra at specific energies. All data is for $^{60}$Co spectra. Datasets followed by an (A) used anti-coincidence techniques, those with (S) used sum-coincidence, and those with (PSA) digital techniques such as pulse shape analysis or gamma ray tracking. Simulated or calculated data is marked with an ‘*’. 
Chapter 4

GEANT4 – A Monte Carlo Toolkit

4.1 Introduction

Monte Carlo methods use random sampling to solve problems that are difficult to consider analytically, and as such are ideal for evaluation using computational models. Results are obtained by repeating the simulation many times, and collating the outputs to provide an answer. As the number of simulations increases, the fractional error will reduce, improving the statistical significance of the results from the computational model. Systematic errors will arise due to inaccuracies in the model, however these can be quantified and propagated through any analysis.

GEANT4[^52] is a publicly available Monte-Carlo toolkit developed at CERN, which enables the user to create accurate simulations of particle propagation through and interaction with matter. Originally it was used to model particle and nuclear physics experiments, however GEANT4 is now used in a wide range of fields including nuclear medicine and radiation protection. As the cost of computing facilities decreases (the main limitation for computer based modelling), Monte Carlo simulations have become essential when developing new equipment, providing substantial cost savings on prototyping and design work. Such simulations also provide a method to improve understanding of existing processes, and of critical points in an experimental setup.
Alternatives to GEANT4 were considered, including MCNP6\textsuperscript{53} and FLUKA\textsuperscript{54}, however neither is as versatile as GEANT4. This is due to the unique ability to modify both the code base of the GEANT4 toolkit and of the simulations themselves, something that is not possible with the other two Monte Carlo packages. MCNP6 also requires a license and is a trademark of Los Alamos National Security (LLC, Los Alamos National Laboratory), and FLUKA is requires code to be written in FORTRAN 77 – a somewhat outdated version of the FORTRAN language that lacks many of the features available using more modern approaches. For comparison, GEANT4 is open-source and freely available, and written in c++ (a more modern, widely used programming language). GEANT4’s flexibility, large user-base and comprehensive physics models ultimately make it ideal for the analysis of existing systems, and the development of a new, Compton Suppressed Low Energy $\gamma$ spectrometer.

4.2 GEANT4 Application Overview

GEANT4 is designed to simulate the passage of particles through matter, and comes pre-configured with many standard options and examples for a user to modify. By default, additional data libraries for certain physics processes are not installed. The user has to specifically request these, or use the standard, reduced data library installed with GEANT4. The typical installation does not come with any Graphical User Interface (GUI), and is instead command line driven. GUI interfaces are available for the GEANT4 toolkit, however this is generally as part of a framework that acts as an interface between the user and GEANT4, which are usually developed for experiments involving large collaborations where some constancy is needed in the structure and definition of the simulations. This reduces the flexibility of the toolkit (as the source code and executable have to be built within the limitations of the framework) however such simulations are far easier to perform for non-expert users.

Once installed, the GEANT4 toolkit is used to compile each simulation from a number of source files, which are typically contained within a directory
for that project/simulation. This creates an executable that can be run in interactive or batch mode, where the user can issue commands one at a time, or specify a ‘macro’ file (which contains a list of commands) respectively. The modification of simulations between runs can be achieved using specific messenger classes for each component (geometry, particle generation, etc.) or via the use of scripting languages such as BASH\(^{55}\) on a linux system (and tools such as SED\(^{56}\) and AWK\(^{57}\)), which can generate macro files, edit source files, recompile and run the simulations as necessary (see figure 4.1).

Figure 4.1: A screenshot of a typical GEANT4 run, showing the terminal in the background on the left (used to run GEANT4 via the command line), a data file used to input source geometry parameters (top right), an example macro file with some common GEANT4 commands (bottom right), and a visualisation of the simulation being run. There are many predefined commands that can be used within the macro files, however the user can also create their own, such as enabling outputs and defining the source.

To create a simulation, the user must define a world, and populate this with materials and geometries. Primary particles (e.g. a radioactive source, proton beam, \(\gamma\) spectrum etc.) must then be defined, and the physics processes required for the simulation added. GEANT4 will then simulate the passage of the primary particles through the world, transporting each particle via a
series of steps.

For each step through the simulated geometry, GEANT4 calculates the mean free paths for any competing physics processes, and ‘chooses’ a process based upon the relative strengths of each interaction channel and a random number generator. This then determines the step length and the physics process to be simulated. Step lengths can also be defined by the user, and will be limited if the step encounters a physical boundary.

Each primary particle is known as an event, and GEANT4 stops tracking it when the particles kinetic energy reaches a cut-off threshold, or it exits the world volume. A user-defined number of events are run, creating the Monte Carlo simulation.

Information about each particle can be obtained at both the pre-step and post-step points, including the energy deposited per step, the type of particle, number of secondaries, position, trajectory etc. By combining the information across all steps, information for each event can be obtained, and therefore used to create useful outputs, such as energy spectra.

\section*{4.3 Implementation}

\subsection*{4.3.1 Physics Lists}

Low energy electromagnetic physics models (valid from 250 eV - 100 GeV) are used to model photon interactions with materials throughout all the simulations. This is implemented via a modular physics list, which uses the built-in GEANT4 particle definitions. This includes common bosons, leptons, mesons, baryons and an ion constructor. Modular physics lists are useful because they allow you to turn on/off certain processes, and even select different sub-lists to include (such as Livermore or Penelope based lists, which may use different models for photoelectric absorption, Compton scattering, etc.).

Within the physics list, all available electromagnetic processes are defined, as well as transportation through the geometry, radioactive decay, and an explicit stepping process. Fluorescence photons, Auger emission and Photon Induced X–ray Emission (PIXE) are all enabled by default, while the mini-
mum energy for the electromagnetic physics tables is reduced to 10 eV (from 100 eV). The stepping process allows the user to modify the length of the step for each particle type (which can be thought of as increasing/decreasing the ‘resolution’ of the simulation). The cuts used (which defines the cut–off threshold for each process), are also be defined here.

To enable the user to modify these parameters (and select different physics sub–lists), a messenger class has been implemented which allows changes to be made without re–compiling the simulation.

### 4.3.2 Primary Particle Generation

Primary particles are those that are defined within the primary generator, and are then propagated throughout simulated geometry. To define a particle, the type is specified ($\gamma$, neutron, etc.), and given a position and initial momentum/direction. To generate radioactive decays, ions are created using $Z$, $A$, and excitation values, and placed with no initial momentum. These are then disintegrated using two methods.

The first uses the radioactive decay module built into GEANT4, which (if the optional radioactive decay library is installed) disintegrates each primary event probabilistically to reproduce the decay behaviour seen in the laboratory. This module is data–driven, and utilises the ENSDF$^{58}$ (Evaluated Nuclear Structure Data File) library. Version 10.0 of GEANT4 uses the ENSDF library updated in August 2012, however versions prior to this use a 2006 release. The radioactive decay module includes decay cascades, where an excited nucleus may emit multiple $\gamma$–rays before it reaches the ground state, and the emission of multiple decay products, such as $\beta$ and $\gamma$ radiation. This method decays the selected isotope to stability, and therefore if the nuclide is part of a decay chain, multiple isotopes will be sequentially decayed within the same event until stability is reached. This is often not the intended effect, and therefore decay chains can be limited by specifying where the chain should end within the macro file that is used to run the simulation (when including the radioactive decay process within the physics lists, the messenger class required to do this will be automatically loaded).
The second method uses a framework developed for the input of decay particles. In this mode, the user can specify their own set of decay libraries, which reside in the same folder as that particular project. Multiple nuclides can be specified in a macro file (with the appropriate weightings) and the particle generator will search the user-defined library for each isotope. If no corresponding library file is found, the simulation will use the GEANT4 decay libraries, however if it is available then the particle generator will generate the specified particles individually by firing them isotropically. Currently, branching ratios (if \( > 10^{-9} \) per decay) are manually imported using the NuDat2 tool\(^{[59]}\), which extracts data from the ENSDF for each nuclide of interest. This method reduces the computing time required to simulate an \(^{241}\)Am source by a factor of 300, but does not take into account any angular or temporal correlations between emissions. This approximation contributes only a small percentage to the overall error within any typical simulation, as the accuracy is dominated by geometry errors. Using this method, a wide range of artificial complex sources can be created, utilising any of the particles defined internally within the GEANT4 libraries, user-defined branching ratios and multiple decay types within a single source.

### 4.3.3 Source Reproduction

The source matrix is reproduced by defining the source size, position and material (or mixture of materials), and then creating the object within the simulated geometry. The geometry information for the source is currently supplied by an external library file, which allows access to this information for particle generation. A decay can therefore be generated at a random coordinate within the source geometry, which can also be supplemented by casings/source holders as necessary. Summing effects due to the source activity’s are simulated by calculating the probability of a detector seeing from 1-20 events within the detectors characteristic decay time, and using this probability to generate extra decays in a corresponding number of events.
4.3.4 Recreating the Experimental Geometry

The geometry of the simulation is recreated as fully and accurately as possible. Complex shapes are created using boolean unions, and all major collections of components are implemented as assembly volumes. This allows the user to create all the individual parts of a detector, assemble them in their correct positions, and then position the entire assembly as one object. The benefits of this are two-fold, firstly the source code is much easier to manage, and secondly, the user can place as many copies of the detector in the simulation as is needed (especially useful when modelling large arrays, for example). An example simulated geometry is shown in figure 4.2.

A materials library is defined in the geometry file, which allows the user to assign a material to each object once it has been created. These can be created by specifying a material from the GEANT4 material database, which is comprised of pure materials (elements), NIST compounds, HEP (high energy physics) and nuclear materials, space materials, and bio–chemical materials. If the required material is not available from this library, the user can also define a customised material based on Z number, atomic weight, and density.

The simulations developed have progressed greatly since the original validation project. Initially all the geometry information was contained in one file, however a framework has now been created to allow different detectors/objects to be defined in separate files and included with a single line of code in the main geometry file. This has allowed a library of detector files to be created, which can be included/swapped/modified in a coherent, self–contained manner. This has greatly aided simulation development as the geometry and particle generators can now be maintained separately from the rest of the code, which deals with the physics, simulation structure and output (this generally does not change between simulations, and instead only has to be modified when changes are required due to an update of the GEANT4 toolkit).
4.3. Implementation

Figure 4.2: An example GEANT4 geometry, showing a cross-section of a coaxial HPGe detector (stripped of the cold finger and associated electronics). The main crystal is created using a single cylinder, from which a smaller cylinder and sphere are subtracted to create the void for the cold finger. A smaller cylinder is placed on top of this, and a rounded edge created by producing a quarter torus. All these geometrical shapes are joined with boolean unions to create one physical volume, which can be placed within the simulated geometry.

4.3.5 Extracting Information

There are two main methods for extracting information from a simulation. The first can be controlled via a macro file, and allows the user to create a scoring mesh. This places a grid of a defined size, position and resolution over the geometry, and accumulates the required quantities over the simulation (such as energy deposited, the dose deposited, the number of steps recorded, surface flux, etc.). These values can then be written to a file for later processing. The second method involves setting sensitive regions in the geometry, and implementing code to read out the energy deposited within these sensitive
regions every step/event as required.

For the majority of the simulations in this thesis, the second method is used, with the detector crystals set as sensitive volumes. The energy deposited at each step by an impinging particle is recorded, and these steps are then summed to calculate the total energy deposited per event (which is equivalent to the energy deposited in a detector within the laboratory). GEANT4 simulations, however, record the energy exactly, giving the simulated detector a perfect resolution. This must therefore be ‘smeared’ to approximate the detector output in reality. Once the detectors real resolution is measured using standard sources (usually over a range of 30–3000 keV), it is approximated with a function of the form $y = a + b.Ln(cx + d)$. The energy deposited in the simulated detector at the end of each event is then ‘smeared’ using a Gaussian function. This uses the function determined from the experimental data to calculate a resolution for that specific energy, and reproduce the detectors resolution in the simulated data.

### 4.4 Tuning the Models

There are many sources of error when creating a model of a detector system, however they are often dominated by unknowns within the source and the absolute geometrical accuracy of the detector reproduction. Such inaccuracies include unknowns in the detector (such as crystal positioning and alignment), and the determination of the crystals dead layers, which may vary across the detector surface[60]. Errors in the uniformity of the source matrix may also cause unwanted effects, and the thickness of attenuators within the detector are often only approximately known. Simulations can be carried out with manufacturer supplied dimensions, however they are unlikely to be accurate as the detectors low–energy response is particularly sensitive to geometry changes.

To minimise these errors, secondary measurements are made of all components to verify their dimensions. This involves precision measurements of all external dimensions, and if possible, radiography of the detector. The re-
sults of a x–ray can be scaled to the external dimensions measured, and also allow the operator to check crystal alignment and positioning. Even with radiography however, certain internal dimensions cannot be determined in this way, (such as the crystal dead layers), and so have to be ‘tuned’ to match the experimental data[61]. The most critical of these involve any materials that are between the crystal and the source, the exact positioning and shape of the crystal, and the extent of the dead layers within the crystal. When considering a Compton suppression system, the dead layers between the primary and secondary crystal also become crucial, which makes the simulation far more complex as these typically include the primary crystal holder & electrodes as well as the additional casings for the secondary crystal.

4.5 Simulating Coincidence Systems

In the laboratory, coincidence detectors (γ–γ systems, Compton suppression systems, etc.) are controlled using complex electronics with highly accurate timing systems. These synchronise the two detectors, allowing a gate to be set (in either hardware or software) that records/vetos coincident events (depending on the system). The timing of this gate is dependent on the electronics of each system, and the charge–collection time of the detector. Scintillation detectors are generally much faster than semi–conductor based systems, however charge–collection in either type of crystal is far slower (typically by up to $10^3$ times) than radiation transport within the system. Monte Carlo models can therefore replicate coincidences by recording when energy is deposited in multiple detectors within the same event. This would be equivalent to a perfect system, as there will be no losses due to electronics that would be seen in the laboratory. For a well set up system, however, these losses are minimal.

In the simulations presented, both the energy deposited and the volume in which the interaction took place are recorded at step level. The energy is summed separately for each volume, and the order of the interactions is also recorded. If energy is deposited in multiple volumes within a single event, then these are treated as coincident.
Chapter 5

Experimental Details

5.1 Experimental Overview

Environmental radiation detectors typically monitor unknown radioactive sources, and must accurately quantify the nuclides present. To achieve the levels of consistency required, several calibrations must be carried out before any measurements take place. This applies to all detection systems used in this thesis, including a variety of scintillation and semi–conductor crystals.

5.1.1 Detector Background

As well as detecting radiation emitted from the radioactive source, all detector systems will register events that originate from the surrounding environment. Provided this is relatively constant, measurements of ‘empty’ sources can be made to quantify the levels of background radiation expected. Before all experiments, background spectra were acquired (see figures 5.1 & 5.2), which were subtracted from the experimentally observed spectra with the source in place.

5.1.2 Energy Calibration & Peak Fitting

To define the energy range each detector operated over, numerous calibration sources were used. All were sources with a known activity and included single emitters as well as complex mixtures of nuclides that covered the entire energy
5.1. Experimental Overview

Figure 5.1: An example background collected over 6 days. This used a low-energy HPGe detector, which was optimised for photon energies <500 keV. The one peak seen is due to annihilation radiation at 511 keV.

Figure 5.2: The rate of acquisition for the spectrum in figure 5.1. Even with the bins set to two minute intervals, a large amount of statistical variation is seen.
range of interest. Once the data had been acquired, spectra were loaded into ROOT\cite{ROOT}, where peaks are identified using the TSpectrum class (see figures 5.3 & 5.4).

Figure 5.3: The peak searching routine is designed to find up to 200 peaks, which can then each be selected for fitting.

Figure 5.4: The fitted gaussian is used to calculate both the centroid of the peak, and the total area underneath it.

Once a peak has been identified with the analysis routine, it is fitted with a gaussian curve. The centroids can then be used to calibrate the channel
5.1 Experimental Overview

numbers, and therefore identify energies of unknown peaks. A linear calibration equation is used (of the form $y = mx + c$), with a minimum of 5 points for this process. The complex $\gamma$ source is NIST (National Institute of Standards and Technology) traceable, and comprised of $^{241}$Am (59.54 keV), $^{109}$Cd (88.03 keV), $^{57}$Co (122.06 keV), $^{139}$Ce (165.86 keV), $^{113}$Sn (391.68 keV), $^{137}$Cs (661.67 keV), $^{54}$Mn (834.84 keV), $^{88}$Y (898.04 and 1836.06 keV), $^{65}$Zn (1115.54 keV), and $^{60}$Co (1173.23 and 1332.49 keV).

5.1.3 Determining the Full Photopeak Efficiency

The same tools are used as for the calibration process, however this time the areas under the peak are used to calculate the full photopeak detection efficiency. For each peak, the underlying continuum is calculated using algorithms described in [63, 64] with an inbuilt function of the TSpectrum class. This can then be approximated with a linear function across the range of the peak, with the integral of this subtracted from the integral of the gaussian peak itself. Where multiplets could not be resolved, these peaks were fitted with overlapping Gaussians, as in figure 5.4.

Figure 5.5: Multiple gaussian functions are used to fit multiplets where the separate centroids can be identified. If this is not the case (and the separate contributions cannot be estimated by observing other $\gamma$ decays from the same nuclide), the peak is omitted from the results.
The resulting integrals are combined with the source information (activity, branching ratio) and data acquisition time to estimate the efficiency of that photon being detected. Note that single $\gamma$ emitting nuclides and sources of low activity are useful for this process, as this minimises losses due to cascade summing and accidental summing, respectively. For calibration sources where cascade summing cannot be avoided (such as $^{60}$Co), the detector–source distance was increased until the peak count plateaued.

Efficiency curves were then fitted to the data to describe the peak efficiency across the entire energy range. These were of the form described by equation [5.1], where $\text{Eff}$ is the efficiency, $E$ is the energy of the decay, and $c_n$ is a parameter dependent on the order of the equation (usually up to 5 terms were used). An example efficiency fit is shown in figure 5.6.

$$\frac{\text{Eff}}{E} = c_1 \ln(E) + c_2 \ln^2(E) + c_3 \ln^3(E) + \ldots + c_n \ln^n(E) \quad (5.1)$$

Figure 5.6: An efficiency fit to GEANT4 data for a HPGe detector. This particular function used a 7th order equation of the form described above, and was fitted using GNUPlot[65].
5.2 Acquisition Electronics

All data is collected using a common acquisition process, which involves routing the detector output (directly for semi–conductors, and from Photo–Multiplier Tubes (PMT’s) for scintillators) through a preamplifier to increase the voltage to the required range for the subsequent electronics. These consisted of four distinct systems; a series of NIM units for amplification, shaping and collection of the pulse, and three digital ‘all–in–one’ systems. These are an OSPREY™ Digital MCA Tube Base from Canberra Industries (Meriden, US), a LYNX™ Digital Signal Analyzer from Canberra Industries (Meriden, US), and a CAEN Digital Multi Channel Analyzer (CAEN S.p.A., Italy). The various systems are shown below, in figure 5.7.

![Figure 5.7: The different data acquisition systems used and evaluated within this work. From left to right: the CAEN Digital MCA, a LYNX™ Digital Signal Analyzer, an OSPREY™ Digital MCA Tune Base, and a series of NIM units. These devices increase in complexity and functionality from right to left, however require increased programming and data processing knowledge to extract such performance.]

5.2.1 Detector Dead Time

For low activity samples, dead time is not an obvious concern. Percentage losses of up to two percent are common, and easily accounted for when considering the total acquisition time (and resulting radionuclide abundances of a sample). Where resolution is a critical factor however, increased shaping times (which ensure the full signal is collected and minimise ballistic deficit) can drive up the dead time to unacceptable levels. This becomes a partic-
ularly acute problem when large crystal sizes are involved, and especially so in Compton suppression systems, where dead time losses can result in missed coincident events. A balance must therefore be struck, where the resolution is sufficient to resolve any radionuclides of interest, while the dead time of the detector is minimised to increase throughput. For guard detectors used in Compton Suppression systems, minimal shaping was used for the pulses, resulting in a slightly degraded resolution but a highly effective veto detector.

5.2.2 Ballistic Deficit

Ballistic deficit refers to the loss in signal amplitude that can occur as a result of signal shaping times that are significantly shorter than the preamplifier pulse rise time. Where charge–collection times are fairly constant, ballistic deficits can be tolerated as each signal amplitude will be attenuated by a similar percentage. Where this is not the case however, (such as in large coaxial HPGe crystals), the amount of signal lost due to short shaping times will vary slightly, degrading detector resolution. This problem can be alleviated by using a trapezoidal shaping filter, which has a flat top. If this ‘flat–top’ is longer than the variation in the rise time of the preamplifier pulse, ballistic deficit can be avoided. Shaping times were therefore carefully optimised to maximise both the resolution and throughput of the detector.

5.2.3 Pole–Zero Correction

Pole–zero correction was performed using inbuilt circuitry within the acquisition electronics. This involves using an attenuated input pulse (often created by adding a resistor in parallel to the capacitor within the shaping circuit) to correct for the undershoot of the pulse that arises due to the decay time of the preamplifier pulse. The correction required is dependent on the preamplifiers decay time, and is set with an automatic routine run with a characteristic source in place (to provide representative preamplifier signals).
Chapter 6

Data Processing

6.1 Acquisition & Storage

This project is concerned with the use of Compton Suppression systems, and therefore coincidence measurements between different detectors. Such data is normally collected using analogue electronics, with coincident signals vetoed (discarded) to reduce the contribution from the Compton continuum. Spectra can then be collected into histograms using a Multi–Channel Analyser (MCA), with appropriate binning (typically 8192 channels) to cover the required energy range and fully utilise the resolution of the detector. This results in very small (∼kB) files, which contain the total counts seen in each detector channel. These can then be calibrated/analysed (either using commercially available software or analysis suites developed in–house) to create a Compton suppressed energy spectrum and extract the required information. This process, however, discards all data from coincident events, all timing data from the events that are collected, and relies on the initial delay gate (which is used to accept/reject coincident events) being accurately calibrated. Furthermore, once a delay gate is set, it cannot be changed for that experiment, resulting in the loss of all additional coincidence information.

An alternative method that does not discard events is to collect the data in ‘List–mode’. This synchronises the data collection units, and time-stamps the events in each detector for later analysis[15]. The independent recording of each event allows the data to be re–sorted numerous times, and therefore for
multiple coincidence signals to be extracted. The evolution of peaks (during the data acquisition) can also be observed, as well as coincidence backgrounds (which result from random coincidences between the detectors) estimated and reduced.

An acute problem with ‘List-mode’ data acquisition is the size and subsequent processing of the data files, which require up to $\sim 0.6$ GB of storage space per detector per hour when observing a standard 37 kBq source at 0 cm detector–source separation. Not only does storage become a concern, but this data must then be post-processed to both sort and extract the relevant coincidence information. Only once this is complete can energy spectra be constructed and then analysed in the traditional fashion. At the start of this project, processing 2 GB of coincidence data required up to half a day, and 6–10 times the storage space of the original data. Through several advances developed as a result of this project, the same amount of data now takes $\sim 1$ minute to process, and requires less than a tenth of the storage space used by the raw (unprocessed) data.

6.2 Data Formats

Initially, all List–mode data were acquired in an ASCII format, which is a human readable text format. Standard List–mode data were written out from the acquisition system as a string of four comma separated values, which recorded the channel number, clock time, real time in seconds, and live time in seconds. A separate file was written for each detector, with a two–line header holding the IP address of the detector and the time/date that the acquisition was started. During testing and operation of the post-processor (see below) it was found that the use of ASCII formats significantly slowed the sorting process. A binary format was therefore developed, along with a converter for historical data.

Binary formats encode information as a series of bytes, and are therefore not human readable. Due to the sequential and known values of each byte, however, significant performance improvements can be gained when compared
to an ASCII format (where the program has to interpret numbers from a text file). The information required from the detectors is the channel number, clock time and live time (the clock time is equivalent to the real time, and ‘roll–over’ – where the clock resets – is eliminated by outputting the total clock time since the start of the measurement from the acquisition software).

The format developed for this research encodes a 512 byte header, which includes the IP address, acquisition start time, and formatting information for the data that follows. The header itself is encoded as a character string, so that it can be easily inspected (either using existing command–line tools or with customised software) to extract the data formatting information. The format of the data depends on the information encoded, however a typical file will contain repeating 20–byte blocks (4–bytes for the channel number, and 8–bytes each for the clock time and live time respectively). By moving to a binary format for all acquisition, sorting, and binning of the data (using ROOT), processing speeds were increased by a factor of 8–14.

6.3 Post-Processing & Analysis

The LIst-Mode Processor (LIMP) provides the same functionality as that seen in typical Compton Suppression systems\cite{14, 49} (the ability to veto events based on their timing), although with much more flexibility. In traditional systems, coincident events are vetoed in real time using electronics, and all information about these events discarded. By recording all the events and post-processing them, the data can be searched as many times as necessary to extract far more coincidence information than is typically available.

LIMP is written in c++, and re-processes list-mode data files into binned spectra in both ASCII and ROOT formats. The input data can be in either ASCII or binary formats, as it includes a conversion module that can process 1 GB of ASCII data per computing core in $\sim 30$ seconds (tested on an Intel Core$^\text{TM}$ i7-2600 CPU @ 3.40GHz). Input formats can be modified here (or additional formats defined) allowing it to be used for a wide range of current and future datasets.
6.3. Post-Processing & Analysis

Figure 6.1: Inset: An example coincidence time-distribution for two detectors using a combined $^{155}$Eu, $^{22}$Na ($\beta^+$) source, with the time window set from -8 to +8 $\mu$s. There are 3 clearly distinct regions, the prompt coincidences between -250 and 250 ns, delayed coincidences from (±) 600 to 1200 ns, and the background (random) coincidences.

Main: The coincidence matrix for the aforementioned source, with the time window on the ‘prompt’ -250 to 250 ns peak. The $^{22}$Na (511 keV) decays are clearly seen in coincidence due to the annihilation of the $\beta^+$, while the $^{155}$Eu decays (45.3, 60.0, 86.6, 105.3 keV) are not.

Coincidence searching can be performed on multiple detectors, which extracts all coincident events within a user-defined time window. The time window is defined with respect to one detector, which is used as a base ($t_0$) for the coincidence search. Any events in additional detectors that are within this time window are extracted to a coincidence matrix (for two detectors, coincident events would be recorded into a 3D (energy, energy, time) matrix). Events where no coincidences are found are recorded into an anti–coincidence spectrum. The coincidence searching routine is extremely efficient (over an order of magnitude quicker than the existing processor), and is scalable to large datasets (currently tested up to 100 GB on a Intel Core™ i7-2600 3.40GHz system with 8 GB of RAM). The processor can also run on multiple cores, after which the analysis can be undertaken in ROOT or any other suitable software package.

The ability to gate on specific time and energy windows allows the user to ‘scan’ the coincidence space (as in figure 6.1 inset), and then select regions for further investigation (figure 6.1 main), projecting spectra after applying
time and/or energy gates. It was originally developed to suppress Compton scattered events (which have a characteristic scattering time between detectors), although it can process any dataset that contains events in coincidence (and the appropriate temporal information).
Chapter 7

GEANT4 Validation Studies

7.1 NaI(Tl) Detectors

7.1.1 Scope

This preliminary project had two main parts. Firstly, it involved developing and testing the post-processing program for use with coincidence measurements, and secondly, to create Monte-Carlo simulations that could be validated against detectors in the laboratory. The results from this project were published in the Journal of Radioanalytical and Nuclear Chemistry, and a copy of this can be found in Appendix E.

7.1.2 Equipment & Method

The NaI(Tl) detectors and associated electronics are supplied by Canberra UK (Harwell, Oxfordshire). Each detector has a cylindrical crystal of dimensions \( \theta = 76.2 \text{ mm}, \ l = 76.2 \text{ mm} \) (model 802-3x3), and the pre–amplifier and MCA are combined into a single unit that mounts onto the back of the detector body (model Osprey\textsuperscript{TM}). This also controlled all further amplification and digitisation of the pulse, saving both space and expense as separate NIM bins are not necessary. The detectors are partially enclosed in 40-60 mm of lead shielding, which served to reduce the number of background counts by 54% and also act as a support. For the purposes of this work the detectors were set up in a ‘back-to-back’ configuration (both looking directly at the source and
7.1. NaI(Tl) Detectors

separated by 180°). A simulated image of the experimental setup is shown below, in figure 7.1.

Figure 7.1: A GEANT4 simulated geometry of the experimental setup. The yellow cylinder is the source, the red cylinders the detector caps, the grey cylinders the detector assemblies, and the grey (transparent) boxes are the lead shielding. The Osprey units are not shown here, but would mount onto the back of the detector assemblies.

7.1.3 Model of the Detector

This used a simpler version of the simulation discussed in chapter 4 (the main difference being the lack of a decay library), and was built using version 9.4.p01 of the GEANT4 toolkit. The description of the detector geometry consisted of the crystal itself, the optical coupler between the crystal and the Photo–Multiplier Tube (PMT), and the Aluminium canister that surrounded these components. These were accurately represented, while everything that sat behind the optical coupler was approximated with an Aluminium cylinder to save computing and development time (this was found to have no measur-
able adverse effects on the spectra produced). The lead shielding and source housings were also reproduced as accurately as possible.

The Aluminium outer canister is of nominal thickness 1.5 mm, although the thickness of the front face was tuned to match the experimental data. The distance between the front face of the canister and the crystal was initially set to 2 mm, and also optimised.

### 7.1.4 NaI(Tl) Results

A typical spectrum and background are shown in figure 7.2. The background consists primarily of 75 keV X-rays from lead, and the 1461 and 2614 keV peaks from $^{40}$K and $^{208}$Tl respectively. Data were collected for long enough to provide sufficient statistics for comparison with the simulated spectrum, while minimising the simulation time required. These counts ranged from 300-1200 s in both a ‘close’ and ‘extended’ geometry (50 mm and 150 mm source-detector separations).

![Figure 7.2: An example energy spectrum for a $^{137}$Cs source irradiating a single 3x3\" NaI(Tl) detector. Lead shielding slightly increased the background at low energies (due to 75 keV X-rays), however reduced the overall number of background counts by 54%. The background only makes a substantial contribution at energies greater than the main photopeak energy (at 661.7 keV), where statistics for summing effects are considerably lower.](image)

All sources were also simulated, and the efficiencies for both experimental and simulated data are presented in figure 7.3. Note that due to the resolution
of NaI(Tl) (6-7%), some low energy and closely packed peaks could not be resolved, and were also obscured by X-ray fluorescence from the shielding. Where multiplets could be resolved, these were fitted with overlapping Gaussians accordingly, otherwise, they were omitted from the efficiency calculations.

![Graph showing detector efficiency as a function of energy](image)

**Figure 7.3:** The detector (peak) efficiency as a function of energy for experimental and simulated data. In the simulated data, the only substantial errors are due to the uncertainty in the number of counts in a peak (plus the standard error), whereas the experimental data also contained errors due to both the source positioning and activity. All efficiencies are for a 50 mm source–detector separation.

The simulated and experimental peak efficiencies were compared using a statistical z-test\[66\] for each energy point. These were then averaged for the entire dataset, resulting in a maximum error margin of 9% at a 93.0% confidence level. This is valid from $32.2 \leq E \leq 2505.7$ keV, and is adequate given the small sample size. A comparison between simulation and experiment for the extended geometry is shown in figure 7.4 below.

Many aspects of GEANT4 were tested and optimised for future work, including event generation, treatment of summing effects and the physics models used. The simulation output was also tailored to allow compatibility with the experimental post-processor, which proved to be both highly efficient and flexible, with data compression of up to 12 times, and a coincidence searching routine that is both a order of magnitude faster than the existing processor and scalable to very large datasets. This allows much more information to be
7.2. BEGe Detector

7.2.1 Scope

The Broad Energy Germanium (BEGe) project extended the GEANT4 simulations that were developed for the NaI(Tl) crystals, adding a decay library and advanced geometry features such as assembly volumes and boolean unions. Due to the comparative complexity of High Purity Germanium detectors, these modifications were necessary to accurately reproduce the detector response. Once optimised, the simulation was used to generate an efficiency for a distributed NORM source. This was then used to calculate the abundances of any radionuclides present, and compared to results that were obtained using proprietary software that had been previously validated. The results from this project were published in the Journal of Radioanalytical and
Nuclear Chemistry, and a copy of this can be found in Appendix E.

7.2.2 Equipment & Method

The BEGe detector \(^{67}\) (model BE3825) and associated electronics are supplied by Canberra UK (Harwell, Oxfordshire). The detector has an efficiency of 28% relative to a standard 3x3 NaI(Tl) crystal at 1.33 MeV, and a carbon epoxy window to minimise the attenuation of photons at low energies. The crystal is also relatively thin when compared to a standard coaxial design, minimising the contribution from higher energy decays (and therefore improving low energy efficiency). The cryostat (model 7500SL-RDC-6-ULB) is specifically designed to minimise background by using low-background components and offsets so that more active parts can be effectively shielded. A preamplifier (model 2002C SL) processes the initial signal data. All detector dimensions are taken from the manufacturer provided documents, and these are summarised in table 8.1.

The preamplifier output is sent to a LYNX\textsuperscript{TM} digital signal processing unit from Canberra, which controls all further amplification, pole-zero correction and digitisation of the pulse. Data were collected in ‘list mode’, and the post-processor used for analysis (without the coincidence functions).

<table>
<thead>
<tr>
<th>Component</th>
<th>Dimension (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ge crystal diameter</td>
<td>71.0</td>
</tr>
<tr>
<td>Ge crystal length</td>
<td>26.5</td>
</tr>
<tr>
<td>Aluminium endcap thickness</td>
<td>1.5</td>
</tr>
<tr>
<td>Endcap window thickness</td>
<td>0.5</td>
</tr>
<tr>
<td>Top dead layer</td>
<td>0.0004</td>
</tr>
<tr>
<td>Side dead layer (each side)</td>
<td>0.6</td>
</tr>
<tr>
<td>Crystal distance from outside</td>
<td>5.0</td>
</tr>
</tbody>
</table>

Table 7.1: A summary of the BEGe (model 3825) detector dimensions from Canberra. These were used as initial values for the simulations, before being ‘tuned’ to reproduce the detector response.

The detector is surrounded with a 105 mm thick graded lead shield, with liners of tin and copper to reduce fluorescence from lead x-rays. This created a low background environment for the BEGe detector, with an ambient
background of 2 counts per second (cps). A simulated image of the detector (including the lead cave, but excluding some internal detail of the detector due to graphics limitations) is shown below in figure 7.15.

Figure 7.5: A simulated cross-section of the experimental setup. The detector is shown in the centre, with an example source in an extended geometry above (the small yellow cylinder). The surrounding lead shield and liners are also shown, with grey corresponding to lead, silver to tin and a deep orange for copper. Some detail is missing from the electrodes in this representation, and the dead layers in the Germanium crystal are not shown (this detail is present in all simulations).

7.2.3 Model of the Detector

This encapsulated all of the advanced features discussed in chapter 4 and was built using version 9.4.p01 of the GEANT4 toolkit. The description of the detector geometry consists of the crystal itself, the outer and inner electrodes, the aluminium canister and carbon epoxy window. The cold finger and vacuum spaces within the detector were also included, and to complete the model, the tin and copper lined lead shielding and source housings were recreated. NIST compounds were used where available, and all components were reproduced as accurately as possible according to manufacturer specifications.
The shape of the crystal is well known, as it is a simple cylinder with flat, non-bulletised edges, however the exact dimensions of the rear electrode are not. The dead layer in the front face is very thin (≈0.4 µm), and predominantly affects the low energy part of the spectrum. The side dead layers are somewhat larger (≈600 µm), and of importance as this project also considers unusual geometries and side mounted sources (such as Marinelli beakers).

7.2.4 BEGe Results

A comparison between experimental and simulated data for a complex γ source is shown in figures 7.6 & 7.7. Data were collected for ≈20 minutes, and experiments were carried out with multiple sources (in several geometries) to validate the model.

When tuning the model, a range of rear electrode parameters were tested (assuming it was cylindrical in form). A larger rear contact would act to reduce the efficiency of the higher energy decays slightly, while not affecting the lower energy part of the spectrum. No significant change could be found, however, and the variation produced was insignificant when compared to the errors from the positioning and definition of the sources (typically ±5%). This is assumed to be because it is not a ‘critical’ component (i.e between the source and the crystal), and therefore has a minimal effect on the detector response. The distance from the carbon epoxy window to the crystal was revised down to 4.7 mm, and the dead layers remained unchanged from the manufacturers specifications.

The simulated and experimental peak efficiencies were compared using a statistical Z-test [66], which yields a maximum error between experiment and simulation of 3% with a 95% confidence level for the standard geometry. The only significant discrepancies are at low energy, and are due to the sensitivity of the detector response to small geometrical errors. The side mounted geometry has far greater attenuation between the source and crystal, reducing the detectors sensitivity to low energy decays. The simulated data matches this extremely well, with a maximum error across the energy range in the side mounted geometry of less than 1%. 
Figure 7.6: Simulated (red) and experimental (black) data for a complex $\gamma$ source. Data were taken 5 mm from the top of the detector and collected for twenty minutes. The top image shows the spectrum collected and simulated, while the bottom shows a peak efficiency plot for the same geometry.

Once these levels of accuracy were achieved, a NORM sample of granite was simulated to determine the efficiency for a range of energies. The size, shape, chemical composition and density of both the granite and its container were reproduced, and photons from 30 keV to 3000 keV were fired from a random position within the source. The results of this efficiency calculation, which includes self-attenuation of the source, are shown in figure 7.8.

The sample was measured for 24 hours, and analysed using LIMP. The isotopes identified and their abundances are recorded in table 7.2.

Most radionuclides were identified using multiple peaks, and only $^{40}$K,
210Pb, 224Ra, 226Ra, and 228Th were seen as single emitters (increasing the errors in these abundances substantially). The radionuclides within the sample appear to be in secular equilibrium, as the majority of the identified isotopes in each decay chain have around the same activity (the only exception is 208Tl, however this is due to the 232Th decay chain splitting at 212Bi, with only 36 % of the decays going to 208Tl). The population of 238U was therefore inferred from the daughter decay of 234Th, giving a $^{235}\text{U}/^{238}\text{U}$ abundance of $1.07 \pm 0.36 \%$, which is in fair agreement with the natural abundance of 0.72 %. The abundances also agree with a prior analysis using proprietary software,
Figure 7.8: Peak efficiency for an encapsulated granite source, for randomly (and evenly) distributed decays as a function of energy. The chemical composition of granite was taken from reference [68], and errors are due mainly to the uncertainties in this composition.

Figure 7.9: Energy spectrum acquired from the granite NORM source. Data were collected for 24 hours with the source positioned at the top of the detector.
Table 7.2: Isotopic abundances from a granite NORM sample measured for 24 hours, using a GEANT4 based efficiency characterisation. A '*' denotes an isotope that was not be measured directly, but inferred from the daughter decay populations.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Decay chain</th>
<th>Abundance (PPM)</th>
<th>Activity (Bq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{228}\text{Ac}$</td>
<td>$^{232}\text{Th}$</td>
<td>$7.61 \pm 0.03 \times 10^{-13}$</td>
<td>$20.4 \pm 0.4$</td>
</tr>
<tr>
<td>$^{212}\text{Bi}$</td>
<td>$^{232}\text{Th}$</td>
<td>$1.2 \pm 0.2 \times 10^{-13}$</td>
<td>$22 \pm 4$</td>
</tr>
<tr>
<td>$^{214}\text{Bi}$</td>
<td>$^{238}\text{U}$</td>
<td>$8.88 \pm 0.03 \times 10^{-14}$</td>
<td>$47 \pm 3$</td>
</tr>
<tr>
<td>$^{40}\text{K}$</td>
<td>-</td>
<td>$4.2 \pm 0.6 \times 10^{0}$</td>
<td>$358 \pm 49$</td>
</tr>
<tr>
<td>$^{210}\text{Pb}$</td>
<td>$^{238}\text{U}$</td>
<td>$4.5 \pm 0.7 \times 10^{-8}$</td>
<td>$41 \pm 6$</td>
</tr>
<tr>
<td>$^{212}\text{Pb}$</td>
<td>$^{232}\text{Th}$</td>
<td>$1.2 \pm 0.3 \times 10^{-12}$</td>
<td>$19 \pm 3$</td>
</tr>
<tr>
<td>$^{214}\text{Pb}$</td>
<td>$^{238}\text{U}$</td>
<td>$1.08 \pm 0.01 \times 10^{-13}$</td>
<td>$42.4 \pm 1.6$</td>
</tr>
<tr>
<td>$^{224}\text{Ra}$</td>
<td>$^{232}\text{Th}$</td>
<td>$1.280 \pm 0.002 \times 10^{-11}$</td>
<td>$25 \pm 4$</td>
</tr>
<tr>
<td>$^{226}\text{Ra}$</td>
<td>$^{238}\text{U}$</td>
<td>$3.7 \pm 0.5 \times 10^{-6}$</td>
<td>$44 \pm 6$</td>
</tr>
<tr>
<td>$^{228}\text{Th}$</td>
<td>$^{232}\text{Th}$</td>
<td>$2.9 \pm 0.8 \times 10^{-9}$</td>
<td>$28 \pm 8$</td>
</tr>
<tr>
<td>$^{234}\text{Th}$</td>
<td>$^{238}\text{U}$</td>
<td>$1.720 \pm 0.006 \times 10^{-10}$</td>
<td>$47 \pm 2$</td>
</tr>
<tr>
<td>$^{208}\text{Tl}$</td>
<td>$^{232}\text{Th}$</td>
<td>$2.23 \pm 0.03 \times 10^{-15}$</td>
<td>$7.9 \pm 0.6$</td>
</tr>
<tr>
<td>$^{235}\text{U}$</td>
<td>$^{235}\text{U}$</td>
<td>$1.3 \pm 0.5 \times 10^{-1}$</td>
<td>$3.3 \pm 1.2$</td>
</tr>
<tr>
<td>$^{238}\text{U}^*$</td>
<td>$^{238}\text{U}$</td>
<td>$1.2 \pm 0.5 \times 10^{1}$</td>
<td>$47 \pm 2$</td>
</tr>
</tbody>
</table>

further validating the GEANT4 efficiency calculation that was used for the abundance calculations.

7.2.5 Summary

Broad-energy HPGe spectra for single and complex sources were simulated using the GEANT4 Monte Carlo toolkit, with the geometry parameters optimised to obtain detection efficiencies within 3% of those determined from the experimental data, with a 95% confidence level. Multiple geometries have been explored, characterising the response of the detector for sources in standard and non-standard configurations, with no loss of accuracy. Small deviations were found at low energies (<100 keV) in the standard (detector top) source geometry, and are due to the limited accuracy of the detector reproduction.

A granite NORM sample has also been analysed using a GEANT4 based efficiency, allowing abundance estimates to be calculated for a range of radionuclides in the sample. Where multiple peaks could be identified for a single radionuclide, the abundances calculated across the energy range showed no sign of systematic error (that would arise if the efficiency calibration had
been wrong), and the abundances also agree with a prior analysis using proprietary software. The simulation allows much more flexibility than the current proprietary software, and this validated model can now be used to characterise the detector response for a variety of complex geometries and source configurations.

7.3 Cascade Summing Corrections

7.3.1 Scope

Cascade summing occurs where an unstable nuclei emits multiple photons or x-rays in coincidence/cascade. If multiple photons deposit their energy in the detector crystal, a single output pulse will be generated as the charge collection times are typically much greater than the corresponding nuclear lifetimes. These events will therefore be interpreted as a full energy peak with the energy equal to the sum of the cascade photons. This is known as ‘summing out’, and reduces the apparent efficiency of the detector to that nuclide. It is also possible for multiple photons to sum to an equivalent energy of the photon of interest, increasing the apparent efficiency of the detector, and is known as ‘summing in’. Such correction factors can be estimated empirically [69], however they will change for each new geometry and source counted. It is therefore more efficient to calculate the correction factors numerically [70, 71, 72], or use Monte Carlo simulations to determine the appropriate correction factors [73, 74]. The latter method also allows for additional complexity in the source matrix that may be difficult to achieve numerically.

This project builds upon the previous BEGe simulations (section 7.2), and aimed to use the GEANT4 detector model to calculate correction factors for radionuclides that are sensitive to cascade summing effects. By comparing simulated results using the full GEANT4 gamma libraries to those using an efficient (user-defined) decay library, the cascade summing correction factors can be determined, and compared to experimental data and an alternative (previously validated) calculation method. Due to the sensitivity of Cascade correction factors on the source and detector geometry, such a calculation
using GEANT4 provides a robust test of the simulations validity. The results from this project were published in the Journal of Radioanalytical and Nuclear Chemistry, and a copy of this can be found in Appendix E.

7.3.2 GEANT4 simulation

GEANT4 models during this work were built using version 9.5 of the toolkit. The description of the detector geometry consists of the BEGe detector, associated shielding and the source, and used the models developed in section 7.2, with the decay libraries and primary particle generators extended to account for coincident radiation. To efficiently simulate (and subsequently analyse) the environmental sources, many of the automation and analysis routines developed for LIMP were written during this work.

Cascade summing is simulated using the GEANT4 radioactive decay data libraries. A radioactive isotope is disintegrated per event, with the decay radiation selected based upon the relative branching ratios for that nuclei. Multiple decay modes are possible, including situations where multiple photons are emitted in cascade. Radiation from these decays is propagated throughout the simulated volume, and any energy deposited within the active detector crystal recorded. These are summed on an event by event basis, and so reproduce the cascade summing seen in the laboratory. Additional simulations were also completed using a user-defined gamma library. This approximates each source as a single particle emitter, and generates one photon per event. Branching ratios (if greater than $10^{-9}$) are obtained using the NuDat2 tool\cite{59}, and energy deposited in the detector was calculated as before.

In both modes, complex sources could be created by selecting multiple radioactive nuclides and giving each a relative weighting. Summed events that are due to the activity of the source are also simulated as this is based upon a separate programming loop that can generate multiple decays per event.
7.3.3 Experimental Method

The BEGe detector (model BE3825) is also the same as that used in section 7.2. Apparent peak efficiencies and isotopic abundances were calculated as before, and all data were analysed using the post-processor developed.

Calibration sources were chosen to cover the 10-3000 keV energy range, and included NIST (National Institute of Standards and Technology) traceable complex $\gamma$ sources. The isotopes that comprised these were $^{241}$Am (59.54 keV), $^{109}$Cd (88.03 keV), $^{57}$Co (122.06 keV), $^{139}$Ce (165.86 keV), $^{113}$Sn (391.68 keV), $^{137}$Cs (661.67 keV), $^{54}$Mn (834.84 keV), $^{88}$Y (898.04 and 1836.06 keV), $^{65}$Zn (1115.54 keV), and $^{60}$Co (1173.23 and 1332.49 keV).

Two complex $\gamma$ sources were used to validate the cascade summing corrections; a cylindrical compressed air filter (type half-RASA), and a 15-layer reference filter pack (type CINDERELLA). Both are used for the operation of the IMS (International Monitoring System), which undertakes radionuclide monitoring for verification of the CTBT (Comprehensive Nuclear-Test-Ban Treaty). The half-RASA (Radionuclide Aerosol Sampler/Analyser) source measured approximately 72 mm diameter by 18 mm thickness, and is from IMS station JPP38. This was exposed to some of the environmental radiation from the Fukushima incident (March 2011), and as such contains a high proportion of $^{134}$Cs, (a nuclide that requires substantial cascade summing corrections). The second source is a CINDERELLA type, and measures approximately 83 mm diameter by 8 mm thickness. The CINDERELLA source contains multiple radionuclides, the activities of which were accurately characterised at NPL (National Physics Laboratory). The performance and behaviour of these source types is well understood, and they therefore provide an excellent test of both the GEANT4 efficiency characterisation and the cascade summing corrections.

7.3.4 Calculating the correction factors

Cascade summing is dependent on both the decay of the radionuclides, and the geometry of the detection system. The correction factors are therefore
different for each variation in source matrix and position, and only comparable in specific geometries. The cascade summing sources have been previously analysed, and the concentrations of the radionuclides present are well known. The correction factors and efficiency characterisations used in the previous analysis were calculated using the Geometry Composer module in Canberra’s GENIE™ 2000 software (version 3.6). This had been previously validated, and is used as the basis for comparison with the GEANT4 calculated values (as well as the NPL provided activities for the CINDERELLA source).

Simulations were performed using the full GEANT4 decay libraries, and repeated using the user-defined gamma libraries. The total number of events was calculated such that the total number of photons from each source was the same, however in the latter case these were all emitted individually, with no coincidence data (see figure 7.10). The full energy photopeaks can then be counted in each simulation, and the ratio of the peak areas used to determine the correction factors appropriate for the specific detector, source, and experimental geometry used.

Figure 7.10: An example decay scheme (\(^{134}\)Cs) used in the Cascade summing correction calculations. The user-defined source uses the gamma energies from the transitions in the (excited) \(^{134}\)Ba daughter nucleus, and branching ratios from the NuDat2 database \([59]\). No coincidence data is included in this library, allowing direct comparison with the full GEANT4 decay libraries, where such information is included.
7.3.5 Results & Discussion

7.3.5.1 Efficiency of the Sources

To allow a comparison between the radionuclide abundances in both samples previous and current analyses, the peak efficiency of the detection system must be determined. As the GEANT4 model of the detector has been validated and is accurate to within 3%, simulations were run using photons of energy from 10-2500 keV. Both sources were reproduced according to their size, chemical composition and density, and the photons fired from randomised coordinates within this volume. The determined efficiencies are shown in figure 7.11 below. All errors are quoted at the 1 sigma significance level.

![Figure 7.11: The BEGe detector peak efficiencies as a function of energy for both the half-RASA and CINDERELLA sources. The error in these efficiency calculations is ±5%, which includes both the error in the detector model and the error in the composition of the source.](image)

7.3.5.2 Calculated Correction Factors

An example simulated spectrum using the full GEANT4 libraries is shown below, in figure 7.12. Corresponding simulations were also performed using data-files that contained no coincidence information, and the calculated correction factors for the main \( \gamma \) decays in the half-RASA source are summarised in table 7.3. A full list of the calculated correction factors for the CINDERELLA source is available in Appendix C.
Figure 7.12: The energy spectrum obtained from the simulation using the full GEANT4 decay library, with a pure $^{134}$Cs source in the half–RASA geometry. The events were generated at a randomised coordinate within the source geometry, and sum peaks where two or more photons have ‘summed out’ can be clearly seen.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy (keV)</th>
<th>Cascade Correction Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{134}$Cs</td>
<td>475.37</td>
<td>0.728</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>563.25</td>
<td>0.686</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>569.33</td>
<td>0.692</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>604.72</td>
<td>0.797</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>795.86</td>
<td>0.786</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>801.95</td>
<td>0.742</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>1038.61</td>
<td>0.955</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>1167.97</td>
<td>1.207</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>1365.19</td>
<td>1.326</td>
</tr>
</tbody>
</table>

Table 7.3: A summary of the correction factors calculated for the half–RASA source. Some correction factors were not calculated in the Canberra software due to those peaks not being present in the preliminary analysis. Errors in the calculated correction factors are ± 6%, and were dominated by the variability in the source geometry/composition.

For the half-RASA source, the results show excellent agreement between the calculated correction factors and those determined using Canberra’s GENIE™ 2000 software, with a average deviation of 3% and a maximum of 6%. Similar agreement is also found in the CINDERELLA geometry. For $^{140}$Ba, $^{134}$Cs, $^{140}$La, $^{147}$Nd and $^{103}$Ru the two calculation methodologies had an average de-
viation of 4%, and a maximum of 8%. Reasonable agreement was found for \(^{99}\text{Mo}\), \(^{132}\text{I}\), and \(^{132}\text{Te}\) with an average deviation of 8% between the GEANT4 and GENIE\textsuperscript{TM} 2000 correction factors.

Overall, the two approaches yield values that are in close agreement for a variety of radionuclides. As they use completely different methods, this confirms both the validity of the GEANT4 model and the approach used to determine the correction factors. It is worth noting that the errors in this calculation are fairly large, as true coincidence summing is highly sensitive on the geometry of both the detector and the source (as well as their relative positions).

### 7.3.5.3 Analysis of Realistic Sources

Analysis was completed for both the half-RASA and CINDERELLA sources. Figure 7.13 shows the complex gamma spectrum obtained from the half-RASA source, while figure 7.14 shows the spectrum for the CINDERELLA source. For both samples, radioisotopes were identified using their characteristic energies and a weighted mean taken if the isotope emitted multiple photons. The radioisotopes detected in the half-RASA source and their (cascade corrected) abundances are summarised in table 7.4, alongside the accepted values for this source (corrected for radioactive decay between the previous analysis and the latest measurement). The same comparison between the CINDERELLA source and the NPL calibration values is shown in table 7.5.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Accepted Activity (Bq/m(^3))</th>
<th>Calculated Activity (Bq/m(^3))</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{7}\text{Be})</td>
<td>(4.10 \pm 0.25 \times 10^{-3})</td>
<td>(3.8 \pm 0.4 \times 10^{-3})</td>
</tr>
<tr>
<td>(^{134}\text{Cs})</td>
<td>(1.05 \pm 0.02 \times 10^{-4})</td>
<td>(1.04 \pm 0.05 \times 10^{-4})</td>
</tr>
<tr>
<td>(^{137}\text{Cs})</td>
<td>(1.44 \pm 0.04 \times 10^{-4})</td>
<td>(1.39 \pm 0.07 \times 10^{-4})</td>
</tr>
<tr>
<td>(^{210}\text{Pb})</td>
<td>(4.6 \pm 0.5 \times 10^{-4})</td>
<td>(4.4 \pm 0.4 \times 10^{-4})</td>
</tr>
</tbody>
</table>

Table 7.4: A summary of the radioisotopes detected in the CTBT half-RASA source, and their abundances. The ‘calculated activity’ is from the present study, and was determined solely using Monte-Carlo methods (GEANT4). The ‘accepted activity’ is from a previous study, and is time corrected to account for the radioactive decay between the collection of the two datasets.
Figure 7.13: The energy spectrum obtained from the half-RASA source. This was placed upon the detector (with a sheet of polyethylene film between the two to prevent contamination), and counted for 7 days.

Figure 7.14: The energy spectrum obtained from the CINDERELLA source. This was placed upon the detector (with a sheet of polyethylene film between the two to prevent contamination), and counted for 7 hours.
<table>
<thead>
<tr>
<th>Isotope</th>
<th>NPL Calibration value (Bq/m³)</th>
<th>Calculated Activity (Bq/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{140}$Ba</td>
<td>$4.77 \pm 0.10 \times 10^{-3}$</td>
<td>$4.43 \pm 0.26 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{141}$Ce</td>
<td>$3.13 \pm 0.06 \times 10^{-3}$</td>
<td>$3.05 \pm 0.30 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{144}$Ce</td>
<td>$4.97 \pm 0.15 \times 10^{-4}$</td>
<td>$4.84 \pm 0.47 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>$1.383 \pm 0.027 \times 10^{-3}$</td>
<td>$1.28 \pm 0.16 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>$2.22 \pm 0.07 \times 10^{-5}$</td>
<td>$2.23 \pm 0.31 \times 10^{-5}$</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>$0.87 \pm 0.03 \times 10^{-3}$</td>
<td>$0.84 \pm 0.15 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{99}$Mo</td>
<td>$3.81 \pm 0.12 \times 10^{-4}$</td>
<td>$4.24 \pm 0.38 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{95}$Nb</td>
<td>$8.23 \pm 0.61 \times 10^{-4}$</td>
<td>$7.4 \pm 0.4 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{147}$Nd</td>
<td>$1.48 \pm 0.10 \times 10^{-3}$</td>
<td>$1.52 \pm 0.07 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{103}$Ru</td>
<td>$7.72 \pm 0.17 \times 10^{-4}$</td>
<td>$7.48 \pm 0.41 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{132}$Te</td>
<td>$4.67 \pm 0.14 \times 10^{-4}$</td>
<td>$4.89 \pm 0.26 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{95}$Zr</td>
<td>$2.476 \pm 0.048 \times 10^{-3}$</td>
<td>$2.53 \pm 0.10 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

Table 7.5: A summary of the radioisotopes detected in the CTBT CINDERELLA source, and their abundances. The calculated activity was again determined solely via Monte-Carlo methods. This is compared to the (time-corrected) activity values determined during the initial calibration at NPL (National Physical Laboratory). Nuclide activities were also corrected to account for the feeding of different isotopes from other decays (such as $^{140}$Ba, which decays into $^{140}$La).

In the half-RASA data, it is clear that there is a large background continuum from higher energy decays. This reduces the sensitivity slightly, however mBq detection levels are still attainable. The CINDERELLA spectrum was far more complex, and some energy peaks could not be resolved (increasing the errors in the activities of these radioisotopes). For both sources, the abundances determined are in good agreement with the previously reported values, suggesting that the sample efficiency and cascade correction factors are accurate. Where radioisotopes were identified using multiple peaks, good agreement was also seen in the calculated abundances across the energy range, which would not be the case if some systematic error had been present in the analysis process. A full list of the radionuclides detected in the CINDERELLA source is available in Appendix D.

7.3.6 Summary

The GEANT4 based Monte-Carlo simulation developed for a BEGe detector was successfully utilised to generate peak efficiency characterisations and mul-
tiple cascade summing corrections in two source geometries commonly used for environmental monitoring. The cascade summing corrections calculated were in excellent agreement for many radionuclides (within 10%) of values generated using an existing (validated) system (GENIE™ 2000). The calculated correction factors and peak efficiencies were also tested using actual half-RASA and CINDERELLA type sources, in which the abundances of the radioisotopes detected matched those previously determined.

The process of generating the efficiencies relies upon an accurate simulation of the detector system and source, and as such requires a working knowledge of GEANT4 and c++. The software from Canberra requires no such expertise, and is inherently more user-friendly. The Monte-Carlo method also requires more time to complete, unless an accurate simulation is already available (in this case the methods take a similar amount of time). A major benefit of using the GEANT4 method, however, is the complexity possible in both the detector model and the source matrix. This is required for applications involving multiple detector set-ups, such as Compton Suppression systems.

7.4 Low–Energy Coincidence Systems

7.4.1 Scope

This project utilised a Compton suppression system for the first time, and so provided an opportunity to gain experience with a fully operational coincidence system, test both the required experimental equipment and data processing software, and develop preliminary coincidence simulations using GEANT4. The coincidence system used a Low–Energy HPGe (LEGe) primary crystal with a NaI(Tl) annulus, and the results from this project were published in the Nuclear Instruments and Methods in Physics Research Section A (a copy of this can be found in Appendix E).
7.4.2 Experimental Setup

The LEGe detector (model GL0510) and associated electronics are supplied by Canberra UK (Harwell, Oxfordshire). The detector is optimised for low energy photons, and set up to cover a range of 10-700 keV (above this value the efficiency is $<0.5\%$). The detector also has a carbon epoxy window to minimise the attenuation of low energy photons, which allows $\sim82\%$ transmission at 10 keV [75]. The crystal is fairly small (an active diameter of 25.5 mm, and a thickness of 10.5 mm), minimising the contribution from higher energy decays. An ultra-low background cryostat (model 7915-30-ULB) is used, and a preamplifier (model I-TRP) processes the initial signal data.

The preamplifier output is sent to a LYNX™ digital signal processing unit from Canberra, which controls all further amplification, pole-zero correction and digitisation of the pulse. Data is again collected in ‘list mode’.

The NaI(Tl) shield detectors are from Scionix (Utrecht, Netherlands), and comprise of a cylindrical annulus with a removable plug that is positioned above the primary detector. Due to this geometry, the efficiency for suppressing Compton Scattered events is very high, however there may be a large number of additional coincidence signals due to $\gamma$ radiation emitted in cascade, and chance coincidences due to both the activity of the source and background radiation. Note that while the BEGe has a thin entrance window at the top, Compton scattered photons will typically exit the detector through the Aluminium casing, and may therefore be attenuated before entering the NaI(Tl) veto detector (through another thin layer of Steel).

The cylinder has five Photo-multiplier Tubes (PMT’s), and the plug has a separate PMT for efficient charge collection. The outputs are combined from all of the PMT’s, and passed to a preamplifier (model 2005). Again, the preamplifier output is sent to a LYNX™ unit, where the data is collected in ‘list-mode’. A synchronisation cable also runs between the two LYNX™ units to allow synchronisation of the clocks used to record the events in each detector.

The LEGe and NaI(Tl) combination is placed within a 80 mm thick lead shield, with tin and copper liners to reduce fluorescence from lead and tin
x–rays. This reduces the background to \( \sim 2 \) counts per second (in the primary detector). A simulated image of the setup (including the lead cave, but excluding some internal detail of the detectors due to graphics limitations) is shown below in figure 7.15.

![Figure 7.15: A simulated image of the experimental setup. The primary detector is shown in the centre, with an example source (the small yellow cylinder) above. The cylindrical NaI(Tl) crystal is shown in green (with a thin grey aluminium casing), and the PMT’s are shown above the crystal as light grey cylinders. The NaI(Tl) plug is shown in the centre of the NaI(Tl) annulus as a dark grey cylinder. The surrounding lead shield and liner is also shown, with grey corresponding to lead, silver to tin, and red to copper.](image-url)
7.4.3 Measurement & Analysis

Apparent peak efficiencies and isotopic abundances were calculated as in section 7.2. Peak to count ratios and Compton suppression factors were calculated as in section 3.3. Calibration sources were chosen to cover the 37-662 keV energy range to fully characterise the energy response and efficiency of the LEGe crystal. Several single $\gamma$ emitters were used, as well as a NIST (National Institute of Standards and Technology) traceable complex $\gamma$ source. The isotopes that comprised these were $^{241}$Am (59.54 keV), $^{109}$Cd (88.03 keV), $^{57}$Co (122.06 keV), $^{139}$Ce (165.86 keV), $^{133}$Ba (302.85 keV, 356.01 keV), $^{113}$Sn (391.68 keV), and $^{137}$Cs (661.67 keV).

Analysis was completed using the post-processor to sort the data, and ROOT used for matrix manipulation and peak searching/fitting. Coincidences were identified by searching for all events in the NaI(Tl) detector with a time delay ($t$) between -10 $\mu$s and +10 $\mu$s (with $t = 0$ defined as the interaction time in the LEGe). These coincidences were then recorded into a 3D matrix with the energy deposited in the LEGe, energy deposited in the NaI(Tl) detector, and the time delay on each axis ($E_{\gamma 1}$, $E_{\gamma 2}$, $t$). This allows a delay spectrum to be created, with peaks identifying characteristic delays where there are a large number of coincident events. Time and energy gates can then be set to extract information from the dataset.

7.4.4 Results & Discussion

7.4.4.1 Full Coincidence Search

Data were acquired for one hour using a complex $\gamma$ source comprised mainly of $^{241}$Am, $^{109}$Cd, $^{57}$Co, and $^{137}$Cs. The energy spectrum for the LEGe with this source is shown below, in figure 7.16. The full delay window for this source (between -10 $\mu$s and +10 $\mu$s), is shown in figure 7.17.

In the time coincidence spectrum, there are four distinct regions; a well defined coincidence peak at $\sim$2 $\mu$s, a rising peak at $\sim$5 $\mu$s, the random coincidence background before the $\sim$2 $\mu$s peak, and a slightly increased coincidence background after the $\sim$2 $\mu$s peak. The random background coincidences are
7.4. Low–Energy Coincidence Systems

Figure 7.16: The energy spectrum recorded in the LEGe during one hour of data acquisition. The energy range was restricted to 700 keV to exploit the detectors low–energy performance.

Figure 7.17: The time distribution of the coincidences seen in the NaI(Tl) detector, when observing an event in the LEGe at $t=0$. Coincident events were extracted between -10 µs and +10 µs, and the resulting time delay spectrum shows several distinct event types.
due to chance coincidences from the source and additionally from background radiation. The raised background is due to the time-walk of low energy interactions \cite{76}, which causes coincidences to appear later than they would at higher energies. The coincidence peak at $\sim 2 \mu s$ is the standard signal used for Compton suppression, and is the result of a photon scattering out of the LEGe into the NaI(Tl) detector ($\text{LEGe} \rightarrow \text{NaI(Tl)}$). As the NaI(Tl) scintillation process is relatively fast (and this interaction happens after the interaction in the Germanium crystal), the coincidence peak is well defined. The rising peak at $\sim 5 \mu s$ is far less well defined, which is due to the order of the interaction process. This peak represents events that have entered the NaI(Tl) detector first, and then interacted with the LEGe ($\text{NaI(Tl)} \rightarrow \text{LEGe}$). The ‘rising’ shape is caused by the rise time of a pulse in the LEGe, which was set to $2.2 \mu s$ (and is consistent with the rise time in the coincidence time distribution).

### 7.4.4.2 Single Delay Gate

For the complex $\gamma$ source, the delay gate was set from $1.5 \mu s$ to $3.0 \mu s$ to cover the main coincidence peak. The spectra extracted are shown below, in figure 7.18.

![Figure 7.18: The raw, coincidence and anticoincidence spectra for the complex $\gamma$ source, with the delay gate set on the 1.5 $\mu s$ to 3.0 $\mu s$ coincidence peak. Note, that due to the little amount of coincidence seen above 300 keV, the anticoincidence spectrum overlaps with the raw spectrum.](image)

The coincidence spectrum clearly shows Compton scattered photons from
the various decay lines (\(^{241}\)Am at 59.5 keV, \(^{57}\)Co at 122.1 and 136.5 keV, \(^{137}\)Cs at 661.7 keV) in the complex source. Almost no full photopeak counts are seen in coincidence (which would only occur due to false coincidences). By suppressing these events, the suppression will match that of a typical electronic system, and the (P/C) and (P/T) ratios are substantially improved over the raw spectrum.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>( \text{P/C} )</th>
<th>( \text{P/T} )</th>
<th>CSF</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{241})Am (59.5)</td>
<td>67.66</td>
<td>169.14</td>
<td>171.22</td>
</tr>
<tr>
<td>(^{109})Cd (88.0)</td>
<td>10.65</td>
<td>26.50</td>
<td>26.63</td>
</tr>
<tr>
<td>(^{57})Co (122.1)</td>
<td>2.72</td>
<td>7.00</td>
<td>7.38</td>
</tr>
<tr>
<td>(^{57})Co (136.5)</td>
<td>2.23</td>
<td>3.13</td>
<td>3.13</td>
</tr>
<tr>
<td>(^{137})Cs (661.7)</td>
<td>16.04</td>
<td>15.96</td>
<td>39.2</td>
</tr>
</tbody>
</table>

Table 7.6: A summary of the suppression ratios calculated for the LEGe utilising single and multiple time delay gates. Values for unsuppressed spectra are denoted by a ‘U’, values for a single time gate (on the \( \sim 2 \) \( \mu s \) peak) with ‘G1’, and values for both time gates with ‘G2’.

### 7.4.4.3 Multiple Delay Gates

As there is another peak at \( \sim 5 \) \( \mu s \), additional suppression can be achieved if these events add to the background seen in the original spectrum. The coincidences were extracted from the peak at -6.2 \( \mu s \) to -3.2 \( \mu s \), and the resulting energy spectra are shown below, in figure 7.19.

![Figure 7.19: The raw, coincidence and anticoincidence spectra for the complex \( \gamma \) source, with the delay gate set on the -6.2 \( \mu s \) to -3.2 \( \mu s \) coincidence peak.](image-url)
Again, almost no full photopeak counts are seen in coincidence, however those events that are seen in coincidence are characteristic of the continuum from a higher energy decay or the natural background. As these events interact with the NaI(Tl) shield before the HPGe crystal, possible sources include Compton scattered photons (that interact with the NaI(Tl) shield and then backscatter into the LEGe), and Cosmic radiation such as high energy muons, fast and thermalised neutrons. A seven day background was taken to estimate the amount of coincidences caused by these events, and while some were seen, background radiation could only account for \( \sim 0.8 \% \) of the events in the 2 \( \mu \)s peak, and \( \sim 0.5 \% \) of the -5 \( \mu \)s peak. The extracted spectrum when suppressing both of these coincidence peaks is shown in figure 7.20. All suppression ratios are detailed in table 7.6.

With both coincidence peaks suppressed, the P/C, P/T, and CSF ratios are slightly improved for a range of nuclei in the low energy regime (typically by 2-10 \%). The suppression ratios for a higher energy decay (\(^{137}\text{Cs}\)) are improved by up to 144 \%. This is because the continuum counts at this energy are fairly low, and therefore the background reduction is far more dramatic than at lower energies. In the complex \( \gamma \) source used, there are higher energy decays. Compton suppression of these, however, cannot account for the reduction in the high energy background (when suppressing the -6.2
μs to -3.2 μs time gate), as these events would interact with the HPGe crystal first and produce the time delay signal seen at ~2 μs.

### 7.4.4.4 GEANT4 Simulations

To corroborate the findings in table 7.6 and confirm the origins of the -5 μs peak, GEANT4 simulations were performed to determine the proportion of coincident events that would be expected to interact in the LEGe first, and vice-versa. These were built using version 9.5.p01 of the GEANT4 toolkit.

The detector geometry was reproduced as accurately as possible, including the LEGe, both NaI(Tl) detectors and the shielding. The LEGe contained the detector crystal, outer and inner electrodes, the aluminium canister and the carbon epoxy window. The NaI(Tl) detectors included the crystal, detector casings and Aluminium cylinders (to approximate the PMT’s). To complete the model, the tin and copper lined lead shielding and source housings were also recreated. All simulations were based upon the previous models developed, with additional outputs to accommodate the required coincidence information.

The source was recreated in the simulation, and 10^6 events were run. Coincident events in the GEANT4 simulations (when energy was deposited in LEGe and NaI(Tl) crystals during the same event) showed that ~63 % of coincidences were due to a LEGe→NaI(Tl) interaction path, with the remaining 37 % interacting via NaI(Tl)→LEGe. This compares well to the value of 66 % for the LEGe→NaI(Tl) path, and 34 % via the NaI(Tl)→LEGe path seen in the data.

### 7.4.5 Summary of Results

List mode acquisition was used to run a full Compton suppression system with a low energy, hyperpure Germanium detector. This was achieved with no loss of coincidence data, and a greatly simplified experimental setup. Coincidence information has been successfully written to a 3D interaction matrix, allowing the efficient gating and extraction of events.

Analysis of the time delay spectrum also identified a coincidence peak be-
fore the interaction in the HPGe crystal (in addition to the standard Compton suppression peak). The energy spectrum of these coincidences is also substantially different to that seen in the standard delay gate. A major source of these coincidences has been identified as photons that are scattered out of the active NaI(Tl) shield (and into the LEGe), and GEANT4 simulations of the detector setup agree with the proportion of coincident events seen in each delay peak. This validates the use of GEANT4 for coincidence simulations, allowing further development to fully simulate a Compton suppression system.

Excellent suppression was found in the ‘standard’ Compton suppression configuration (with a single time gate), and a slight improvement on this was obtained at low energies using multiple time gates. A 144% improvement in the Compton Suppression Factor was achieved for the $^{137}$Cs peak when utilising multiple time gates to suppress coincident events. Multiple delay windows should therefore be considered in any coincidence system, as there may be substantial additional suppression to be gained.

### 7.5 BEGe Compton Suppression System

#### 7.5.1 Scope

The usual range for environmental analysis is from 30 keV - 3 MeV, however many applications require increased sensitivity in the low to medium energy (20 keV - 1.5 MeV) region to detect specific radionuclides. It is therefore desirable to use a BEGe detector for this analysis, and in particular a BEGe detector in conjunction with a Compton suppression system. This project utilised a BEGe detector (similar to that described in section 7.2) that was positioned within the Compton suppression system detailed in section 7.4. GEANT4 models were developed that contained all the features discussed so far, including advanced geometries, cascade summing calculations and coincidence simulations. These were then validated using the experimental equipment, proving the ability of GEANT4 to accurately simulate advanced detector systems. The results from this project were published in the Journal of Radioanalytical and Nuclear Chemistry, and a copy of this can be found in
7.5.2 Experimental Setup

The BEGe detector (model BE3825) and associated electronics are supplied by Canberra UK (Harwell, Oxfordshire), and is the same model as that used in the previous studies. LYNX™ digital signal processing units were again used for data acquisition, with all event information stored in list-mode. The NaI(Tl) veto detector used is the same as that in section 7.4.

7.5.3 Measurement & Analysis

Apparent peak efficiencies, isotopic abundances, peak to count ratios and Compton suppression factors were all calculated as before. Calibration sources were chosen to cover the 10-2000 keV energy range to fully characterise the efficiency of the BEGe crystal. Several single $\gamma$ emitters were used, as well as a NIST (National Institute of Standards and Technology) traceable complex $\gamma$ source. The isotopes that comprised these were $^{241}$Am (59.54 keV), $^{109}$Cd (88.03 keV), $^{57}$Co (122.06 keV), $^{139}$Ce (165.86 keV), $^{113}$Sn (391.68 keV), $^{137}$Cs (661.67 keV), $^{54}$Mn (834.84 keV), $^{88}$Y (898.04 and 1836.06 keV), $^{65}$Zn (1115.54 keV), and $^{60}$Co (1173.23 and 1332.49 keV).

Analysis was completed using the post-processor to sort the data, and ROOT for matrix manipulation and peak searching/fitting in a similar fashion to that in section 7.4.

7.5.4 GEANT4 simulation

The description of the detector geometry consists of the BEGe, both NaI(Tl) detectors (annulus and plug) and the shielding. The BEGe contains the detector crystal, outer and inner electrodes, the aluminium canister and the carbon epoxy window. The cold finger and vacuum spaces within the detector are also included, as are the dead layers within the crystal. The NaI(Tl) detectors were recreated with a NaI(Tl) crystal, the detector casing and Aluminium cylinders (to approximate the PMT’s). To complete the model, the tin and
copper lined lead shielding and source housings were also recreated. NIST compounds were used where available, and all components were reproduced as accurately as possible according to manufacturer specifications. These simulations were built using version 9.5.p02 of the GEANT4 toolkit.

7.5.5 Results & Discussion

7.5.5.1 Delay Window Analysis

A delay spectrum was created by searching the data for coincidences between -10 and +10 µs (see figure 7.21). As in section 7.4, multiple coincidence peaks were found when analysing this data, indicating the presence of multiple event types occurring with characteristic delays. When extracting the coincidence spectrum for each detector, the time window was further refined to only include events seen within the main coincidence peak.

![Figure 7.21: The time distribution of the coincidences seen in the NaI(Tl) detector, when observing an event in the BEGe at t=0. The insert shows the time distribution from section 7.4 where the two distinct peaks can be seen more clearly.](image)

The peak in the delay spectrum at ~0 µs is a convolution of a rising sawtooth shape peak, and a well defined, Gaussian shaped peak. The Gaussian peak is the standard signal used for Compton suppression, and is the
result of a photon scattering out of the BEGe into the NaI(Tl) detector (BEGe→NaI(Tl)). As the NaI(Tl) scintillation process is relatively fast (and this interaction happens after the interaction in the Germanium crystal), the coincidence peak is well defined. The rising sawtooth shaped peak is far less well defined, which is due to the order of the interaction process. This peak represents events that have entered the NaI(Tl) detector first, and then interacted with the BEGe (NaI(Tl)→BEGe). The ‘rising’ shape is caused by the rise time of a pulse in the BEGe, which is consistent with the rise time seen in the coincidence time distribution.

7.5.5.2 Efficiency Comparisons

Peak efficiencies were calculated for multiple sources, and are compared to the simulated values in figures 7.22 & 7.23.

![Graph showing efficiency vs energy](image)

Figure 7.22: The actual and simulated peak efficiencies for the BEGe detector. A variety of sources were each counted for 1 hour to minimise statistical error.

Excellent agreement was found across the energy range for the BEGe detector, with the detector response accurate to within 3 % for the efficiency of each peak.

In the case of the NaI(Tl) guard detector, only a limited number of efficiency points could be used as many peaks were unresolvable (the detectors poor resolution was due to both the crystal material and the difficulty in gain matching multiple PMT’s). The efficiencies for the NaI(Tl) detector are
accurate to 3% at 661.66 keV and over, however the GEANT4 model overestimates the NaI response at low energies. This is due to the gain setting for the NaI(Tl) detector, which notably reduced the signal rate below 100 keV.

For $^{60}$Co, significant amounts of cascade summing were seen due to the geometry of the NaI(Tl) detector. To correct for this, a cascade summing correction factor was calculated using GEANT4 as in section 7.3, which gave a value of $0.23 \pm 0.01$. When applied to the data, the efficiency for the $^{60}$Co source matched that of the GEANT4 calculated peak efficiencies. This provides further evidence that the model accurately reproduces the detector response, as cascade correction factors are highly sensitive to the detector geometry.

### 7.5.5.3 Compton Suppression

Both detectors are accurately (individually) characterised, however they must perform together to achieve Compton suppression. A source containing $^{241}$Am, $^{109}$Cd, $^{57}$Co, $^{139}$Ce, $^{137}$Cs and $^{60}$Co was used to calculate Compton Suppression Factors (CSF’s) for the Compton suppression system. The same factors were calculated for the simulated results, and these are compared in table 7.7. Good agreement was found between the CSF values for actual and simulated data,

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Figure 7.23: The actual and simulated peak efficiencies for the NaI(Tl) detector. A variety of sources were each counted for 1 hour to minimise statistical error.
and the unsuppressed and suppressed spectra for the $\gamma$ source used are shown in figures 7.24 & 7.25 respectively.

<table>
<thead>
<tr>
<th>Nuclide (energy in keV)</th>
<th>CSF Actual</th>
<th>CSF Simulated</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}$Am (59.5)</td>
<td>1.26</td>
<td>1.14</td>
<td>0.91</td>
</tr>
<tr>
<td>$^{109}$Cd (88.0)</td>
<td>2.61</td>
<td>2.47</td>
<td>0.95</td>
</tr>
<tr>
<td>$^{57}$Co (122.1)</td>
<td>2.88</td>
<td>3.17</td>
<td>1.10</td>
</tr>
<tr>
<td>$^{57}$Co (136.5)</td>
<td>1.99</td>
<td>2.12</td>
<td>1.06</td>
</tr>
<tr>
<td>$^{137}$Cs (661.7)</td>
<td>2.61</td>
<td>2.79</td>
<td>1.07</td>
</tr>
<tr>
<td>$^{60}$Co (1173.2)</td>
<td>1.68</td>
<td>1.67</td>
<td>1.00</td>
</tr>
<tr>
<td>$^{60}$Co (1332.5)</td>
<td>1.06</td>
<td>1.07</td>
<td>1.01</td>
</tr>
</tbody>
</table>

Table 7.7: A summary of the suppression ratios calculated for the BEGe with the delay gate set on the $\sim 0$ $\mu$s peak. These are compared to the simulated values from the GEANT4 model, and a ratio of the simulated to actual values is given in the final column. All values agree to within 10 %, and the errors range from 4-12 % (at 1 sigma). These are dominated by the contribution from the number of counts in the continuum, which may contain as few as 20 counts per channel.

Figure 7.24: The actual and simulated spectra for a complex $\gamma$ source. The full photopeaks and the continuum are well reproduced, and shown in more detail within the insert.

Both the unsuppressed (figure 7.24) and suppressed (figure 7.25) spectra show excellent agreement between the simulated and actual data for this source. The continuum is slightly under-produced in the suppressed (simulated) spectrum, and this is due to the over-suppression of the continuum in the simulations. This is primarily caused by the activity of the source ($\sim 3$ kBq); as the source activity increases, the dead time of the acquisition system
Figure 7.25: The actual and simulated suppressed spectra for a complex $\gamma$ source. The full photopeaks are well reproduced, however the continuum is slightly underestimated due to the activity of the source. A close up of the peaks are shown in the insert.

can cause it to miss coincident events [77]. When calculating the Peak to Count ratios, a large variation was seen in the continuum where the number of counts per channel was low (<20). To mitigate this, continuum counts were averaged over several bins, which greatly reduced the variation seen. All CSF values agree within 10%.

7.5.5.4 Simulation of a Realistic Source

To verify the accuracy of the GEANT4 simulation, a reference source was also simulated. This was comprised of a thin 12 mm diameter filter placed upon a 25 mm diameter steel disc. The isotopic composition was known to within 5%, and recreated within the GEANT4 simulation. The source included up to 30 distinct isotopes, which were primarily fission fragments with a combined activity of $\sim$6 kBq. The unsuppressed and suppressed spectra are shown in figures 7.26 & 7.27.

Excellent agreement was found between the simulated and actual results, however accidental summing between multiple events (due to the activity of the source) was not included in these simulations. This is because by default, GEANT4 decays radioactive isotopes until a stable isotope is reached, which
Figure 7.26: The actual and simulated spectra for a surrogate $\gamma$ source. The full photopeaks and the continuum are well reproduced, and shown in more detail within the insert. For a full colour image please see the online version of this article.

Figure 7.27: The actual and simulated suppressed spectra for a surrogate $\gamma$ source. The full photopeaks are well reproduced, however the continuum is slightly underestimated in the simulation due to an oversuppression of coincident events. A close up of the peaks are shown in the insert. For a full colour image please see the online version of this article.
therefore includes all subsequent emissions in a decay chain. You can limit this by specifying an end to a decay chain (typically the isotope that the user wishes to decay), however this cannot be applied to the complex sources here due to the way primary events are created in the simulations. The events from each nuclide were therefore simulated separately (where this limit could be applied), and brought together in post-processing to produce the final dataset.

The original anti-coincidence spectrum (figure 7.27) initially showed poor agreement with the data from the Compton Suppression system. This was due to both the low ($<150$ keV) and high ($>1.6$ MeV) energy coincident events being missed by the NaI(Tl) crystal due to the gain settings of the electronics. The lower energy events were missed due to difficulties in gain matching multiple PMT’s, while the higher energy events were missed as they were simply outside of the electronics acquisition range. Both of these event types were still classed as coincident in the GEANT4 simulations, causing an over-suppression of the continuum when compared to the data. By adding coincident events in the simulation that had an energy of $<150$ keV or $>1.6$ MeV in the NaI(Tl) detector back into the anti-coincidence spectrum of the BEGe crystal, this discrepancy was resolved.

The lack of accidental coincidences in these simulations will increasingly affect the accuracy of the model as the count rate increases (and sum events between uncorrelated emissions become more likely), however this is a small and often negligible effect in environmental samples. As with the previous source, the continuum was slightly over-suppressed in the simulation, which is likely due to a combination of the dead time in the detectors and inefficiencies in the acquisition electronics (the coincidence acquisition system also has a small ($<2\%$) inherent dead time). Given these limitations, simulations to predict the performance of a coincidence system should be treated as a best case scenario, as several factors may artificially limit the suppression achieved. It is worth noting that with standard electronics (running an online veto), the dead time can be greatly reduced as the incoming pulse need not be shaped, but simply used as a logic pulse to veto coincident events.
7.5.6 Summary

GEANT4 Monte Carlo simulations were successfully utilised to characterise a Compton suppressed broad-energy HPGe detector. The detector setup has been fully recreated in the simulation, which has been optimised to consistently reproduce the detector response. The peak efficiencies for both the BEGe and NaI(Tl) detectors agree with the simulated values for multiple test sources within 3 %, and cascade correction factors calculated using the model are consistent with the experimental data.

Compton suppression has also been simulated, with good agreement seen between the simulated and actual CSF values (<10 %) for multiple radionuclides. Simulations were also performed of a complex γ reference source, which contained up to 30 radionuclides with a total activity of 6 kBq. Excellent agreement was found for the detector response in both the suppressed and unsuppressed modes.

The model has been shown to be highly accurate, and can reproduce many aspects of the spectrum seen when performing environmental analysis. This includes peak efficiencies, the Compton continuum, true-coincidence (cascade) summing, and Compton suppression for a variety of source matrices and geometries. The accuracy of the model may be reduced when source activities increase, however this should not be an issue in the low activity regime that is typically measured. It has also been found that the effects of the systems electronics must be taken into account to fully reproduce the coincidence behaviour of the detectors. This model can now be used to fully characterise the detector response for any environmental source.
Chapter 8

Shielding Optimisation

8.1 Scope

The sensitivity of $\gamma$ (gamma) spectroscopy relies upon the absolute efficiency of the detector, and the ability to identify decays of interest above the natural background. This consists of terrestrial sources, including those within the detector, cryostat, shielding and surrounding materials, and secondary radiation produced by the interactions between the source radiation and the detector/shielding. Fast neutrons and muons also cause background counts in the detector, and are produced by Cosmic-ray interactions within the upper atmosphere.

The focus of the shielding work is the evaluation of different materials and geometries for a radiation shield to be used with HPGe detectors. Monte Carlo simulations were used to achieve this, and are based upon the previous simulations developed for the NaI(Tl) and HPGe based systems (using version 9.4.p01 of the GEANT4 toolkit). The results from this project were published in the Journal of Radioanalytical and Nuclear Chemistry, and a copy of this can be found in Appendix E.

8.2 Materials & Design

Several designs and materials were simulated, and all are be based around a ‘standard’ configuration of (from the outside in) lead (Pb), aged lead (low-Pb),
tin (Sn) or cadmium (Cd), and copper (Cu). The lead makes up the primary shield with a nominal thickness of 102 mm, and the Sn/Cd and Cu liners are 1.0 mm and 1.5 mm respectively. The combination of these liners have been shown to reduce the x-rays from the lead by up to 98\%\cite{29}. Cadmium is often used instead of Sn for its higher density, however it has a high neutron capture cross-section and emits a 558 keV photon when absorbing a neutron, which may adversely affect detector performance.

Plastic neutron moderators and absorbers were also investigated but not simulated, as these were found to have been studied extensively in a previous publication by Stewart et al\cite{78} (the results of which were in agreement with preliminary simulations carried out). Neutron moderators and absorbers must minimise the neutron flux and any subsequent photon production near the detector, and absorb thermalised neutrons to reduce neutron capture reactions within the detector or nearby materials. This rules out the use of internal moderators/absorbers, as thicknesses less than \(\sim\)15 mm produce additional neutrons, and all thicknesses produce additional \(\gamma\) radiation when subjected to a neutron flux\cite{78}. Large internal thicknesses can not be used as the plastic is a low-Z material, and will therefore create a large continuum from scattered photons. The best performing shields in reference\cite{78} (for stopping both neutrons and gammas) had separate neutron moderators and attenuators, used lithium doped PE (PE:Li), bismuth doped PE (PE:Bi) and Pb in a ratio 3:8:4.

8.3 Shielding Results

8.3.1 Terrestrial Radiation

Background radiation in the laboratory is equivalent to a \(\sim\)30 kBq (distributed) source with energies up to 3 MeV (assuming a 10\% efficiency for detection, and a 1 count per second, per keV average detection rate). This rate was verified experimentally, and is similar to that reported in reference\cite{29}. The decays seen consist of various radionuclides over the 30 keV - 3 MeV energy range, and the rate can change on a daily basis (mainly due to
Table 8.1: A summary of the materials considered in this study. Premadex, PE:B and water were eliminated as possible neutron shielding materials due to better performing alternatives. \( \gamma \) production values are for source neutrons interacting with 1 mm of material, and describe the amount of photons that penetrate the shielding per incident neutron.

As expected, larger shielding thicknesses provide greater protection from terrestrial radiation. The amount that penetrated the shielding is reduced to
8.3. Shielding Results

Figure 8.2: This spectrum is for $10^7$ 2.5 MeV photons fired directly at the detector from outside the shielding. There is a clear advantage to increasing the shielding to $\sim 150$ mm, although this may increase the number of interactions with muons and neutrons from cosmic sources.

0.01% of the initial flux (a factor of 10000) at $\sim 150$ mm Pb. This is an order of magnitude greater than for the standard, 102 mm case, and is preferred for larger detectors as they are increasingly likely to pick up high energy gammas that penetrate the shield.

8.3.2 Fluorescence and Compton Scattering from Internal Sources

Quantifying the levels of Compton scattering and Fluorescence within the shielding is a difficult process. The Compton scattered events must be discerned from scattered events in the detector, and the fluorescence must be counted above the continuum and/or peaks from other radionuclides. The former is prohibitively difficult, while the latter suffers from large errors or unreasonably large count times.

These obstacles can be overcome in Monte Carlo simulations by defining the source in such a way that it does not interact with the detector directly. This is achieved by limiting the initial photon directions to the $2\pi$ of the sources solid angle that is not subtended by the detector (see figure 8.3). Only secondary events are then seen, and an example spectrum is shown in
8.3. Shielding Results

Figure 8.3: The simulated geometry for a BEGe–6530, in a lead cave. By firing the photons away from the detector, no primary events are seen.

Figure 8.4: An example spectrum, showing the Fluorescence and Compton scattering from the shield and liners. The simulation included a 0.2 mm Cd liner, and ran for $2.0 \times 10^7$ events. Inset – a zoomed section showing the Pb (72–87 keV) and Cd (23–29 keV) x-rays in more detail.
The number of scattered photons seen in the simulations (minus the Pb x-rays) was normalised to the case where no liners were included. The levels of fluorescence seen were also recorded, however the statistics were too low to calculate the relationship to liner thickness with any accuracy. A separate artificial source was therefore created that only consisted of the x-rays required, and this was fired from outside the liners towards the detector. A summary of these results is shown in figures 8.5 & 8.6 and simulations comparing the inner diameter of the Pb cave to the background seen are shown in figure 8.7.

![Graph](image)

Figure 8.5: The percentage of 75-85 keV fluorescence photons seen as a function of liner thickness (normalised to a setup with no liners) for cadmium, tin and copper. The attenuation of 23-29 keV Sn and Cd x-rays with a Cu liner is also shown.

From figures 8.4, 8.5, 8.6 & 8.7 it is clear that the liners composition, thickness and the inner radius of the shield have an important effect on the amount of background seen. Cu is the worst performing material due to its low z-value, increasing the scattering seen by 128% ($\sim 1.6 \times 10^4$ events) and reducing the fluorescence seen by 80% ($\sim 1.7 \times 10^3$ events) at typical liner thicknesses (3 mm). Sn and Cd both perform well, reducing the fluorescence by 99.78% and 99.85% respectively at 3 mm. Scattering was increased by 49% and 64%, however Sn and Cd both emit further x-rays in the 23–29 keV range. These can be attenuated by a thin layer of Cu, however this should be minimised due to the additional Compton scattered events this will cause.
8.3. Shielding Results

Figure 8.6: The percentage of scattered photons seen as a function of liner thickness (normalised to a setup with a bare Pb cave) for cadmium, tin and copper.

Figure 8.7: The number of secondary events seen as a function of the shieldings inner radius. Liners were set to 1.0 mm Sn & 1.5 mm Cu for these simulations to mimic the current set-up and minimise fluorescence.
8.3. Shielding Results

If the x-rays are tolerable (or not within the energy range of interest) it is preferable to omit one or both liners completely, otherwise, 2.5 mm Sn/Cd with 0.5 mm Cu is optimal, as the Pb and Sn/Cd x-rays will be attenuated by >99.5%. Greater liner thicknesses also increase the chance of interactions with Cosmic neutrons and muons\textsuperscript{[79]}, and should therefore be kept to a minimum. The inner shield diameter should be set as large as is reasonably practical; a radius increase of 20% reduced the number of counts seen from scattered photons by 33%.

8.3.3 $^{210}$Pb Decay in the Shield

The lead shielding is layered, with standard lead used for the outside, and aged lead used for the inner shield, which must therefore be thick enough to attenuate the radiation from the outer shield. This was simulated by populating the outer lead shield with 500 Bq/kg $^{210}$Pb isotopes, and as expected, no radiation could be seen through aged lead (of at least 5 mm).

Further simulations were also undertaken to evaluate the thickness of the liners, and whether these could attenuate radiation from the inner (aged lead) shield. These results need to be considered within the context of the section 8.3.2 as increased liner thicknesses will also increase the amount of Compton scattering and Cosmic interactions seen. These results are shown in figure 8.8.

The 47 keV gamma decay is suppressed by increasing liner thicknesses as expected, with the intensity reduced to 0.001% for a 2 mm Cu liner. The $\beta$ decay of the $^{210}$Bi daughter causes a Bremsstrahlung continuum up to the endpoint energy at 1.162 MeV, which can be seen as the scattered photons in figure 8.8. These are not shielded as effectively as they are from a higher energy decay. To reduce the contribution from these events, the internal diameter of the lead should therefore be increased, or the $^{210}$Pb concentration minimised.

8.3.4 Neutron Absorption in the Liners

Neutron radiation from cosmic events is spread over a range of energies, and varies with both location and altitude. As cadmium has a high neutron in-
8.3. Shielding Results

Figure 8.8: The total number of events (normalised to the case where no shielding is used) seen in the detector from $^{210}$Pb decays in the inner shield, as a function of total liner thickness. Liners were split in a 3:2 ratio of Cu:Sn up to 2.5 mm total thickness, after this 1.0 mm Sn was used with increasing thicknesses of Cu.

Interaction cross-section, simulations were performed to investigate whether the liners could attenuate the neutron flux reaching the detector. This would be at the expense of a 558 keV $\gamma$-ray emitted when cadmium absorbs a neutron, however this may be outside the energy range of interest, and so be of no further consequence.

Figure 8.9: The neutron flux (normalised to the case where no liner is used) penetrating the liner, as a function of total liner thickness. 0.1, 1 and 10 MeV neutron sources were used with both Cd & Sn liners.
At 1 MeV and 10 MeV, neutron production is seen with greater liner thicknesses, but for energies less than this a reduction in neutron flux is observed (see figure 8.9). Due to its higher cross-section for neutron interactions, these effects are exaggerated for cadmium when compared to tin. For ‘real’ neutron spectra from cosmic sources, the neutrons are mainly of an energy <100 keV\[80, 81\]. Simulations were therefore performed using this spectrum, and the results for Sn and Cd are shown below in figure 8.10. The $\gamma$-rays produced with increasing thicknesses of Sn and Cd are also shown in figure 8.11.

![Figure 8.10](image)

**Figure 8.10:** The neutron flux (normalised to the case where no liner is used) penetrating the liner, as a function of total liner thickness. Both Cd & Sn liners were tested for neutron flux attenuation using a realistic cosmic neutron source from reference [80].

With a realistic neutron spectrum, both Sn and Cd liners reduce the neutron flux incident on the detector. Cd reduced the neutron flux much faster than Sn, however this reduction plateaued after a few mm of either material. This is due to the lower energy part of the incident neutron spectrum being absorbed by the liners, but the higher energy neutrons continuing through the material and producing more neutrons (as in figure 8.9). The reduction in background due to neutron absorption may however be offset by the increase in $\gamma$ production, which is up to 3 times greater in Cd than Sn. At 1 mm liner thicknesses, Cd reduced the neutron flux by $\sim 27\%$ and Sn by $\sim 10\%$. The corresponding values for $\gamma$ production are 1.39 and 0.55 $\gamma$-rays penetrating the liner per source neutron for Cd and Sn, respectively.
8.3. Shielding Results

8.3.5 Muon Absorption in the Shield

To simulate the effects of a Cosmic muon flux on a typical lead shield, the Cosmic muon spectrum from reference [82] was used to generate $10^6$ events for the geometry shown in figure 8.12. Substantial amounts of both muons and secondary radiation penetrated through the shielding to the detector, and the effects of increasing the shielding’s lid thickness are shown below in figure 8.13. The effects of increasing the body’s thickness was also investigated, however above 100 mm (the minimum needed to effectively shield terrestrial radiation), no statistically significant levels of additional radiation were found to enter the chamber.

Increasing the thickness of the shielding showed a corresponding increase in the total number of events seen in the chamber. This included muons that penetrated the shielding, but also neutrons, $\beta$-particles, gamma radiation and neutrinos. These are created by the muons interactions within the shielding, and the proportion of radiation seen at each lid thickness is shown in figure 8.13.

The major components that will interact with the detector (muons, $\beta$-particles, neutrons and gammas) maintain roughly the same percentage of
Figure 8.12: Cosmic muons are fired from a randomised coordinate in the x-y plane above the detector (the ‘sky’), and cover the entire area of the shielding. This allows a realistic flux to be simulated, and the effects of increasing the lid and body thickness to be independently estimated. The image shows a single muon track through the shielding, producing secondary gamma and neutron radiation.

the total number of particles seen irrespective of shield thickness, however the total number of events seen increases by $\sim 50\%$ in the first 50 mm of shielding, and a further 10% over the next 150 mm. This suggests that even small shielding thicknesses will increase the Cosmic muon derived background substantially in a detector, however further increases beyond the first 50 mm only increase the background seen by a relatively small amount.

8.3.6 Minimising the External Background Radiation

The simulations suggest that while increasing shield thicknesses reduce the impact of radiation from Terrestrial and Cosmic neutron sources, the background
8.3. Shielding Results

Figure 8.13: The percentage and type of secondary particles created by muon interactions in the shielding, as a function of shielding lid thickness. The percentage of each radiation type is normalised to the total number of events at that thickness, to show how the distribution of secondary radiation changes. The total number of events entering the detector chamber is also shown, which increases by \( \sim 60\% \) when the thickness is increased from 0 mm to 150 mm.

will be increased by Cosmic muon sources. By using a realistic Cosmic muon spectrum and an estimate of the Terrestrial background rate, an optimum shield thickness can be estimated. This result is shown in figure 8.14 which presents the relationship between shielding thickness and the total background that can be expected in a laboratory setting.

As the Terrestrial background rate is much greater than the Cosmic muon and neutron flux (by \( \sim 2 \) orders of magnitude) the effects of Cosmic radiation do not become significant until the shielding reaches a thickness of 80 mm, where the Cosmic muon derived background approaches 25% of the total seen. Even at the full 200 mm simulated however, the reduction in Terrestrial background outweighs the increase in Cosmic muon derived radiation. One other subtlety involves the type of radiation produced, as Cosmic muons cause additional neutrons to enter the chamber as well as the gamma and beta radiation that is typically seen from a Terrestrial source. This is a small fraction of the radiation produced however (\( \sim 0.2\% \)), and therefore the small increases seen at large shielding thicknesses will not produce a measurably greater neutron flux.
8.4 Discussion & Recommendations

Plastic neutron moderators and absorbers were initially investigated, but not fully simulated due to the comprehensive nature of previous publications (the results of which were found to be in agreement with preliminary simulations of neutron absorption). The best performing shields (for stopping both neutrons and gammas) have separate neutron moderators and attenuators, used lithium doped PE (PE:Li), bismuth doped PE (PE:Bi) and Pb in a ratio 3:8:4. It is therefore recommended that future shielding designs use this specification where the shielding of neutrons is critical, as it is known to be both commercially available and the most effective solution. Internal neutron moderators and absorbers are not recommended as these greatly increase the amount of neutron and $\gamma$ radiation seen in the detector.

From the simulations, it is clear that increasing the inner radius and thickness of the shielding are effective measures for reducing the background that a detector will see. Increasing the shield thickness from 100 to 150 mm will reduce the amount of terrestrial radiation seen by an order of magnitude, while a 30 mm increase in inner radius reduces the background seen by 33%.

Figure 8.14: The total background seen as a result of external factors, including Cosmic muons and Terrestrial radiation. As the thickness of the shielding increases, the Cosmic muon derived background contributes a larger proportion of the background radiation seen in the detector chamber. The errors in the total background count are dominated by variations in both the Cosmic and Terrestrial background rates.
To reduce fluorescence x-rays, liners of lower Z materials are needed, although these increase the overall level of background seen due to their relatively high Compton scattering cross-sections. The liners are not effective shields for radiation from $^{210}$Pb decays, as the higher energy component is not attenuated by the relatively thin layers. Liners of the thicknesses needed to achieve this would substantially increase the number of background events due to the reduced internal diameter of the cave, increased Compton scattering and increased interactions with Cosmic radiation.

Currently, Cu and Sn is used in a 3:2 ratio (typically 1.5 mm and 1.0 mm thicknesses respectively), however using a thicker liner of Sn or Cd would be of great benefit, as these are far more effective than Cu for shielding Pb x-rays, and only 0.5 mm of Cu is required to remove 99.5% of Sn/Cd x-rays. Minimising the thickness of the liners (especially Cu) in the shielding also significantly reduces the level of Compton scattering seen (Compton scattering causes $\sim$ an order of magnitude more background events than the x-rays themselves).

Liner materials (Cd and Sn) were also tested for their neutron absorption properties, with cadmium outperforming tin for a realistic Cosmic neutron spectrum. The net effect for both materials is neutron attenuation, reducing the flux by up to 28% at 3 mm. Additional $\gamma$ radiation is produced, as the liners are typically thin components, and cadmium also emits a 558 keV $\gamma$-ray from the $(n, \gamma)$ interaction. Depending on the experimental requirements, Sn and Cd both have their respective advantages and disadvantages, and will provide some added benefit for the reduction of the Cosmic neutron flux incident on the detector.

As expected, there is no way to substantially reduce the Cosmic muon flux (and associated secondaries) over the relatively short material thicknesses involved in a typical Pb shield. Any thickness of shielding that is practical in a laboratory will actually increase the background seen, however this effect is most crucial over the first 50 mm of material, where the background derived from Cosmic muons increases by $\sim$50%. Above this, the level of background radiation entering the detector chamber is only increased by a further 10%, and produces a much smaller effect than that caused by Terrestrial radiation.
and Compton scattered photons/fluorescence from the source.

As a result of the differing contributions, the overall background radiation levels entering the detector chamber continue to fall up to the 200 mm of Pb shielding simulated. The main contribution is from Terrestrial radiation up until 100 mm, after which the Cosmic muon derived background increasingly dominates. The majority of background (both Terrestrial and Cosmic muon derived) is in the form of gamma radiation, however a significant neutron flux is also present (mainly from Cosmic neutrons, with some contribution from neutrons created by impinging Cosmic muons). This cannot be effectively shielded in 200 mm, however external neutron moderators and absorbers may reduce this contribution. For a standard setup, the fluorescence/Compton scattering from source photons will make up the vast majority of background events seen, which can only be minimised by reducing the amount of material in the immediate vicinity of the detector.
Chapter 9

Optimisation of Future Compton Suppression Systems

9.1 Primary Detector Crystal

9.1.1 Scope

The CTBT network consists of seismic and atmospheric monitoring stations across the globe, which monitor for 83 key radionuclides that may be indicative of nuclear weapons tests and/or reactor incidents, including $^{140}$Ba, $^{95}$Zr, $^{99}$Mo, $^{141}$Ce, $^{147}$Nd, $^{131}$I, $^{134}$Cs and $^{137}$Cs [26]. Note that these are often very small signals amongst the natural background present in the atmosphere, which is dominated by isotopes from the natural Uranium ($^{238}$U) and Thorium ($^{232}$Th) decay chains, and cosmogenic radionuclides produced by Cosmic radiation (e.g. $^7$Be). In particular, $^{241}$Am (59.54 keV), $^{144}$Ce (133.52 keV), $^{99m}$Tc (140.51 keV), $^{141}$Ce (145.44 keV), $^{235}$U (143.76 keV, 185.72 keV), and $^{95m}$Nb (235.68 keV) are difficult to accurately detect and quantify due to the Compton continuum from higher energy $\gamma$-rays, which may obscure lower energy (and low activity) signals. One possibility to increase the detectors sensitivity in this range is to reduce the thickness of the detector material, which detrimentally affects detection efficiency for higher energy photons far more acutely than those of a lower energy.

This project aimed to evaluate a number of designs for a proposed environ-
mental γ spectroscopy system, optimising the material and thickness of the detector crystal to achieve the highest sensitivities for those radionuclides that may be obscured by higher energy photons. These results were published in the Journal of Environmental Radioactivity, and a copy of this can be found in Appendix E.

### 9.1.2 GEANT4 Simulations

All simulations were carried out using the GEANT4 Monte Carlo toolkit (version 9.6.p02), and based upon the simulations that were developed and validated in section 7. All dimensions and specifications were derived from manufacturer supplied values except for the detector crystals themselves. The radius of the crystal was set to 40 mm (equivalent to a 50 cm$^2$ crystal — a commonly available size), while the thickness was varied to obtain the optimum thickness for the radionuclides of interest. A simulated image of a HPGe detector is included as figure 9.1.

![A simulated image of a HPGe detector. Not all detail in the simulated geometry (dead layers, the full detector cradle) is visible in this representation due to graphics limitations. The carbon fibre window is shown in black, the casing in light grey, the cold finger and crystal holder in red, and the crystal itself in yellow.](image-url)
9.1.3 Material Choice

For $\gamma$ spectroscopy systems, the most common scintillation based crystals used include NaI(Tl), LaBr$_3$(Ce), and BGO, while the semi-conductor based materials are dominated by HPGe and Silicon. For the assay of environmental samples, there are several requirements for a detection system. The detector crystals must be able to be produced in large sizes, with a high Z material to maximise the efficiency of the system. The material must be stable and have low levels of radioactive contamination, and the resolution must be good enough ($\leq 3\%$) to separate multiple peaks within a complex $\gamma$ spectrum. The detector does not have to be particularly fast (one of the main advantages of LaBr$_3$(Ce), which has a light decay time of 35 ns\cite{83}), due to the low count rates experienced.

These requirements rule out NaI(Tl) and BGO, which have resolutions of $\sim 7\%$ and $\sim 16\%$ respectively, and Silicon detectors are primarily of interest in the 5–50 keV region due to their low Z value and the sizes of crystal available. This leaves HPGe and LaBr$_3$(Ce), which have resolutions of $\sim 0.2\%$ and $\sim 3\%$ respectively. LaBr$_3$(Ce) does have a substantial internal background from $^{138}$La ($788.7$ keV $\gamma$ from $\beta$ decay, and a 1435.8 keV $\gamma$ from $e^-$ capture), $^{227}$Ac (which decays via a series of five $\alpha$ decays to $^{207}$Pb), and Barium K x-rays from 31–38 keV\cite{84, 85}, however it can be used at room temperatures, allowing a much simpler system than HPGe (which requires cooling apparatus often several times the size of the detector itself).

Simulations were developed to evaluate the full photopeak efficiency of both HPGe and LaBr$_3$(Ce) materials with respect to both material thickness and photon energy. Up to 400 simulations were performed for each material, and the results are presented as efficiency maps in figures 9.2 & 9.3. Note that the efficiency is shown as both a contour (thin lines across the plot), and as a graded colour map to allow some interpolation of the contours.

Figures 9.2 & 9.3 provide minimum sizes to detect photons of a variety of energies. Both materials are effective absorbers, however LaBr$_3$(Ce) outperforms HPGe due to its larger Z number. Despite the combined advantages of additional efficiency and room temperature operation, the detector perfor-
Figure 9.2: The efficiency of a full energy deposition for a photon within a HPGe crystal for a range of photon energies and material thicknesses (absorption lengths). Thin (<20-30 mm) crystals of HPGe are effective absorbers for up to \(~300\) keV photons (with an efficiency of up to 50%).

Figure 9.3: The efficiency of a full energy deposition for a photon within a LaBr$_3$(Ce) crystal for a range of photon energies and material thicknesses (absorption lengths). The same efficiency performance as a 30 mm HPGe crystal can be achieved at 300 keV with only 15-20 mm of material.
mance does not offset the reduced resolution and increased background levels seen in a LaBr$_3$(Ce) crystal. HPGe will therefore be selected as the primary crystal, but the authors would like to note that in a $\gamma-\gamma$ system where coincidences are utilised (reducing the importance of the resolution and additional background), LaBr$_3$(Ce) would be the crystal of choice.

### 9.1.4 Crystal Design

Due to the cooling requirements of HPGe, the crystal itself must be in contact with a heat sink (often a copper based ‘cold-finger’). Usually (in a co-axial design) this is emplaced within a cutout in the crystal, with the electrodes on the outer and inner surfaces of the crystal respectively. Depending on the electrode design, this can result in a relatively thick ($<700$ $\mu$m) dead layer either on the outside or the inside of the crystal, which will attenuate low-energy photons. The electrodes also have to be carefully designed, as poor charge collection will reduce the resolution and performance of the detector. For this particular system, a large-area planar crystal is required to maximise efficiency for low-energy photons, while minimising interactions with higher energy $\gamma$’s. The outer dead layers must also be minimised, with only a small volume of the crystal lost to the heat sink.

The closest commercially available detectors that follow these design principles are the Broad-energy range from Canberra UK (Harwell, Oxfordshire), which can operate from 3 keV to 3 MeV[67]. Ortec (Oak Ridge, Tennessee) also produce a planar HPGe detector (the SLP and GLP range), however these are designed specifically for low energy ($<300$ keV) detection[86]. Planar crystals differ from the traditional co-axial design as they use a small electrode implanted into the back of a thin crystal. They also have a very thin external dead layer ($<1$ $\mu$m), increasing sensitivity to low energy photons. As the Canberra design has been shown to work (from a charge-collection and engineering point of view), and has a greater energy range (useful when requiring greater flexibility from a detection system), this will be used as a basis for the following simulations.
9.1.5 A Representative Source

When calculating the MDA for each radionuclide, the background is defined as any energy deposition that is not part of the full energy photopeak for that particular $\gamma$-emission. In a sample with multiple radionuclides, this includes the full energy photopeaks and Compton continuum from other photons. To accurately calculate the MDA, it is therefore important to have a representative background.

To simulate the background for each detector thickness, two reference sources were created using GEANT4. The first represents a worst case scenario for the continuum measured, where the background is dominated by photons from the highest energy $\gamma$ emitter routinely measured, $^{208}$Tl (2.614 MeV). As the proportion of photons that Compton scatter out of the crystal is greater at higher energies, the continuum that results from such emissions will be maximised. The second source contained multiple radionuclides that may be seen in CTBT reference samples, including $\gamma$ emitters such as $^{40}$K, $^{95}$Zr, $^{134}$Cs, $^{137}$Cs, $^{140}$La, $^{140}$Ba, $^{208}$Tl, $^{210}$Pb, and $^{226}$Ra. An average activity of 3 kBq was calculated from a number of such samples in the laboratory, and then used to estimate the number of radioactive disintegrations necessary to simulate a 7 day acquisition period.

The source geometry was defined as a compressed air filter (a geometry commonly used for CTBT measurements, 70 mm diameter and 26 mm thickness). When simulating photons of specific energies, photons were emitted in a randomised isotropic direction from this geometry, otherwise the ‘G4RadioactiveDecay’ module was used to fully recreate each radioactive decay in the simulation.

9.1.6 Optimising the Thickness of the Crystal

Simulations were initially carried out using both the $^{208}$Tl and CTBT background sources, with the background spectrum generated for every thickness of detector simulated (which ranged from 1 mm to 100 mm). Further GEANT4 simulations were then carried out to establish the full photopeak efficiency
for each $\gamma$-ray of interest (note that these fired $\gamma$ radiation with no coincidence information, and so cascade summing effects are not included in these calculations). By combining the efficiency, branching ratio and background information with the length of acquisition, an MDA can then be calculated for each nuclide of interest and detector thickness. Simulated background spectra for the CTBT source are shown in figure 9.4.

![Figure 9.4: A subset of the 'background' spectra simulated for 1–100 mm thick HPGe detectors with the CTBT source. This contains multiple radionuclides that are of interest to CTBT measurements, and therefore allows the estimation of a typical background for the calculation of the MDA’s.](image)

Increasing the thickness of the HPGe detector clearly improves the overall efficiency of the system, however there is a limit to the improvement that is achievable, with little efficiency gain seen above a detector depth of 50 mm for this particular source. The increased efficiency at larger thicknesses drives up the total number of events seen, and therefore the MDA for lower energy radionuclides. Efficiencies were generated for each radionuclide and material thickness, and are shown in figure 9.5.

By combining the efficiencies calculated and the background seen, the MDA’s can then be calculated for the radionuclides of interest (figures 9.6 & 9.7).

From the MDA’s calculated for a range of nuclei, it is clear that there is no or very little detrimental effect on the nuclides of interest when increasing
Figure 9.5: The efficiencies calculated for each permutation of detector thickness and nuclide of interest. Photons of the required energies were fired isotropically from the test source, and the full photopeak efficiency calculated for the main $\gamma$ decays in $^{241}$Am, $^{144}$Ce, $^{99}$Mo, $^{141}$Ce, $^{235}$U, $^{95m}$Nb and $^{99m}$Tc. Note that lines between the points are shown to aid visualisation of each series, and do not represent interpolated values.

Figure 9.6: The MDA’s calculated for each radionuclide using a pure $^{208}$Tl source for the background calculation. Errors are dominated by the uncertainties in the source, such as its positioning, the source matrix itself and the average source activity.
9.1. Primary Detector Crystal

Figure 9.7: The MDA’s calculated for each radionuclide using a reference CTBT source. Note that the MDA’s calculated are much higher than those for the $^{208}$Tl source due to the increased continuum seen throughout the energy range.

The crystal size. For the pure $^{208}$Tl source (which represents the worse case scenario due to the high energy $\gamma$ decays), a minima in the calculated MDA is seen at $\sim 15$ mm for a range of nuclei, however the subsequent increase seen in MDA with increasing crystal thickness is minimal. For the CTBT reference source (which is much more typical of a reference sample), no minima is found in the calculated MDA’s, with the sensitivity achievable levelling off past 20–30 mm.

The overall ‘background’ with each source increased as expected, however only to a limiting point where the efficiency is then dominated by the radius of the crystal, not the depth. Also, at lower energies (where the continuum level is most crucial for the nuclides of interest) the ‘background’ levelled off after 30–40 mm of HPGe. This is due to a combination of effects that act to stabilise the continuum seen. With increased crystal thickness, higher energy events are more likely to interact with the detector, increasing the Compton continuum as these photons scatter out of the crystal. The proportion of low and medium energy photons Compton scattered out of the crystal, however, will fall due to the increased detector material, therefore
9.1. Primary Detector Crystal

reducing the continuum.

The MDA’s achieved for each permutation of detector are minimised for the nuclides of interest with a 30 mm HPGe crystal, and there is no obvious benefit (or detriment) to increasing the crystal depth further. The only nuclide that showed a different behaviour with the CTBT source is $^{241}$Am, where the energy of the $\gamma$-emission is far lower than that of the other nuclides evaluated. Again, however, there was very little detrimental effect to increasing the crystal depth.

For higher energy decays, increased crystal depth has obvious benefits. For this particular system however, it is recommended that the crystal depth does not exceed 30 mm, as no additional sensitivity can be gained, and larger crystals will suffer from increased interaction from conventional background sources, such as the cosmic muon and neutron flux.

9.1.7 Summary

Monte-Carlo simulations were been utilised to determine the optimum material and thickness for a $\gamma$ spectrometer to be used in environmental monitoring. HPGe and LaBr$_3$(Ce) were initially considered for use, however the additional background radiation and lack of resolution in the latter drove the selection of HPGe for further optimisation.

Multiple thicknesses were considered for the HPGe detector, with the aim of maximising the sensitivity of the system for radionuclides in the 50–300 keV energy range. By restricting the thickness of the HPGe crystal to 30 mm (currently the largest thickness routinely produced for the crystal design considered), the system has been optimised to both reduce the continuum from higher energy photons, and maximise the efficiency of the detector. While no obvious detrimental effects were observed when increasing the crystal depth past this size, it is recommended to minimise the amount of HPGe used as an increased volume of material will suffer from a greater interaction rate with the cosmic and terrestrial background.


9.2 Secondary Veto Crystal

9.2.1 Scope

This project aimed to build upon the results of the previous section, and optimise a secondary ‘veto’ detector system to be coupled with the optimised HPGe primary detector. This involved an evaluation of possible veto detector materials and designs, and the subsequent optimisation of component thicknesses to deliver a design that could substantially improve upon the current system. These results were published in Nuclear Instruments and Methods in Physics Research Section A (a copy of this can be found in Appendix E).

9.2.2 Overview of materials considered

The secondary detector crystals must be able to be produced in large volumes (>100 cm$^3$), allow relatively fast signal formation (as the volumes will be large, the count rate will be high), and the Z values and material densities must also be as high as possible to reduce the amount of material needed to absorb scattered γ radiation. As all detectors must fit inside the lead shield, plastic scintillators were ruled out (due to their low Z-values, and large resulting sizes). To minimise cost and maximise efficiency, NaI(Tl), BGO, and LaBr$_3$(Ce) scintillators were selected for further evaluation.

To test each materials effectiveness as a detector, simulations were created in which the number of interactions could be recorded as a function of both impinging γ energy and material thickness. By combining the results of each simulation set, the materials effectiveness as a detector can be depicted below, in figures 9.8−9.10.

BGO is clearly the most effective material for interacting with γ’s across the entire energy range. To interact with 50 % of impinging γ’s at 1 MeV, the amount of material required would be ∼30 mm of NaI(Tl), ∼21 mm of LaBr$_3$(Ce), or ∼13 mm of BGO. For the current design (which contains 80 mm of NaI(Tl)), the same performance (for γ’s of energy 1 MeV) could be obtained with ∼52 mm of LaBr$_3$(Ce), or ∼38 mm of BGO. Alternatively, the performance of the NaI(Tl) shield (again for 1 MeV photons) could be
Figure 9.8: The efficiency of an interaction (not necessarily a full energy deposition) for a photon within a NaI(Tl) crystal for a range of photon energies and material thicknesses (absorption lengths). While low energy photons (<100 keV) are absorbed with high efficiency in only a few mm of material, 1 MeV photons require $\sim$145 mm of NaI(Tl) to interact with a probability $>$95%.

Figure 9.9: The efficiency of an interaction (not necessarily a full energy deposition) for a photon within a LaBr$_3$(Ce) crystal for a range of photon energies and material thicknesses (absorption lengths). LaBr$_3$(Ce) has a greater stopping power than NaI(Tl), and a minimum of 100 mm of material would be required to stop 1 MeV $\gamma$’s with a 95% probability.
Figure 9.10: The efficiency of an interaction (not necessarily a full energy deposition) for a photon within a BGO crystal for a range of photon energies and material thicknesses (absorption lengths). Medium energy photons (<500 keV) are absorbed with far higher efficiency than for NaI(Tl) or LaBr$_3$(Ce), and 1 MeV photons require up to ~65 mm of BGO to interact with a probability >95%.

improved by up to 14% if LaBr$_3$(Ce) was used, or 25% if BGO was used.

Despite NaI(Tl) performing the worst of the three materials, it is the cheapest and most widely available. Resolution for the three materials ranges from 3-4% for LaBr$_3$(Ce), 6-8% for NaI(Tl), and ~15% for BGO, however LaBr$_3$(Ce) is currently restricted in size to 75 x 75 mm crystals. LaBr$_3$(Ce) also has much faster timing properties, with a typical decay time of 35 ns [83], compared to 230 ns for a NaI(Tl) crystal, and 300 ns for a BGO crystal [18].

If the secondary detector is used for purely as a veto, then BGO is the best option, as this minimises the material needed for the veto, and maximises interaction efficiency. If the secondary crystal is also required to output a usable spectrum, then (if 75 mm crystals are efficient enough) LaBr$_3$(Ce) would be a far better choice due to its impressive energy resolution, timing characteristics, and stopping power. Where crystals are required to be more efficient than 75 mm of LaBr$_3$(Ce), the best compromise would use NaI(Tl) to veto/record coincident events.

For this project, the suppression system is to be used solely in veto mode, with no spectral information required. BGO will therefore be selected as the
principle material for the secondary crystal.

### 9.2.3 Overview of the design considered

Several possible designs were considered, and infeasible, problematic or undesirable designs were ruled out. This includes conical secondary detectors, as there is no need to fit many Compton Suppressed HPGe detectors in close geometry around a single focal point. Such a requirement is useful for large arrays of detectors, but in the domain of single HPGe crystals it would only serve to artificially limit the efficiency of the secondary detector. Any designs that required a source to be collimated, or that prevented a source from being placed in direct contact with the primary detector were also eliminated, as these would severely limit the efficiency of the system (which is the most critical factor when counting environmental radiation). Any designs that were too large to fit into a standard Pb cave (of inner diameter 230 mm) were also eliminated, as this is required to substantially lower the background seen in the detector system. The largest environmental source that must fit within the system is a compressed air filter, which can measure up to 70 mm diameter $\times$ 26 mm thickness.

Re-designing the primary detector was considered, which is an especially attractive prospect when considering the possible advantages of creating a veto system with no dead layers between primary and secondary crystals. This idea was rejected however, as not only is the design and manufacture of such a system fraught with engineering challenges, but the removal of the copper crystal holder and cold finger (which make up the majority of the dead material within the immediate vicinity of the HPGe detector) would require the entire apparatus to be cryogenically cooled for the operation of a HPGe detector. The system envisaged will instead use a commercially available primary detector that has been designed to minimise dead layers and photon attenuation within the system.

Three main components of a veto detector were considered for this project, including the main cylinder around the primary detector, a base plate for forward scattered $\gamma$ radiation$^{[87]}$, and a lid for backscattered radiation. All
9.2. Secondary Veto Crystal

designs are based upon BEGe 5030 primary detector from Canberra UK (Harwell), which has a 110 mm outer casing. This detector has been selected as a result of section 9.1, which showed a 30 mm thick, planar crystal to be the most effective size for maximising the sensitivity of the primary detector to environmental radiation. Each component will be evaluated for a variety of material sizes and offsets, with the most effective design defined as having the greatest suppression ratios up to 1.5 MeV.

9.2.4 GEANT4

The GEANT4 (version 9.6.p02) simulations were based upon the previous models developed and validated, with the geometry changed to accommodate the different detector setups.

The description of the detector geometry consists of the BEGe, secondary detectors and the shielding. The BEGe contains all previous internal detail, including the electrodes, vacuum spaces and dead layers within the crystal. The secondary detectors were recreated with a crystal, the detector casings and Aluminium cylinders (to approximate the PMT’s). To complete the model, the tin and copper lined lead shielding and source housings were also recreated, with NIST compounds used where available. The primary detector components were reproduced as accurately as possible according to manufacturer specifications.

For the majority of the simulations, multiple sources were considered. These include $^{241}$Am, $^{134}$Cs, $^{137}$Cs, $^{54}$Mn, and $^{60}$Co. All were simulated in the geometry of a compressed air filter, as this represents the largest, most dispersed source that would be routinely used. Due to the finite size of the crystal, geometrically large sources will increase the amount of Compton Scattering seen.

9.2.5 A typical event

Simulations were performed for a range of energies from 20 keV - 1.5 MeV. These fired photons directly at the primary detector crystal from a coordinate 10 mm above the detector. Each event was tracked to build up a map of where
the photons typically scatter at each energy. The results of these are shown below, in figure 9.11.

![Figure 9.11: A series of plots showing the distribution of photons that interact with the primary detector at a variety of energies. Low-energy photons are mostly backscattered, while higher energy photons tend to either penetrate the detector crystal completely, or interact and scatter in a forwards direction. For the final (2 MeV plot) the current Compton Suppression system (including the annulus and plug) is overlaid for reference.](image_url)

In figure 9.11, the plots show two main interaction points for γ’s emitted towards the crystal. These are the crystal itself, and the copper cold finger
below the crystal (especially for high-energy photons). The final plot (with the
current system overlaid) shows that there is significant amount of scattered
events missed by the current system. While some of these scattered photons
will be due to events that interact with (and scatter from) the cold finger,
there is clearly a benefit to placing additional vetoing material in the area
below the main detector.

### 9.2.6 Current Performance

The current system, (described in section 7.5), uses multiple large volume
NaI(Tl) crystals to veto coincident events. Due to the geometry of the system,
it also heavily suppresses radiation emitted from the source in cascade. This
is desirable in some situations, however it does reduce the statistics available
for identifying radionuclides. The primary detector is also smaller than the
BEGe 5030, however this has been replaced in these simulations to make the
results comparable.

Simulations were run with the full set of sources listed above, and the
suppression factors achieved are detailed below in table 9.1 along with the
peak efficiency in the unsuppressed and suppressed modes at each point. As
these simulations reproduce the radioactive decays of each nuclide using the
GEANT4 decay librarys, cascade summing is also included in the efficiency
values calculated. Suppression factors were also calculated for a range of
incident $\gamma$ energies, and are detailed in figure 9.12. An example spectrum for
both the unsuppressed and suppressed modes of operation is shown in figure

It is clear from table 9.1 that Compton Suppression is only beneficial for
certain radionuclides. Any that decay via a cascade of $\gamma$ emission ($^{134}\text{Cs}$,
$^{60}\text{Co}$) are suppressed as multiple coincident $\gamma$ emissions cannot be discrimi-
nated from a Compton Scattered event. Detector dead layers, crystal holders,
casings and endcaps all attenuate events that do escape, limiting the amount
of suppression achievable.

The large peak CSF values calculated for low energy emissions (figure 9.12
are due to the relative reduction of the Compton edges for each energy of
Figure 9.12: The Peak and Total Compton Suppression Factors calculated for the current Compton Suppression system. These represent the system response for singular $\gamma$ emissions, and therefore the theoretical potential of the system at each energy in the case where no accidental or cascade coincidences are emitted from the source.

Figure 9.13: A simulated spectrum for a $^{60}$Co source in the compressed air filter geometry, showing both unsuppressed and suppressed modes of operation for the current NaI(Tl) based Compton Suppression system.
Table 9.1: A summary of the suppression ratios and peak efficiencies calculated for a variety of sources. For nuclei that decay via a singular $\gamma$ emission, the Compton continuum is suppressed by up to a factor of four. Nuclei that decay in cascade still show some improvement for most major lines, however the loss of counts in the peak limit the suppression factors achieved.

incident photon. $\gamma$ emissions that are particularly low in energy (for example $^{241}$Am) are unlikely to scatter out of the crystal, and the small amount of radiation that does is extremely limited in geometry. As low energy emissions do not penetrate far into the crystal, the vast majority of scattered events escape backwards, away from the primary crystal and system dead layers. This allows extremely effective suppression of these photons with a detector above the primary crystal.

The values in table 9.1 and figure 9.12 represent a baseline for the performance of the proposed Compton Suppression systems. Note that these simulated values represent the peak performance of the Compton Suppression system, as additional dead time and acquisition electronics effects can reduce the performance seen in the laboratory.

### 9.2.7 Proposed Configuration

The most effective configuration for Compton Suppression is an all enclosing geometry, where scattered photons (at all angles) are intercepted by the veto detector. As the system will be primarily tasked with environmental studies (where sources have low activities) there is no detrimental effect to surrounding the source with secondary crystals (as in the current design). If the secondary crystal were modular in design, then parts of the veto system (for example those crystals surrounding the source), could be turned off to
minimise the veto of nuclei that decay via a cascade of $\gamma$ radiation.

The proposed system will use the minimum amount of material to veto the maximum amount of events. As the height of the cylindrical crystal is increased, there will be diminishing returns as the cross-sectional area decreases for each increment in height. It is therefore desirable to minimise the height of the main secondary crystal, and then include lid and base detectors of appropriate thicknesses.

### 9.2.7.1 Main body

The chamber required for sources is 100 mm high, and the width of the detector (110 mm) across. The top of the secondary crystal will therefore be set to this height, and the base of the crystal in line with the base of the detector casing (making the total length $\sim$240 mm). The thickness of the BGO cylinder can then be varied to establish the optimum amount of material. The thicknesses evaluated range from 30 mm to 70 mm, as 30 mm gives the minimum level of performance needed (from figure 9.10), and 70 mm is the maximum possible size within the shielding. These results are presented in figure 9.14.

![Figure 9.14: A comparison between the peak and total Compton Suppression Factors for varying thicknesses of a BGO secondary crystal. This contained a cylindrical annulus from the base to 100 mm above the face of the HPGe detector.](image)

Increasing the thickness of the BGO veto detector improves the CSF val-
ues as expected. The additional material particularly improves low to medium energy suppression, as scattered events that deposit a small amount of energy in the HPGe detector typically scatter with a medium to high energy remaining. The energy range from 10−1000 keV, which is the major beneficiary of an increased cylindrical size, is critical for observing many important radionuclides. Typical spectra for a $^{60}$Co source in a system with a variety of cylinder thicknesses are shown in figure [9.15]

![Graph showing spectra comparison](image)

Figure 9.15: A comparison between the suppressed spectrum when simulating a $^{60}$Co source. The thickness of the main veto detector was varied between 30−70 mm for these runs.

The benefit of the main secondary crystal is realised across the entire energy range, as this intercepts the majority of scattered events from the HPGe primary crystal. Due to the cross-sectional area it subtends for scattered events from the primary crystal, this should always be the first component of a Compton Suppression shield to be implemented. While the optimum thickness is around 50−60 mm, improvements in the suppression ratios are seen up until the full 70 mm thickness for the energies simulated.

9.2.7.2 Lid

To veto γ’s that have scattered from the primary detector back towards the source, a ‘lid’ can be placed on the system. In the simulations of this configuration, the ‘lid’, main secondary crystal body, and the primary detector
are all present in the geometry, and the performance of the system with a variety of ‘lid’ thicknesses is compared to the performance of a 70 mm thick annulus (with no lid present). This allows a realistic estimation of the suppression ability of the ‘lid’ to be determined, while retaining any effects that the main veto detector will have. The ‘lid’ was placed 100 mm above the primary crystal, with a diameter equal to the outer edge of the main annulus. Compton Suppression Factors for this setup are shown in figure 9.16 and example spectra in figure 9.17.

![Figure 9.16: A comparison between the peak and total Compton Suppression Factors at a variety of photon energies and ‘lid’ thicknesses. A substantial improvement in the peak Compton Suppression Factors is seen at the Compton edges due to the capture of backscattered photons. Note that this happens across the energy range for backscattered photons, however it makes up a far larger proportion of the continuum counts at lower energies, hence the dramatic improvement in this region.](image)

The lid thickness was varied from 10–50 mm, with the majority of the additional suppression achieved seen at the Compton edges, (where photons deposit the maximum amount of energy when scattering, and are therefore likely to be scattered through \(\sim 180^\circ\) towards the ‘lid’). While this equates to a small improvement in the peak and total Compton Suppression Factors, it is a substantial improvement in a specific part of the spectrum. Improved suppression was seen for all ‘lid’ thicknesses, however it was very slight above 30 mm.
9.2. Secondary Veto Crystal

9.2.7.3 Base

High energy $\gamma$ radiation is more likely to be forward scattered, and therefore additional suppression may be achieved with a ‘base’ detector situated below the primary detector. As with the ‘lid’ simulations, the main crystal was included in the geometry, and used in conjunction with the base detector. Results from these simulations are shown in figures 9.18 & 9.19.

Similar levels of suppression are achieved with a ‘base’ detector, however it is concentrated in the lower energy region of the spectrum. This is a result of high energy photons depositing a small amount of energy in the primary crystal, and then interacting with the ‘base’ detector after scattering forward, beyond the primary crystal. The ‘peak’ at $\sim 250$ keV is a result of photons that backscatter out of materials surrounding the detector, including the aluminium canister and copper crystal holder/cold finger. This can only be minimised by reducing the amount of dead material surrounding the detector. An effective thickness for the ‘base’ detector was found to be 40 mm, with little additional suppression achieved above this (for a $^{60}$Co source).
Figure 9.18: A comparison between the peak and total Compton Suppression Factors at a variety of energies and ‘base’ thicknesses. The thickness of the ‘base’ detector was varied between 20–60 mm for these runs.

Figure 9.19: A comparison between the suppressed spectrum when simulating a $^{60}$Co source. Most of the additional suppression achieved with the base detector is at low energies, which is a critical range for environmental monitoring.
9.2.7.4 Combination

So far, effective sizes have been determined for the main body (50 mm), the ‘lid’ (30 mm), and the ‘base’ (40 mm) secondary detectors, which can now be utilised to evaluate the performance of a combined system. The suppression factors achievable are detailed in figure 9.21, with example suppressed spectra in figure 9.22. These are also compared to the current Compton Suppression system, the proposed system with only the main body, a system with the maximum current sizes available (70 mm cylinder, 50 mm lid, 40 mm base) and an oversize system (100 mm cylinder, 60 mm lid, 50 mm base) that would require a new lead cave. The envisaged design is shown in figure 9.20.

The main cylinder significantly lowers the continuum seen in the primary detector to a value less than that seen in the current NaI(Tl) based system, however it does not perform as well at the Compton edges as it cannot intercept photons that are backscattered. The ‘lid’ and ‘base’ detectors, however, significantly improve the coverage of the veto detector. This is most obvious at the Compton edges, however additional performance is seen throughout the energy range. While the current system achieves continuum reductions of a factor of \(\sim 30\)–\(40\), a BGO based system (with a 70 mm cylinder, 50 mm lid, and 40 mm base) can reduce the continuum by up to a factor of 240, with additional material in the three components of the secondary detector improving performance further.

The fundamental limit to the suppression ratios achievable arises due to dead layers in the system (crystal dead layers, detector casings, crystal holders etc.), which cannot (currently) be avoided. These layers are detrimental in a variety of ways. Primarily, dead layers reduce the chance of a photon that is Compton scattered from the HPGe detector interacting with the shield detector (it is worth noting that the reverse process, which contributes a significant fraction of coincident events to the total seen, is also reduced in efficiency). Source photons interacting directly with the dead material (especially the crystal holder, cold finger, and shield liners) may also scatter into the primary detector, and therefore contribute to the continuum (an obvious manifestation of this is the backscatter peak at \(\sim 250\) keV).
Figure 9.20: A cross-section of the proposed geometry for a high-efficiency Compton suppression system (for clarity, the PMT’s and Pb shielding are not shown here). The primary detector is shown in the centre, with the copper crystal holder and cold finger (in red) surrounding the HPGe crystal (in green). The veto crystals are also shown in green, and the casings for all detectors are drawn in grey (note that for the veto detectors, only the inner casings are shown). When considering how to extract the signals from each veto detector, it may be necessary to modify the main annulus and lid design, such that the main cylinder is taller and the ‘lid’ becomes a plug, (as in the current system), which would allow PMT’s to be mounted as shown in figure 7.15 The ‘base’ detector will require additional space below the system for PMT’s, or alternatively, a light guide may be used to allow the PMT to be more conveniently positioned.
Figure 9.21: A comparison between the peak and total Compton Suppression Factors at a variety of energies and system configurations. All systems perform well for low energy emissions except a cylinder with no lid, which cannot suppress backscattered photons.

Figure 9.22: A comparison between the different systems when suppressing a $^{60}$Co source. The inclusion of a ‘lid’ is crucial to minimise the Compton edges, while the ‘base’ veto detector substantially reduces the low–energy region of the spectrum.
### Table 9.2: Peak Compton Suppression Factors achievable with the different designs evaluated in this study. These include the current system, a cylindrical BGO shield, and a cylindrical shield with a base and lid at various thicknesses.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Energy (keV)</th>
<th>Current system</th>
<th>Main cylinder, lid and base thicknesses (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>50 (no lid/base)</td>
<td>50, 30, 40</td>
</tr>
<tr>
<td>$^{241}\text{Am}$</td>
<td>59.54</td>
<td>1.00</td>
<td>1.03</td>
</tr>
<tr>
<td>$^{134}\text{Cs}$</td>
<td>569.26</td>
<td>0.62</td>
<td>1.31</td>
</tr>
<tr>
<td>$^{134}\text{Cs}$</td>
<td>604.71</td>
<td>2.77</td>
<td>8.20</td>
</tr>
<tr>
<td>$^{134}\text{Cs}$</td>
<td>795.91</td>
<td>1.62</td>
<td>2.83</td>
</tr>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>661.66</td>
<td>4.28</td>
<td>3.83</td>
</tr>
<tr>
<td>$^{54}\text{Mn}$</td>
<td>834.83</td>
<td>4.00</td>
<td>7.87</td>
</tr>
<tr>
<td>$^{60}\text{Co}$</td>
<td>1173.24</td>
<td>2.84</td>
<td>8.26</td>
</tr>
<tr>
<td>$^{60}\text{Co}$</td>
<td>1332.5</td>
<td>2.99</td>
<td>7.85</td>
</tr>
</tbody>
</table>

The Compton Suppression factors achieved with realistic sources (and for each permutation of system design) are detailed in Table 9.2. Large improvements over the current design are clearly possible, even for nuclei that decay via a cascade of $\gamma$ emission. If data is collected in both suppressed and unsuppressed modes, then emissions that are detrimentally affected by the suppression system can be analysed from the unsuppressed data, resulting in no loss of information.

#### 9.2.8 Summary

GEANT4 Monte Carlo simulations have been utilised to develop an optimised, next generation design for a Compton suppression system. A realistic model, based upon an existing system and validated with a variety of detectors, has been developed. This was then used to identify the optimum size and material of a veto detector.

Several possible materials were investigated, with BGO identified as the most effective material for a suppression shield due to its excellent interaction efficiency. A routine that tracked where photons scatter in the simulation has also been developed, and used to identify key areas where suppression may be achieved/improved, such as the area below the primary detector where high energy $\gamma$’s tend to scatter.

Simulations were then utilised to estimate the performance of a BGO shield, and optimise the thicknesses and sizes of each shield component. Compton Suppression Factors were improved by a factor of two over the cur-
recent Compton suppression system, and the continuum reduction for common sources (such as $^{60}$Co) is improved by up to an order of magnitude. This equates to a reduction in the continuum of up to 240 times, which is critical for low-energy, low-activity radionuclides that are often swamped by the large continua from higher energy emissions.

9.3 Coincidence Corrections for Multi–Detector Systems

9.3.1 Scope

Following on from the design studies for primary and secondary detector crystals, further research was conducted into optimising the coincidence window, which would improve the suppression of coincident events.

Coincidences are identified by setting a coincidence window (usually within a few $\mu$s of an event registering in a detector) where a coincidence is recorded if signals are seen in any additional detectors. Properly configured (whether in hardware or software) this will contain the coincident events of interest, however it will also contain some random, or false coincidences that are due to the activity of the source, and/or terrestrial and cosmic background radiation. The exact timing of the coincidences seen also has an energy dependence; lower energy photons will register later in the coincidence window than higher energy events, and this effect is known as time–walk\textsuperscript{76}. This section considers a list–mode acquisition system, and exploiting the ability to re–process the raw data to both correct for the time–walk in the detector, and substantially reduce the level of accidental coincidences seen. The results from this project were published in Nuclear Instruments and Methods in Physics Research Section A (a copy of this can be found in Appendix E).

9.3.2 Experimental Setup

A $\gamma$–$\gamma$ coincidence system was utilised for this work, as this method of detection focuses on coincidences (as opposed to anti–coincidences, which are
generally far more numerous). A $\gamma-\gamma$ system will therefore be particularly sensitive to the removal of the coincidence background, and the best candidate to demonstrate a substantial improvement in the resulting spectra.

The $\gamma-\gamma$ system consists of two large-area, planar HPGe detectors (model BE6530, from Canberra UK in Harwell, Oxfordshire) situated in a back-to-back configuration (both facing towards a central source). These are then enclosed within a Pb cave to minimise the effects of terrestrial radiation. Each detector signal is passed through a preamplifier (model 2002), and to a LYNXTM Digital Signal Processor, which controls all further amplification, pole-zero correction and digitisation of the pulse. A synchronisation cable runs between the the LYNXTM units to allow synchronisation of the clocks used to record the events in each detector, and data is collected in list-mode. A schematic of the system is shown below, in figure 9.23.

![Figure 9.23](image)

Figure 9.23: A schematic of the $\gamma-\gamma$ system, with the detector source separation set to 5 cm. The HPGe crystals are shown in green, surrounded with copper crystal holders and a copper cold finger (both in red). The aluminium casings are shown in light grey, with carbon fibre detector faces and the surrounding lead shield (outlined in dark grey).

### 9.3.3 Measurement & Analysis

Calibration sources were chosen to cover the 10-2000 keV energy range to fully characterise each system. Several single $\gamma$ emitters were used, as well as
a NIST (National Institute of Standards and Technology) traceable complex \( \gamma \) source. The isotopes that comprised these were \(^{241}\text{Am} \) (59.54 keV), \(^{109}\text{Cd} \) (88.03 keV), \(^{57}\text{Co} \) (122.06 keV), \(^{139}\text{Ce} \) (165.86 keV), \(^{113}\text{Sn} \) (391.68 keV), \(^{137}\text{Cs} \) (661.67 keV), \(^{54}\text{Mn} \) (834.84 keV), \(^{88}\text{Y} \) (898.04 and 1836.06 keV), \(^{65}\text{Zn} \) (1115.54 keV), and \(^{60}\text{Co} \) (1173.23 and 1332.49 keV). Analysis was completed using the post-processor to sort the data, and ROOT for matrix manipulation and peak searching/fitting in a similar fashion to that described in section 7.4. A typical delay spectrum is shown in figure 9.24.

![Graph](image-url)

Figure 9.24: An example coincidence delay spectrum from the \( \gamma-\gamma \) system. Three distinct regions are identifiable: the main coincidence peak at \( \sim 0 \) \( \mu \)s, the raised background either side of the peak, and the flat background from \( \pm 6 \) \( \mu \)s onwards. The flat background is due to random coincidences between the two detectors, and therefore roughly constant. The raised background is due to time–walk of low–energy events, and therefore dependent on the energy deposited in each detector. Note that this manifests itself either side of the main peak due to the symmetry of the system.

### 9.3.4 Correcting the Time–Walk of Coincident Events

The processing of coincidence signals has a small energy dependence, with low–energy events taking longer to process. This widens the coincidence peak seen in figure 9.24, causing the system to potentially miss coincident events, or include unnecessary ‘background’ coincident events. To correct for this effect, a source with simultaneous \( \gamma \) emission is required (any delay in the emission of multiple \( \gamma \) decays will artificially affect the position of the coincidence
peak), with relatively high–energy decays (to allow characterisation of the time–walk over a large energy range). Both of these requirements are met by $^{60}\text{Co}$ (see figure 9.25), which emits two $\gamma$’s (1173.23 and 1332.49 keV) within a picosecond. The time resolution of the LYNX™ units is limited to 0.1 $\mu$s, and a typical coincidence window may span up to $\sim$3 $\mu$s; by comparison the lifetime of $^{60}\text{Co}$ is negligible.

With a $^{60}\text{Co}$ source in place, data were acquired until the statistical error in each bin was reduced to less than 1%. Once the coincidences are extracted into an ($E_{\gamma_1}$, $E_{\gamma_2}$, $\Delta t$) matrix, an energy gate is placed around the 1332.49 keV decay in the primary detector (set to two times the full width at half maximum). The coincidence spectrum seen in the secondary detector can then be plotted against the time delay to produce figure 9.26.

To correct the time–walk at lower energies, the mean $\Delta t$ is taken in each energy bin, and the resulting plot fitted with a polynomial function. The parameters for these are extracted and the corrections reapplied to the data on an event–by–event basis. The uncorrected and corrected projections of the gated matrix are shown in figures 9.27 & 9.28 respectively, with the resulting delay spectrum in figure 9.29. A substantial improvement is seen in both the width and shape of the delay window, allowing a far more effective time gate to be applied when extracting coincidences. This also minimises statistical
9.3. Coincidence Corrections for Multi–Detector Systems

Figure 9.26: The \((E_\gamma, \Delta t)\) plot extracted from the matrix when gating on a 1332.49 keV \(\gamma\) decay in detector 1. As well as seeing the 1173.23 keV \(\gamma\) decay in coincidence, energy depositions from 1173.23 keV \(\gamma\)’s that Compton scatter out of the secondary detector are also seen. At low–energies, there is a trend where \(\Delta t\) between coincident events rises, which is the result of time–walk.

Figure 9.27: The projection of figure 9.26 fitted with a polynomial function. Note that the points are plotted as crosses, with the height proportional to the error in the mean. Above 1173.23 keV, the errors rapidly increase due to the reduced statistics for coincidences in this region.
uncertainty when attempting to remove the ‘coincidence background’.

Figure 9.28: The resulting projection of figure 9.26 once corrected for low–energy time–walk. This used the parameters from the functions fitted in figure 9.27 to construct the energy dependent time correction, before reprocessing the data and applying these on an event–by–event basis.

Figure 9.29: The delay spectrum for both uncorrected and corrected list–mode data. There is a substantial improvement in both the width and shape of the coincidence peak.

For a combined $^{137}$Cs & $^{60}$Co high–activity (>100 kBq) source, the mean reduction in gate width (and therefore background from accidental coincidences) after the time–walk correction was $18.4 \pm 0.4\%$, with $\sim 5.6\%$ of events being corrected. The time correction was found to be substantial up to $\sim 100$ keV, however non–zero correction factors were present up to $\sim 500$ keV. It was
not expected that the time–walk would have an effect at such high energies, however the time resolution of the LYNX™ acquisition module (100 ns) limits further inspection of the coincidence window structure. The improvements in the delay gate allow cleaner coincidence spectra to be created, as reducing the width of the gate directly excludes ‘background’ coincidence events.

### 9.3.5 Characterising and Removing the ‘Coincidence Background’

To extract coincidences from the delay spectrum, a region is selected that covers the peak whilst minimising the amount of random coincidences (see figure 9.30, pink area). The raised region of the coincidence background includes events that should be in the main peak, but are shifted away from this due to the time–walk of low energy signals. This cannot therefore be used as a representative background region, however the flat region in the time delay spectrum can be used (figure 9.30, light blue area). Events are extracted from this area and normalised to the width of the delay window. These events can then be subtracted from those extracted in the main coincidence gate. The blue hatched area represents the resulting coincidences once the coincidence background is subtracted from the main peak.

For the combined $^{137}$Cs & $^{60}$Co high–activity source, coincidences from the delay gate and background areas were extracted from the matrix, and used to create both a ‘normal’ and a ‘background subtracted’ coincidence gate projection. Note that no energy gates were used for this, and so the spectra contain all the coincidences seen in a detector when observing an event in the other. The resulting spectrum is plotted in figures 9.31 & 9.32 with figure 9.31 showing the complete spectrum, and figure 9.32 showing a series of zoomed spectra to highlight the differences between the two methods.

This method accounts for $\sim$100% of the accidental coincidences, however it only works if the background rate is high enough, and therefore representative of what is intermixed with the original delay gate. For the $^{137}$Cs & $^{60}$Co source, $16.6 \pm 0.7\%$ of events were removed from the coincidence window, three times more than during the time–walk correction. The spectrum shows ex-
9.3. Coincidence Corrections for Multi-Detector Systems

Figure 9.30: A coincidence delay spectrum from the $\gamma-\gamma$ system. The extracted coincidences are in the pink region, the coincidence background in the light blue region, and a representation of the resulting region (once the subtraction is made) within the blue hatched area.

Figure 9.31: The total and coincidence spectra for the $\gamma-\gamma$ system using the combined $^{137}$Cs & $^{60}$Co source. The major features in the coincidence spectrum are the backscatter peak ($\sim$250 keV), Compton edges ($\sim$500, 950, 1150, & 2300 keV), and the two major $\gamma$ decays in $^{60}$Co (1173.23 & 1332.49 keV), which would all be expected in the coincidence spectrum. Features that should not be present in the coincidence spectrum are the 661.66 keV $^{137}$Cs peak, and 2505.72 keV $^{60}$Co sum peak.
9.3. Coincidence Corrections for Multi–Detector Systems

Figure 9.32: A series of zoomed spectra for the three regions that show major differences between the standard gate and the background subtracted gate. The left plot focuses on the low–energy region (\(^{137}\)Cs x–rays), the middle plot on the 661.66 keV decay from \(^{137}\)Cs, and the right on the 2505.72 keV sum peak from \(^{60}\)Co.

Excellent suppression of false coincidences, which are most apparent as a 661.66 keV \(^{137}\)Cs peak (this is a singular \(\gamma\) emission and should therefore have no coincidence signature). Additional signatures were also removed or reduced; the \(^{60}\)Co sum peak at 2505.72 keV does not contribute to the corrected spectrum (by definition, if both \(^{60}\)Co emissions are seen in a single crystal, no other true coincident events would be expected), and a substantial amount of Cs x–rays were removed. As x–rays can be caused by impinging \(\gamma\) radiation from a variety of energies and sources, there will always be some additional number of x-rays that create a valid coincidence signature, and should not be excluded from the coincidence spectrum. No statistical difference could be found in the \(^{60}\)Co peaks or the additional features (such as the Compton edges), which confirms that the coincidence background subtraction is both valid and effective. Particular care has to be taken when selecting the regions for both the total and background subtraction gates; inhomogeneities throughout the delay window, (due to statistical fluctuations/time–walk/delayed coincidences), could cause the subtraction to be over–estimated or under–estimated. The background coincidences also show a similar energy profile to events in each
9.3. Coincidence Corrections for Multi-Detector Systems

detector, such that higher-energy events contribute to a smaller proportion of the spectrum than lower-energy coincidences. This can reduce the statistics available in the higher energy region, causing greater uncertainty when applying the background subtraction.

9.3.6 Summary

List-mode data acquisition has been utilised in conjunction with a high-efficiency $\gamma-\gamma$ coincidence system, allowing data collection whereby both the energetic and temporal information is retained for each recorded event. As this information is not lost during acquisition, the data can be re-processed multiple times to extract the coincidence information, correct for time-walk of low-energy events, and remove accidental coincidences from the projected coincidence spectrum.

The time-walk correction has resulted in a reduction in the width of the coincidence delay gate of $18.4\pm0.4\%$, and thus an equivalent removal of accidental coincidences. The correction factors applied to $\sim5.6\%$ of events up to $\sim500$ keV, however the author would like to note that while these corrections improved both the shape and width of the peak, further investigation was limited by the time resolution of the electronics. Future work is planned to investigate the delay gate using improved electronics that can resolve timestamps at the 10 ns level, as opposed to the 100 ns resolution possible in this work. As well as the aforementioned benefits arising from the use of an optimised decay window, the time-stamp correction is crucial for sources with low-energy decays (which may be missed by an uncorrected delay gate), and the collection of data that relies on accurate timing measurements.

By extracting both the delay gate and a representative ‘background’ region for the coincidences, a coincidence background-subtracted spectrum can be projected from the coincidence matrix, which effectively accounts for $\sim100\%$ of the accidental coincidences (these accounted for up to $16.6\pm0.7\%$ of the events seen during this work). It is important to note that this method only estimates the coincidences present in the spectrum (in contrast to the time-walk correction, which allows the elimination of events directly). This tech-
nique is obviously limited to radiation sources that contribute to a constant coincidence background, and sources where a coincidence background region can be extracted that is representative of this. Such sources would include coincidences arising from terrestrial and cosmic background radiation, and radioactive sources in which the half-lives are much greater than the length of the decay window. This correction is essential for accurate characterisation of the events seen in coincidence systems, as otherwise false coincidence signatures may be incorrectly interpreted.
Chapter 10

Conclusions & Outlook

10.1 Summary of Current Progress

The measurement of radioactive material is a process that aims to reliably and efficiently identify any isotopes present within the source matrix, and accurately quantify these to allow interpretation of a variety of factors. These may include safety concerns, likely doses that exposure may incur, and the origin of the isotopes that are present. For non-destructive testing, $\gamma$-spectroscopy provides the most effective measurement technique, as $\gamma$ emissions may be the only detectable radioactivity to escape the source matrix.

Typically, larger and more efficient detectors are employed to collect more radiation, and therefore enhance sensitivity of the system. There are however, limits to the detector sizes available, and to reduce the ‘background’ (anything that does not contribute towards a full photopeak signal) a far more nuanced approach is required. The aim of this thesis is therefore to bring advanced detection, optimisation, and analysis techniques (that are typically applied by large collaborations to highly focussed, multi-million pound detector arrays) to a laboratory based Compton suppressed $\gamma$ detection system. To improve the measurement process, all aspects of the detection and analysis procedure have been considered, and where possible, optimised.

The collection of coincidence data were typically achieved using an on-line veto, or List-mode files. The former allowed the creation of instant suppressed and unsuppressed energy spectra, however no temporal information
was stored. The latter consolidated all of the energy and time information into a file for post-processing, however the analysis of such files was a complex and time-consuming process that could not be extended to more advanced systems. A binary format and data processor was therefore developed for the List-mode data, which has improved the sorting speed (for producing coincidence matrices and energy spectra) by a factor of $\sim 20000$. This has removed the bottleneck for analysing such datasets, and benefits all current and future coincidence systems. New possibilities, such as the sorting of List-mode data in real time, provide the opportunity to study coincidence data in new ways, and dramatically increase throughput.

For the detector systems themselves, a combination of experimental work and Monte-Carlo simulation has been used to develop and validate computer models of the laboratory systems, which have then been utilised to improve the performance of all system components. Monte-Carlo models (using the GEANT4 toolkit) were initially used to reproduce the detector response of a NaI(Tl) detector system, with the geometry parameters tuned to reproduce the peak efficiencies for a range of photon energies. These models were then extended to accurately reproduce more complex HPGe detector systems, with all detector components, interaction effects, and source matrices fully simulated. This includes accurate reproductions of peak efficiencies, Compton continua, cascade summing effects, x-ray fluorescence, coincidence systems and radioactive sources with both complex geometries and compositions. Once validated, the models developed were used for routine detector characterisations, and proved particularly useful for the efficiency calibration of sources that were supplied in non-standard configurations.

Simulations of the detector shielding materials revealed that the majority of ‘background’ events originate from source photons that scatter out of the shielding, and into the primary crystal. This can be reduced by increasing the interior radius of the cave, and minimising the amount of low-Z material in the immediate vicinity of the detector. Liner thicknesses (which suppress x-ray fluorescence from the shielding material) were optimised to reduce the level of fluorescence seen by an order of magnitude, while simulations of the total lead
thickness showed that a reduction in the background would continue up to the full 200 mm of Pb considered. All events seen in a detector system that do not contribute to the full energy photopeak dramatically reduce the sensitivity of the system, and the information gained from these simulations will allow future designs to substantially increase the sensitivity of the detectors.

Studies into the primary detector considered multiple materials and designs, before identifying a planar HPGe detector as the most effective for environmental radiation. Utilising the validated simulations, the thickness of the detector was optimised for a number of key, low energy radionuclides. This was achieved by simulating a variety of spectra for each detector thickness, and then calculating the MDA in each case for all radionuclides of interest. The maximum sensitivity was achieved by minimising the amount of detector material, such that the low energy efficiency was maximised while minimising the interaction probability for higher energy $\gamma$ emissions.

A similar process identified a three–component, BGO based detector as the most effective design for a Compton suppression system, with the relative geometry and sizes of the components optimised to maximise the suppression achievable. The levels of continuum suppression are calculated to be an order of magnitude greater than for the current NaI(Tl) based system, with a reduction in the Compton continuum of up to $\sim$240 times for a common high–energy emitter such as $^{60}$Co. Such suppression would also apply to other high–energy emitters such as $^{140}$La, and be greatly increased for nuclides that decay via a cascade of radiation, such as $^{134}$Cs. By vastly reducing the continuum seen at low to medium energies, the systems sensitivity to lower energy $\gamma$ emitting isotopes will be greatly increased. The suppression achievable with such a system is only limited by the dead material in the immediate vicinity of the detector, including the cold finger and crystal holder.

Additional studies concerning the structure of the coincidence delay window were also conducted. These leveraged the ability to reprocess List–mode files, with routines developed to both correct for the time–walk of low–energy events, and substantially reduce the amount of accidental coincidences seen. To correct for low–energy time–walk, a $^{60}$Co source was used to probe the
nature of the coincidence window, and then calculate an energy dependent correction factor. The data could then be reprocessed with the correction applied on an event–by–event basis. This resulted in a reduction of the coincidence delay gate width by 18.4±0.4%, and thus an equivalent removal of accidental coincidences. The time–stamp correction is crucial for sources with low-energy decays (which may be missed by an uncorrected delay gate), and the collection of data that relies on accurate timing measurements. By extracting both the delay gate and a representative ‘background’ region for the coincidences, a coincidence background subtracted spectrum can be projected from the coincidence matrix, which effectively removes up to 100% of the accidental coincidences (these accounted for up to 16.6±0.7% of the events seen during the work). This technique is obviously limited to radiation sources that contribute to a constant coincidence background, and sources where a coincidence background region can be extracted that is representative of this. The correction is essential for accurate characterisation of the events seen in coincidence systems, as otherwise false coincidence signatures may be incorrectly interpreted.

10.2 Commercial/Industrial Applications of this Research

Typically, all detector/source characterisation, data processing and data analysis is completed using proprietary software. In the process of developing the proposed Compton suppression system, several commercial advantages have been realised.

Firstly, the software developed to efficiently store events, extract coincidences and produce both time and energy spectra can be applied to all systems in the laboratory. This allows analysis of coincidence data in the traditional sense, however with far larger datasets, and efficient extraction of coincidences as these are stored in a matrix. The modules developed for time–walk and accidental coincidence correction also represent a capability that was not previously available. Processing the data in this way also allows the study of
peak evolution during acquisition, something that could not easily be done
before. Not only does this software (and the ability to extend, modify and re-
purpose it) allow more complex systems to be employed, but it also requires
no proprietary software to run.

GEANT4 has proved an extremely valuable tool for simulating the re-
sponse of both scintillation and semiconductor based detector systems. Given
the range of effects that can be simulated, continued use of this Monte–
Carlo toolkit will greatly aid in future detector/shielding/system research
and design. For the current detectors in use, GEANT4 provides an alter-
native tool for the characterisation of these systems, including peak efficien-
cies and cascade summing correction factors. Again, this is achieved using
open–source (free) software, and provides far more flexibility when consider-
ing non–standard geometries.

Finally, the improved sensitivity achievable with the proposed detection
system will greatly reduce the time needed to quantify radionuclides within
sources, and allow detection of signals that would otherwise be obscured by
the continuum, and which can not currently be detected within a reasonable
timeframe. For example, the current Compton suppression system utilises
a \( \sim 26\% \) relative efficiency primary detector\cite{67}, and a veto system that can
reduce the continuum by up to a factor of 30–40 times\cite{88}. The proposed
detector configuration is designed to accommodate a \( \sim 48\% \) relative efficiency
primary detector\cite{67}, (with the thickness optimised for maximum sensitivity in
the required energy range), and a veto system that can suppress the continuum
by up to a factor of 240. As well as obtaining highly sensitive measurements,
the resulting reduction in acquisition times necessary for standard sources
would enable a greater sample throughput.

\section{10.3 Future Work}

Building upon the tools and designs developed as a result of this project,
several opportunities have been identified that may improve the detection
limits further for high sensitivity \( \gamma \)-spectroscopy systems.
Firstly, the development of the BGO veto designs with a reputable manufacturer will allow a high efficiency Compton suppression system to be built. This can be modular, with a lid, annulus, and base (or a plug, annulus, and base) implemented separately or as a single unit. The only significant challenge with the design is how extract the scintillation light from the base detector, however this may be solved using a light-guide.

The passive materials used in the construction of the primary HPGe detector are the last remaining dead layers within the immediate vicinity of the crystal. These include not only the dead layers within the crystal itself, but the crystal holder, electronics, cold finger and detector canister. This material both absorbs photons scattered from the crystal, and interacts directly with source photons, causing some to backscatter into the primary crystal. By reducing these materials, the continuum would be reduced in the primary detector, and any subsequent Compton suppression would be more effective. This could be particularly useful in the low-energy region, as reduced dead layers behind the crystal would greatly reduce the backscatter peak, and allow far more effective Compton suppression with a base detector (which is designed to veto photons that deposit a small amount of energy, and therefore scatter forward).

Finally, a combination of a Compton and cosmic veto would allow ultra-low backgrounds to be achieved. This would require a three-way veto system, which may be difficult to achieve using standard electronics. By utilising List-mode data acquisition however, coincidences can be extracted during post-processing with separate delay gates set between the primary detector and each respective veto detector. This system would generate a huge amount of data, however by utilising the processing techniques described in this thesis, extraction and analysis of coincidences from such a combination of detectors is now possible.
Appendix A

Publication List

A.1 Journal Papers

DOI 10.1007/s10967-011-1362-x

DOI 10.1007/s10967-012-1811-1

DOI 10.1007/s10967-012-2203-2

DOI 10.1007/s10967-013-2572-1

6. Improving the effectiveness of a low-energy Compton suppression system, R.Britton, J.L.Burnett, A.V.Davies, P.H.Regan (2013), Nucl Instrum Meth A
   DOI 10.1016/j.nima.2013.06.111

   DOI 10.1007/s10967-014-3029-x

   DOI 10.1016/j.jenvrad.2014.02.018

   DOI 10.1016/j.nima.2014.05.113

    DOI 10.1016/j.nima.2014.09.054

A.2 Contributing author

   DOI 10.1103/PhysRevC.87.014323

2. Li-7 induced reactions for fast-timing with LaBr3:Ce detectors (2012), AIP Conf. Proc.
   DOI: 10.1063/1.4764210

4. Precision Lifetime Measurements Using LaBr3 Detectors With Stable and Radioactive Beams (2013), EPJ Web of Conferences. DOI:10.1051/epjconf/20136301008


6. Germanium–gated $\gamma$–$\gamma$ fast–timing in very exotic fission fragments using FATIMA in combination with EXOGAM at the Institut Laue Langevin (2014), Nucl Instrum Meth A. DOI:10.1016/j.nima.2014.06.004


A.3 Conference papers

Appendix B

Presentation List

1. ‘Compton Suppression Systems for Radiological Analysis’ – November 2010 – Materials Science Conference, AWE, UK

2. ‘Designing a Low Background Compton Suppression System’ – August 2011 – Nuclear Physics Summer School, St. Andrews, UK


5. ‘Next Generation Detection Systems for Radioactive Material Analysis’ – March 2013 – Nuclear Data for Science and Technology (ND2013), New York, USA

6. ‘List-Mode’ data acquisition and analysis for a Compton Suppression system’ – April 2013 – IOP Nuclear Physics Conference, York, UK

7. ‘GEANT4 applications for the characterisation of HPGe detectors’ – May 2013 – IOP half-day meeting on GEANT4 in Nuclear Physics, Manchester, UK

8. ‘Monte Carlo simulations of γ–spectroscopy systems’ – January 2014 – NPL/Surrey Nuclear Data & Gamma–ray Spectrometry Array Meeting, University of Surrey, UK

10. ‘Nuclear data and modelling for monitoring compliance with the Comprehensive Test Ban Treaty’ – October 2014 – Nuclear Data: Current Measurements, Uncertainties, Applications and Needs, National Physical Laboratory, UK
Appendix C

CINDERELLA Cascade
correction factors
### Table C.1: A summary of the correction factors calculated for the CINDERELLA source, with ‘G4’ representing GEANT4 values, and G2K representing GENIE 2000\textsuperscript{TM} derived values. Some correction factors were not calculated in the Canberra software due to those peaks not being present in the preliminary analysis. Errors in the calculated correction factors are ± 6%, and were dominated by the variability in the source geometry/composition.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy (keV)</th>
<th>Correction Factor</th>
<th>Isotope</th>
<th>Energy (keV)</th>
<th>Correction Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{140}$Ba</td>
<td>132.69</td>
<td>0.943</td>
<td>$^{140}$La</td>
<td>487.02</td>
<td>0.741</td>
</tr>
<tr>
<td>$^{140}$Ba</td>
<td>162.66</td>
<td>0.932 0.894</td>
<td>$^{140}$La</td>
<td>751.64</td>
<td>0.870 0.878</td>
</tr>
<tr>
<td>$^{140}$Ba</td>
<td>304.85</td>
<td>0.764 0.787</td>
<td>$^{140}$La</td>
<td>815.77</td>
<td>0.982 0.981</td>
</tr>
<tr>
<td>$^{140}$Ba</td>
<td>423.72</td>
<td>0.933 0.962</td>
<td>$^{140}$La</td>
<td>867.85</td>
<td>0.865 0.878</td>
</tr>
<tr>
<td>$^{140}$Ba</td>
<td>437.58</td>
<td>0.992 0.966</td>
<td>$^{140}$La</td>
<td>919.55</td>
<td>1.034 0.985</td>
</tr>
<tr>
<td>$^{140}$Ba</td>
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<td>$^{140}$La</td>
<td>925.19</td>
<td>0.846 0.878</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>475.37</td>
<td>0.673</td>
<td>$^{140}$La</td>
<td>1596.21</td>
<td>0.772 0.809</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>563.25</td>
<td>0.668 0.727</td>
<td>$^{140}$La</td>
<td>2347.88</td>
<td>1.333 1.252</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>569.33</td>
<td>0.650 0.708</td>
<td>$^{140}$La</td>
<td>2521.4</td>
<td>1.116 1.088</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>604.72</td>
<td>0.783 0.816</td>
<td>$^{99}$Mo</td>
<td>40.58</td>
<td>0.666 0.726</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>795.86</td>
<td>0.790 0.811</td>
<td>$^{99}$Mo</td>
<td>140.51</td>
<td>0.880 0.798</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>801.95</td>
<td>0.679 0.726</td>
<td>$^{99}$Mo</td>
<td>181.07</td>
<td>0.868 0.791</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>1038.61</td>
<td>0.862</td>
<td>$^{99}$Mo</td>
<td>366.42</td>
<td>0.692</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>1167.97</td>
<td>1.188 1.213</td>
<td>$^{99}$Mo</td>
<td>739.5</td>
<td>0.740 0.769</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>1365.19</td>
<td>1.326 1.298</td>
<td>$^{99}$Mo</td>
<td>777.92</td>
<td>0.731</td>
</tr>
<tr>
<td>$^{132}$I</td>
<td>262.9</td>
<td>0.507</td>
<td>$^{147}$Nd</td>
<td>91.11</td>
<td>0.980 0.989</td>
</tr>
<tr>
<td>$^{132}$I</td>
<td>284.9</td>
<td>0.628</td>
<td>$^{147}$Nd</td>
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<td>0.679</td>
</tr>
<tr>
<td>$^{132}$I</td>
<td>505.79</td>
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<td>$^{147}$Nd</td>
<td>196.64</td>
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</tr>
<tr>
<td>$^{132}$I</td>
<td>522.65</td>
<td>0.635 0.692</td>
<td>$^{147}$Nd</td>
<td>275.37</td>
<td>0.626 0.657</td>
</tr>
<tr>
<td>$^{132}$I</td>
<td>630.19</td>
<td>0.615 0.676</td>
<td>$^{147}$Nd</td>
<td>319.41</td>
<td>0.670 0.723</td>
</tr>
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<td>$^{132}$I</td>
<td>650.5</td>
<td>0.554 0.574</td>
<td>$^{147}$Nd</td>
<td>398.16</td>
<td>0.802 0.786</td>
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<tr>
<td>$^{132}$I</td>
<td>667.71</td>
<td>0.697 0.743</td>
<td>$^{147}$Nd</td>
<td>410.48</td>
<td>2.308</td>
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<tr>
<td>$^{132}$I</td>
<td>727</td>
<td>0.618 0.711</td>
<td>$^{147}$Nd</td>
<td>439.9</td>
<td>1.000 0.915</td>
</tr>
<tr>
<td>$^{132}$I</td>
<td>772.6</td>
<td>0.673 0.737</td>
<td>$^{147}$Nd</td>
<td>489.24</td>
<td>1.333</td>
</tr>
<tr>
<td>$^{132}$I</td>
<td>809.5</td>
<td>0.568 0.663</td>
<td>$^{147}$Nd</td>
<td>531.02</td>
<td>0.959 1.008</td>
</tr>
<tr>
<td>$^{132}$I</td>
<td>812</td>
<td>0.645 0.699</td>
<td>$^{147}$Nd</td>
<td>594.8</td>
<td>1.438</td>
</tr>
<tr>
<td>$^{132}$I</td>
<td>954.55</td>
<td>0.667 0.711</td>
<td>$^{147}$Nd</td>
<td>685.9</td>
<td>1.118 1.014</td>
</tr>
<tr>
<td>$^{132}$I</td>
<td>1136</td>
<td>0.744 0.833</td>
<td>$^{103}$Ru</td>
<td>294.96</td>
<td>0.991 1.000</td>
</tr>
<tr>
<td>$^{132}$I</td>
<td>1398.57</td>
<td>0.677 0.719</td>
<td>$^{103}$Ru</td>
<td>497.09</td>
<td>0.998 1.000</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>109.42</td>
<td>0.543</td>
<td>$^{103}$Ru</td>
<td>610.33</td>
<td>0.984 1.000</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>131.12</td>
<td>0.480</td>
<td>$^{132}$Te</td>
<td>49.72</td>
<td>0.754 0.799</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>241.93</td>
<td>0.603</td>
<td>$^{132}$Te</td>
<td>111.76</td>
<td>0.665</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>266.54</td>
<td>0.598</td>
<td>$^{132}$Te</td>
<td>116.3</td>
<td>0.626</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>328.76</td>
<td>0.709 0.725</td>
<td>$^{132}$Te</td>
<td>228.16</td>
<td>0.820 0.750</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>432.49</td>
<td>0.753 0.725</td>
<td>$^{132}$Te</td>
<td>228.16</td>
<td>0.820 0.750</td>
</tr>
</tbody>
</table>
Appendix D

CINDERELLA source activity

The activity of additional radionuclides is shown here, as well as a comparison to abundances calculated using Canberra’s GENIE 2000™ software.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>NPL Calibration value (Bq/m³)</th>
<th>Calculated Activity</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>GEANT4 (Bq/m³)</td>
</tr>
<tr>
<td>¹⁰⁹ᵐAg</td>
<td></td>
<td>8.3 ± 2.0 × 10⁻⁶</td>
</tr>
<tr>
<td>²⁴¹Am</td>
<td></td>
<td>2.9 ± 1.2 × 10⁻⁵</td>
</tr>
<tr>
<td>¹⁴⁰Ba</td>
<td>4.77 ± 0.10 × 10⁻³</td>
<td>4.43 ± 0.26 × 10⁻³</td>
</tr>
<tr>
<td>¹⁴¹Ce</td>
<td>3.13 ± 0.06 × 10⁻³</td>
<td>3.05 ± 0.30 × 10⁻³</td>
</tr>
<tr>
<td>¹⁴⁴Ce</td>
<td>4.97 ± 0.15 × 10⁻⁴</td>
<td>4.84 ± 0.47 × 10⁻⁴</td>
</tr>
<tr>
<td>¹³⁴Cs</td>
<td>1.383 ± 0.027 × 10⁻³</td>
<td>1.28 ± 0.16 × 10⁻³</td>
</tr>
<tr>
<td>¹³⁷Cs</td>
<td>2.22 ± 0.07 × 10⁻⁵</td>
<td>2.23 ± 0.31 × 10⁻⁵</td>
</tr>
<tr>
<td>¹³¹I</td>
<td></td>
<td>4.29 ± 0.58 × 10⁻⁴</td>
</tr>
<tr>
<td>¹³²I</td>
<td></td>
<td>1.0 ± 0.6 × 10⁻³</td>
</tr>
<tr>
<td>⁴⁰K</td>
<td></td>
<td>2.07 ± 0.35 × 10⁻⁴</td>
</tr>
<tr>
<td>¹⁴⁰La</td>
<td>0.87 ± 0.03 × 10⁻³</td>
<td>0.84 ± 0.15 × 10⁻³</td>
</tr>
<tr>
<td>⁹⁹Mo</td>
<td>3.81 ± 0.12 × 10⁻⁴</td>
<td>4.24 ± 0.38 × 10⁻⁴</td>
</tr>
<tr>
<td>⁹⁵Nb</td>
<td>8.23 ± 0.61 × 10⁻⁴</td>
<td>7.4 ± 0.4 × 10⁻⁴</td>
</tr>
<tr>
<td>⁹⁵ᵐNb</td>
<td></td>
<td>2.9 ± 1.0 × 10⁻⁵</td>
</tr>
<tr>
<td>¹⁴⁷Nd</td>
<td>1.48 ± 0.10 × 10⁻³</td>
<td>1.52 ± 0.07 × 10⁻³</td>
</tr>
<tr>
<td>¹⁰³Ru</td>
<td>7.72 ± 0.17 × 10⁻⁴</td>
<td>7.48 ± 0.41 × 10⁻⁴</td>
</tr>
<tr>
<td>¹³²Te</td>
<td>4.67 ± 0.14 × 10⁻⁴</td>
<td>4.89 ± 0.26 × 10⁻⁴</td>
</tr>
<tr>
<td>⁹⁵Zr</td>
<td>2.476 ± 0.048 × 10⁻³</td>
<td>2.53 ± 0.10 × 10⁻³</td>
</tr>
</tbody>
</table>

Table D.1: A summary of the radioisotopes detected in the CTBT CINDERELLA source, and their abundances. The calculated activity was determined solely via Monte-Carlo methods, using both GEANT4 calculated Cascade Summing Corrections and peak efficiency curve. This is compared to a previous study undertaken using the Canberra GENIE 2000™ software, and the (time-corrected) activity values determined during the initial calibration at NPL (National Physical Laboratory). Nuclide activities were also corrected to account for the feeding of different isotopes from other decays (such as ¹⁴⁰Ba, which decays into ¹⁴⁰La).
Appendix E

Attached papers

The following appendix includes (for reference) the papers currently published as a result of the project. Please see the relevant journal websites to obtain a full copy.
Compton suppression systems for environmental radiological analysis

R. Britton

Received: 20 July 2011 / Published online: 31 July 2011
© Akadémiai Kiadó, Budapest, Hungary 2011

Abstract Compton suppression (CS) has increased the sensitivity of gamma spectroscopy systems tenfold, and is routinely used in laboratories for environmental analysis and the monitoring of the CTBT. There are several different techniques available, and many more variables to consider when designing or optimising a CS system. An overview and discussion of these is presented here.

Keywords Compton suppression · Coincidence · Anti-coincidence · Veto · Gamma spectroscopy

Introduction

Gamma Spectroscopy has become an invaluable tool for the non-destructive identification of nuclides. Typically, high purity germanium (HPGe) detectors are employed across a wide range of energies, allowing both the identification of radionuclides and an estimate of their abundances [1]. Applications for γ-spectroscopy include food testing [2], neutron activation analysis [3, 4], fundamental physics research [5–8] and environmental analysis [9, 10] for the monitoring of the comprehensive nuclear test ban treaty (CTBT) [11].

In such detector systems, the main source of noise is the Compton continuum arising from photons that scatter out of the detector, depositing only a fraction of their energy. This is a particularly acute problem for the analysis of transuranic nuclides (TRU) [12], as the low energy photons are swamped by the continuum. Other important sources of background include cosmic events, the materials used to construct the detector/laboratory and radon gas. These can be minimised with a cosmic veto and appropriate shielding, however Compton scattered events originate in the detector itself, requiring a more subtle approach. There are many methods to (partially) eliminate or recover these events, known as Compton suppression (CS) systems, and their evaluation will be the subject of this review.

Theory

Ideally, a γ-spectrometer would have a response function consisting of only the full photopeak energies ($E_p$), with no continuum or background. In the real world however, spectra often have very prominent continua, often masking important spectral information [13].

Photons can interact via the photoelectric effect, Compton scattering or pair production, and the probability of each interaction type is energy dependent (see Fig. 1). For environmental analysis, the energy range of interest is between 50 keV and 3 MeV, and so Compton scattering is the dominant mechanism.

The cross-section for Compton scattering ($\sigma_{\text{compton}}$) varies linearly with $Z$ of the detector material, while the cross-section for photoelectric effect varies as $\sigma_{\text{photo}} \sim Z^{4.5}$, and for pair production as $\sigma_{\text{pair}} \sim Z^2$ [15]. Therefore, choice of a high Z material will improve both the full photopeak efficiency and the proportion of photons stopped in the detector.

Several methods are employed to quantify the amount of continuum reduction, often with interchangeable names.
and meanings. The Peak to Count (P/C) ratio is most commonly used, and is defined as the ratio of the counts in the highest photopeak channel to the counts in a typical channel of the Compton continuum. Normal values range from 50 to 75 for unsuppressed coaxial HPGe detectors [13]. The Compton suppression factor (CSF) is the ratio of P/C for unsuppressed and suppressed spectra, and takes into account the reduction in photopeak efficiency as well as the suppression of the continuum.

Methods

Anti-coincidence mode

As crystals are a finite size, scattered photons may escape the detector before depositing their full energy. These events can be preferentially rejected if the escaped photons are detected in coincidence by a surrounding guard detector (GD).

In Fig. 2 photons travel down a heavy metal collimator to prevent direct interaction with the GD. These are incident on the primary detector (PD) in the centre, and a substantial portion of these are scattered back out into the GD. If the GD is an active volume it can generate a signal to prevent any coincident signals from being recorded in the PD. This anti-coincidence setup is the most common for CS systems, and has been studied extensively [3, 16–20], with common suppression factors of 4.1–12.1 for $^{60}$Co [3, 16, 18, 19] peaks, and 3.9–12.7 for the $^{137}$Cs [3, 16–18, 20] peak (Fig. 3).

Reductions of the Compton continuum of up to 85 have also been reported at the Compton edge [21, 22], with significant variance in each systems effective energy range. These differences are primarily due to their geometries and build materials, hence the popularity of Monte-Carlo simulations to optimise CS systems before production.

By vetoing any event that occurs in coincidence with a signal from the GD, some photopeak counts will be lost due to chance coincidences with other Radiation. High multiplicity gammas will also be partially suppressed as they can not be discriminated from normal escape events. Nuclei with such cascade suppressions may gain little or no benefit from CS systems as the continuum reduction is cancelled out by the reduction in photopeak [21]. High count rates can also detrimentally affect CS systems, as the increased dead time and higher flux increases the rate of chance coincidences [23, 24].

A summary of achieved suppression ratios (including different methods) is given in Table 1.
Sum-coincidence mode

Most Compton continuum events are caused by the single scattering of an incoming photon, which subsequently escapes the detector. Full energy events typically involve multiple scatterings (with escape from the detector significantly less likely with each further interaction) [13]. Therefore the continuum can be suppressed by requiring the event to register in two adjacent segments/detectors. This provides excellent suppression at the expense of efficiency; Palms [29] achieved a 25 fold reduction in the continuum with a 5 fold reduction in photopeak, resulting in a CSF of 4–5.

Sum-coincidence counting is most effective when used in conjunction with other techniques (such as anti-coincidence), as this allows additional statistics to be gleaned when working with low count rates or rare events. Several nuclear physics institutes now employ large, Compton suppressed gamma arrays [7, 15, 30, 31] and significant effort is being invested in optimising these setups for a variety of experiments.

Pair spectroscopy

This approach involves recording only the double escape peak (at $E_{\gamma} \sim 1.022$ MeV), therefore limiting it to energies where a significant proportion of interactions will involve pair production. For an event to be recorded in Pair Spectroscopy, the initial event must be detected in the PD, and both the 511 keV annihilation photons must be detected in the GD. This approach sacrifices efficiency but can produce very clean spectra. A small residual continuum is generally caused by interactions near the edge of the crystal (where the likelihood of scattering out is high), and so can be significantly reduced by using pulse shape analysis to discard slow rising charge pulses [32].

Pulse shape analysis (PSA)

The pulse shape from HPGe detectors depends on the charge collection time, which is determined by the initial position of the $\gamma$-ray interaction. Traditional methods of CS rely on ultra-low background techniques and multiple detectors to veto or record an event. PSA uses the information in each charge pulse to infer both where the event occurred in the detector, and the likelihood that it is a full energy or escape event.

Research has focused on the general characteristics of the charge pulse such as rise time and whether the shape was that of a single site or multiple site event [33, 34], although recent efforts also involve simulations [28, 35, 36] to determine likely pulse shapes for each type of interaction. These are then compared to the experimental data using chi-squared analysis to determine the interactions position and energy, at which point an algorithm can be applied to discriminate escape events from those where the full energy is deposited. The improvement of such algorithms is ongoing, with initial simulations for the MAJORANA experiment

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Table 1: A summary of suppression ratios for various experiments

<table>
<thead>
<tr>
<th>Author</th>
<th>CSF</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>100</td>
</tr>
<tr>
<td>Cooper (A) [22]</td>
<td>–</td>
<td>5.2</td>
</tr>
<tr>
<td>Beetz (A) [25]</td>
<td>–</td>
<td>15.0</td>
</tr>
<tr>
<td>Aarts (A) [26]</td>
<td>7.2</td>
<td>–</td>
</tr>
<tr>
<td>Moszynski (A) [18]</td>
<td>12.1</td>
<td>2.5</td>
</tr>
<tr>
<td>Masse (A) [21]</td>
<td>–</td>
<td>23.0</td>
</tr>
<tr>
<td>Mauherhofer (A) [3]</td>
<td>8.7</td>
<td>–</td>
</tr>
<tr>
<td>Lin (A) [27]</td>
<td>–</td>
<td>18.8</td>
</tr>
<tr>
<td>de Voigt (A) [16]</td>
<td>4.0</td>
<td>23.0</td>
</tr>
<tr>
<td>Fukuda (A) [19]</td>
<td>6.0</td>
<td>13.2</td>
</tr>
<tr>
<td>Peerani (A) [17]</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Duchene (A, S) [8]</td>
<td>–</td>
<td>2.0</td>
</tr>
<tr>
<td>Schmacker (A, S) [7]</td>
<td>–</td>
<td>7.8</td>
</tr>
<tr>
<td>Pearson (PSA) [28]</td>
<td>–</td>
<td>7.5</td>
</tr>
</tbody>
</table>

CSF is defined earlier, and the additional data is the ratio of counts in the suppressed to unsuppressed spectra at specific energies. All data is for $^{60}$Co spectra. Datasets followed by an (A) used anti-coincidence techniques, those with (S) used sum-coincidence, and those with (PSA) digital techniques such as pulse shape analysis or gamma ray tracking.

a Simulated or calculated data
showing a 99% reduction of background while preserving 98% of their signal in a highly specialised array [37].

For ‘laboratory scale’ detectors, the potential benefits of PSA include cost, weight and portability, however, the performance of such systems is yet to match those using a multi-detector set-up [35, 36]. Large arrays such as EXOGAM [5], TIGRESS [38], MINIBALL [39] and DESPEC [40], all utilise Clover detectors in conjunction with PSA to estimate the position of the first interaction, allowing improved resolution and statistics, and future arrays plan to use a pure $4\pi$ HPGe sphere, maximising geometric efficiency while digitally tracking each $\gamma$-ray to determine its complete interaction history [41–43]. Generally such advances filter down to smaller systems with time, and so it should be expected that digital PSA will play a key role in future gamma spectroscopy systems at the limits of detection.

Design considerations

Traditionally, CS systems used HPGe as the PD, with an annulus of NaI(Tl) [9, 13, 21] however recent developments in computer modelling, scintillator materials and digital pulse processing have prompted research into other configurations.

Primary detectors

HPGe is almost exclusively used as the PD due to its excellent energy resolution and high Z value. The major drawback (apart from the cost), is the need for liquid nitrogen cooling. This can restrict the geometry, and therefore the efficiency that the GD can achieve, however specifically designed cooling apparatus can minimise this disruption, and mechanically cooled HPGe detectors are now widely available.

While few materials can match the resolution of HPGe ($<0.2\%$), LaBr(Ce) detectors can approach $3\%$, a substantial improvement over NaI(Tl) (6–7%) and BGO ($<16\%$) [13]. This is also achieved at room temperature, allowing a more effective GD than can be achieved with a HPGe PD.

LaBr(Ce) detectors have excellent light yield and timing properties in addition to their resolution. Kulisek [44] calculated a 6.6 factor improvement in the detection limit for $^{239}$Pu over a $^{137}$Cs continuum for a LaBr(Ce) with a NaI(Tl) shield. There should also be room for improvement on this value as the researchers note that low energy photons were escaping out of the top of the simulated detector.

HPGe detectors reached volumes of 300 cm$^3$ in the early 1990s, but the drive for bigger detectors was being hampered by ballistic deficit, neutron damage and Doppler broadening. This led to the development of a new type of composite Clover detector, improving both the efficiency and resolving power of large HPGe arrays [8, 45, 46]. Clover detectors use several smaller crystals in the same cryostat, each of which are further segmented electronically. This improves both the timing and the granularity, decreasing the Doppler broadening effect and improving resolution. Such an arrangement also permits sum-coincidence techniques to improve the P/C ratio, and anti-coincidence processing with a BGO shield to reduce the continuum further. Many CS schemes have been modelled for a variety of energies and geometries [30], clearly showing the adaptability of such detector systems (as only the processing algorithms need to be modified for different experiments).

Alternative materials for the PD include CdZnTe and Si(Li). CdZnTe has both high atomic numbers and a large bandgap (1.52 eV) allowing operation near room temperature, and is 4–5 times more effective for photoelectric absorption when compared to HPGe. Unfortunately, the crystals can only be grown fairly small before charge collection becomes a problem. With electronic pulse and 3D correction techniques, resolutions of 0.76% at 662 keV have been achieved [47], and preliminary work into a $3 \times 3$ array of CdZnTe crystals has shown that CS is viable, if not as efficient as in ‘standard’ setups [48]. Si(Li) is primarily of interest in the 5–50 keV region, as the lower Z value (Z = 14) when compared to Ge (Z = 32) reduces the probability of photoelectric absorption by a factor of 50, and the size of the crystals are limited by the Lithium drift process [13]. Prussin [49] report continuum reductions of up to 31 in the 5–50 keV range, and factors of 10 in the 90–300 keV range.

Guard detectors

NaI(Tl) is an effective low-cost active shield, and has excellent properties where space is not restricted. If this is not the case, BGO is often used [3, 16, 18, 31, 36] as it has three times the linear attenuation coefficient at 500 keV [50], allowing for substantial space savings and/or performance improvements (albeit at higher cost). NaI(Tl) does have a higher resolution and light yield than BGO [13, 51], and these limitations may be of importance for systems that also perform $\gamma$–$\gamma$ coincidence work.

Liquid Argon (LAr) has also been used as a GD, as it can be in direct contact with the Ge crystal and provide the necessary cryogenic cooling [52–54]. Usually for such experiments photo-multiplier-tubes are used to collect the scintillation light [53], however Silicon photo-multipliers have also been employed [52]. Continuum reductions of up to 17 were achieved at 2 MeV, however the light yield and efficiency of such a system was very low.
Geometry's

Geometry is perhaps the most important aspect of CS system design. Inadequate shielding or insufficient detector material can dramatically alter the performance of the system, and these issues are most commonly addressed with MC modelling and optimisation studies [7, 26, 55, 56]. The path of scattered photons is highly energy dependant, and therefore the geometry of the detectors should be tailored to the materials being analysed (Fig. 4).

Monte Carlo (MC) simulations using MCNP [12, 44], GEANT [7, 8, 55], and FLUKA [53] have all been used to optimise the geometry of existing and proposed systems. Aarts [26, 57] used MC calculations to improve the solid angle of their detector. This was achieved by sacrificing shielding at the front of the guard so that the source was closer to the detector, resulting in a 20–32% improvement in the CS ratio. Scates [12] considered many variables, including GD thickness in front of and behind the PD, and the thickness of the aluminium can surrounding the LaCl₃(Ce) crystal, with the latter found to be the most sensitive to small parameter changes.

Mauerhofer [3] used a planar and two coaxial HPGe PD’s, with BGO and CsI(Tl) as GD’s (similar to Fig. 2). The source was also outside the CS system and collimated with a Pb ‘neck’. Peak to Total ratios improved by roughly 50% at 320 keV. For higher energies, this improvement increased to 200–300% for the suppressed system, which may be indicative of the effectiveness of the BGO when the photons are predominantly forward scattered, and the inefficiency of the geometry for low energy (backscattered) photons. Another advantage of this setup is the source being outside the CS system, as high count rates can be mitigated by simply moving the source further away. A major disadvantage of the multi-detector system is the increased dead material for the photons to be scattered and absorbed by.

Masse [21] use a NaI(Tl) ‘plug’ which sandwiches the source between itself and the PD. This has the advantage of substantially improving the efficiency of the GD for low energy photons, although detector efficiency had to be artificially limited to avoid counting rates that were too high.

Electronics/timing circuits

The electronics systems used for pulse discrimination and vetoing vary greatly in cost and complexity. Digital ‘all-in-one’ systems have also become available as an alternative to traditional analogue systems, and each will be briefly described below.

Analogue electronics

Analogue electronics are used for the shaping, delay and discrimination of event signals from the detector. Anti-coincidence setups achieve this by blocking the analogue to digital converter (ADC) from registering the event if a coincident event is detected in the GD.

A study by Canberra [50] evaluated three different analogue electronics setups, ranging from basic systems to those that are more complex. The most basic setup utilised logic pulses from Canberra 2002 Amplifiers which were fed into a Canberra 2040 Coincidence unit. If the two pulses were received within a set resolving time an output was generated, which was shaped in a delay and gate unit and prevented the ADC from recording. The more complex system used timing filter amplifiers (TFA’s) and constant fraction discriminators (CFD’s) for a greater time resolution, before delaying and shaping the pulse that would prevent the ADC from recording. Both systems performed similarly, with the two achieving suppression ratios of 2.37 and 2.45 respectively, suggesting that ultra-fast electronics may not be necessary for some common radioanalytical applications. An excellent guide on how to setup analogue electronics is available from [58].

Digital electronics

Traditionally, analogue electronics were used for the shaping and timing of incoming charge pulses, however, digital systems replace these with numerical analysis of the incoming charge pulse to determine its properties. Such analysis however, can only take place once the signal is digitised using an ADC [59].

The ADC must be accurate enough and have a sufficient sampling rate to preserve the information in the pulse shape, and is the main limiting factor (along with having an
efficient algorithm to assign/discriminate events) for a
digital system. Pearson [28] found that a sampling the pulse
every 5 μs (200 kHz) was enough to give adequate resolu-
tion and minimise the data rate, although this will vary
across different detectors.

Digital systems also offer improved stability as well as
performance over analogue circuity. They have a wider
range and finer steps of adjustment, can tune the peak
processing to individual pre-amplifiers and compensate for
ballistic deficit [60]. The pulse shape is also preserved
when digitised, and therefore no longer subject to distor-
tions such as electronic noise and temperature gradients.
Digital low frequency rejector (LFR) filters have also been
used to reduce microphonic noise from mechanically
cooled HPGe’s, with resolutions improved to 0.17% at
$^{60}\text{Co}$ [61].

Other considerations/shielding

The amount of dead material (material that is passive, i.e.
the end caps, outer contact, support materials etc.) between
the primary and GDs should be minimised, as this can
absorb escaping photons and therefore reduce the effec-
tiveness of a CS system. Often, n-type contacts are used
with HPGe detectors as these can be as thin as 0.3 μm,
whereas p-types are usually around 600 μm [62]. Aarts
[57] report a 32% improvement in suppression with a
fivefold reduction in dead layer.

Other research into improving the signal to noise ratio
includes carefully choosing design materials with low back-
grounds, reducing the background from the environment with
shielding, cosmic veto systems and controlling the atmo-
sphere to prevent contamination with radon gas [31, 55, 63].

For lead shielding, the optimum amount is around 15 cm
[64], as above this, interaction with cosmic events will
increase the background. Low background (old) lead can
also be used to reduce the $^{210}\text{Pb}$ content ($t_{1/2} = 22$ years),
and an internal liner of copper, tin or cadmium can attenuate
the lead X-rays. For very low background systems, cadmium
is not generally used as it has a high cross-section for neu-
trons from cosmic Radiation. Also, the use of a liner will
increase the background continuum at higher energies
($>100$ keV), leaving an important choice in where the low-
background energy range will be [63].

The hadronic component of Cosmic Radiation can be
shielded by the building and lead, and a significant portion
of the muon contribution vetoed with plastic scintillators in
a similar fashion to the Compton continuum [55]. Reduc-
tions of 60% have been reported for the muon component
up to 3 MeV [64], however to reduce this at higher energies
may require hundreds or thousands of meters of overburden
[65], as found in several underground laboratories.

Radon contamination can be reduced by filling the system with an inert gas such as nitrogen. Parus [65] report
a factor of 12 reduction in the peaks of $^{214}\text{Pb}$ and $^{214}\text{Bi}$ with
such controls, but the $^{220}\text{Rn}$ daughters were only slightly
decreased, giving an overall factor of 2 reduction.

Conclusions

The majority of CS setups utilise anti-coincidence tech-
niques, and a factor of 8–12 improvement in the P/C ratio
should be possible with a well designed system. The optimisation of a CS systems geometry is critical, and
Monte Carlo simulations provide an excellent tool for this.

Improvements in detector materials may also benefit CS
systems, with the advent of CdZnTe and LaBr$_3$ for room
temperature applications, and improvements in cooling
mechanisms for HPGe detectors. Segmented, multi-crystal
HPGe detectors may also improve the sensitivity of CS
systems when combined with sum-coincidence techniques
and PSA, although to realise these benefits, digital pulse
processing or fast analogue electronics may be required, as
well as significant research into appropriate algorithms and
processing techniques.

Finally, the sensitivity of CS systems may also be
improved further by employing a low-background, well
shielded setup and a cosmic veto. As shown here, there are
many important variables to consider when designing a CS
system, each perturbing the final systems performance in a
variety of ways. Clearly, the importance of Monte Carlo
simulations to design and optimise any system must not be
overlooked.

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research.

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Preliminary simulations of NaI(Tl) detectors, and coincidence analysis using event stamping

R. Britton · J. Burnett · A. Davies · P. H. Regan

Abstract This paper discusses preliminary work into the modelling and processing of coincidence measurements, for which a Monte-Carlo simulation and a post-processing program have both been developed. In the current work, a GEANT4 code is used to simulate a pair of NaI(Tl) scintillators, which are used experimentally to both develop the post-processing program and validate the GEANT4 model. This is found to be accurate to within 9 % at a confidence level of 93.0 % for energies from 30 to 3,000 keV.

Keywords List mode · Coincidence · GEANT4 · Monte Carlo simulations

Introduction

Gamma (\(\gamma\)) spectroscopy is an important tool for the non-destructive assay of radionuclides, however its sensitivity is limited by many practical factors, including the size, efficiency and resolution of the detector crystals. The Compton continuum (which arises due to the incomplete energy deposition of a scattered photon) can also obscure spectral detail, and coincident (cascade) radiation may be summed out as these are seen as a single decay. The aforementioned factors add noise and uncertainty to an energy spectrum, and must therefore be minimised to achieve the highest sensitivities. This is of particular importance for the assay of environmental samples, where \(\gamma\)-spectroscopy is often used for the detection of transuranics and other artificially created radionuclides. There are several approaches to increasing the sensitivity of \(\gamma\)-spectroscopy [1], and preliminary research into these will be presented here.

The project that this paper introduces has two main parts. Firstly, it involves using coincidence techniques [2–4] to reduce the background and Compton continuum. Secondly, it aims to use Monte-Carlo simulations to optimise detector geometry and designs, improving upon current setups. Initial work has focused on the use of NaI(Tl) scintillation detectors to develop a Monte-Carlo simulation and the post-processing techniques required for Compton suppression.

Experimental setup

NaI(Tl) detectors

The NaI(Tl) detectors and associated electronics are supplied by Canberra UK (Harwell, Oxfordshire). Each detector has a cylindrical crystal of dimensions \(\Theta = 76.2 \text{ mm}, \ l = 76.2 \text{ mm (model 802-3 \times 3)}\), and the pre-amp and multi-channel analyser (MCA) are combined into a single unit that mounts onto the back of the detector body (model Osprey\textsuperscript{TM}). This also controls all further amplification and digitisation of the pulse, saving both space and expense as separate NIM bins are not necessary. The detectors are partially enclosed in 40–60 mm of lead shielding, which serves to reduce the number of background counts by 54 % and also act as a support. For the purposes of this work the detectors were set up in a ‘back-to-back’ configuration (both looking directly at the source and separated by 180\(^{\circ}\), and a (simulated) image of the detector setup is shown below in Fig. 1.
Data acquisition

Data can be read in Pulse Height Analysis (PHA) mode from the Osprey units, but this only gives time integrated information, and no information on coincident events (other than sum peaks). Data is therefore collected in ‘list mode’, which synchronises the Osprey units and time-stamps the events in each detector [5]. An acute problem with this is the size and subsequent processing of the data files (~20 GB for two detectors on a five day count using a standard 37 kBq source). To address this, a post-processor has been developed using C++ and ROOT [6] that can reduce the data footprint by up to 12 times, and spread the data processing across several cores to speed up the analysis process.

GEANT4 simulation

The simulations have been carried out using the GEANT4 simulation toolkit developed at CERN (version 9.04p01) [7]. It is a publicly available software package that can be used to accurately simulate the passage of particles through matter. The definition of the experimental setup (including all shapes, objects and materials) is of particular importance for the accuracy of the simulation, and low energy electromagnetic (EM) physics models are used throughout the simulations presented here.

All simulations were carried out for 300–1,200 s of data collection (dependent on the source strengths and geometry’s replicated). This involved using sources of known activity, and calculating the total number of events expected in the data collection window. Summing effects due to the source activities are simulated by calculating the probability of a detector seeing from 1 to 10 events within the detectors characteristic decay time, and using this probability to generate extra decays in a corresponding number of events.

The GEANT4 default G4ParticleGun generator was used to create light (A < 137) radioactive isotopes in the simulation, which are then automatically disintegrated in GEANT4 according to known branching ratios (generated from the NuDat2 database [8]). Heavier isotopes (including 241Am) are created manually by firing photons isotropically (also using branching ratios from the NuDat2 database). This was primarily to cut the computing time required for a simulation, and ran 300 times quicker for a singular 241Am source than when using the radioactive decay method. The isotope was placed within the source geometry (a cylinder of known size) at a randomised co-ordinate in (x, y, z) each time the event generator was called, and both the isotope and source activity can be specified in a macro file. The resolution of the detectors was measured from 32.2 to 2,505.7 keV using several sources, and then approximated with a function of the form

\[ y = a + b \ln(c \cdot x + d) \]

where the energy deposited in the detector at the end of each event was then ‘smeared’ using a Gaussian spreading function. This used the function determined from the experimental data to calculate a resolution for that specific energy, convert it from FWHM to standard deviation, and reproduce the detectors resolution in the simulated data. The conversion to standard deviation used

\[ \text{FWHM} = 2.\sqrt{2 \ln 2}, \sigma \approx 2.355, \sigma, \text{a standard mathematical relation for a Gaussian peak.} \]

Model of the detector

The description of the detector geometry consists of the crystal itself, the optical coupler between the crystal and the photo-multiplier-tube (PMT), and the aluminium canister that surrounds these components. These were accurately represented, while everything that sat behind the optical coupler was approximated with an aluminium cylinder to save computing and development time (this was found to have no measurable adverse effects on the spectra produced). The lead shielding and source housings were also reproduced as accurately as possible.

The aluminium outer canister is of nominal thickness 1.5 mm, although the thickness of the front face was tuned to match the experimental data. The distance between the

Fig. 1 A GEANT4 geometry of the experimental setup. The yellow cylinder is the source, the red cylinders the detector caps, the grey cylinders the detector assemblies, and the grey (transparent) boxes are the lead shielding. The Osprey units are not shown here, but would mount onto the back of the detector assemblies.
front face of the canister and the crystal was initially set to 2 mm, and also optimised.

Post-Processing program

The post-processor provides the same functionality as seen in typical Compton suppression systems [3, 9, 10] (the ability to veto events based on their timing), although with much more flexibility. In traditional systems, coincident events are vetoed in real time using electronics, and all information about these events discarded. By recording all the events and post-processing them, the data can be searched as many times as necessary to extract far more coincidence information than is typically available.

The post-processor is written in C++, and re-processes list-mode data files into binned spectra in both ASCII and ROOT formats. Coincidence searching can be performed on a pair of detectors, which extracts all coincident events within a user-defined time window. The time window is defined with respect to one detector, which is used as a base (t_0) for the coincidence search. These events are then recorded into a 2D matrix. The coincidence searching routine is extremely efficient (over an order of magnitude quicker than the current processor), and is scalable to large datasets (currently tested up to 60 GB on a Pentium D 2.8 GHz system with 2 GB of RAM). The processor can also run on multiple cores, after which the analysis can be undertaken in ROOT or any other suitable software package.

The ability to gate on specific time and energy windows allows the user to ‘scan’ the coincidence space (as in Fig. 2, inset), and then select regions for further investigation and reprocessing (Fig. 2, main). It was originally developed to suppress Compton scattered events (which have a characteristic scattering time between detectors) although it can process any data set that contains events in coincidence (and the appropriate temporal information).

In the current work, data is run through the post-processor, and all results written to a ROOT file. Peaks are identified using the TSpectrum class in ROOT, and fitted using a Gaussian peak. The underlying continuum is calculated using algorithms described in [11, 12] with an inbuilt function of the TSpectrum class. This can then be approximated with a linear function across the range of the peak, with the integral of this subtracted from the integral of the peak itself.

Results

A typical spectra and background are shown in Fig. 3. The background consists primarily of 75 keV X-rays from lead, and the 1,461 and 2,614 keV peaks from 40K and 208Tl, respectively. Data was collected for long enough to provide sufficient statistics for comparison with the simulated spectra, while minimising the simulation time required. These counts ranged from 300 to 1,200 s in both a ‘close’ and ‘extended’ geometry (50 mm and 150 mm source-detector separations).
The absolute efficiencies (total and photopeak) were measured using a variety of sources, including $^{241}\text{Am}$, $^{133}\text{Ba}$, $^{60}\text{Co}$, $^{137}\text{Cs}$, and $^{22}\text{Na}$. These are defined as:

$$e_{\text{total}} = \frac{\text{total no. of gammas detected}}{\text{no. of gammas emitted by source}}$$

$$e_{\text{peak}} = \frac{\text{no. of gammas detected in the photopeak}}{\text{no. of gammas emitted by source}}$$

These sources were also simulated, and the results are presented in Fig. 4. Note that due to the resolution of NaI(Tl) ($6 - 7\%$), some low energy and closely packed peaks could not be resolved, and were also obscured by X-ray fluorescence from the shielding. Where multiplets could be resolved, these were fitted with overlapping Gaussians accordingly, otherwise, they were omitted from the efficiency calculations.

The simulated and experimental peak efficiencies were compared using a statistical $z$-test [13] for each energy point. These were then averaged for the entire dataset, resulting in a maximum error margin of $9\%$ at a $93.0\%$ confidence level. This is valid from $32.2 \leq E \leq 2505.7$ keV, and is adequate given the small sample size. A comparison between simulation and experiment for the extended geometry is shown in Fig. 5.

**Conclusions**

NaI(Tl) spectra for single and complex sources have been simulated using a newly developed GEANT4 code, with the geometry parameters optimised to obtain detection
efficiency’s within 9% of those determined from the experimental data, with a 93.0% confidence level.

Many aspects of GEANT4 have been tested and optimised for future work, including event generation, treatment of summing effects and the physics models used. The simulation output has also been tailored to allow compatibility with the experimental post-processor.

The post-processor has proved to be both highly efficient and flexible, with data compression of up to 12 times, and a coincidence searching routine that is both an order of magnitude faster than the current processor and scalable to very large datasets. This allows much more information to be obtained about coincident events than with a traditional electronic veto, and should enable the MDA’s of the current detector set-ups to be obtained with a greatly simplified electronics setup, and possibly improved.

Acknowledgments Thank you to the University of Surrey, the AWE Technical Outreach Programme and EPSRC for funding this research.

References

Determining the efficiency of a broad-energy HPGe detector using Monte Carlo simulations

R. Britton · J. Burnett · A. Davies · P. H. Regan

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Abstract Broad-energy HPGe detectors have a useful range of 3 keV to 3 MeV, making them ideal for the assay of environmental samples. Such measurements however, are hindered by variations in the sample matrix, summing effects, and the Compton continuum. Detectors may be characterised by proprietary software in such a situation, however Monte-Carlo modelling is a useful, inexpensive alternative that also provides greater flexibility when determining the detector response and efficiency during a measurement. In the current work, a full GEANT4 model of a broad-energy HPGe detector is presented, and simulations of various samples are compared to experimental data. These are found to be accurate within 3 % at a confidence level of 95 % for energies from 30 to 3,000 keV, with greater variations below 100 keV due to an increased sensitivity to geometrical inaccuracies.

Keywords Gamma spectroscopy · GEANT4 · Monte Carlo · Detector efficiency · NORM

Introduction

High purity Germanium (HPGe) detectors have become the de facto standard for performing gamma (γ) spectroscopy due to their excellent energy resolution (~0.18 % at 662 keV), the availability of large volume crystals, and Germanium’s high Z number (increasing the materials stopping potential). These properties allow the assay of radionuclides over a range of energies, and the useful range for environmental samples extends to ~3 MeV. Broad Energy HPGe (BEGe) crystals have several advantages over standard co-axial and low-energy designs in this range, including improved low-energy efficiency and charge collection at higher energies [1].

Several factors may hinder the assay of environmental samples, including variations in the source matrix, summing effects and the Compton continuum [2–4]. The sample matrix is of particular concern, as it can consist of a variety of materials and arrive in different geometrical configurations. These variations make the efficiency of a detector difficult to characterise, and can therefore introduce large errors when quantifying radionuclides. The current work presents a GEANT4 based Monte Carlo model of a BEGe detector and associated shielding, which is used to characterise the efficiency of a NORM (naturally occurring radioactive material) source, allowing the activity and abundances of radionuclides present to be deduced.

Materials and methods

Experimental setup

The BEGe detector (model BE3825) and associated electronics are supplied by Canberra UK (Harwell, Oxfordshire). The detector has an efficiency of 28 % relative to a standard 3 × 3 NaI(Tl) crystal at 1.33 MeV, and a carbon epoxy window to minimise the attenuation of photons at low energies. The crystal is also relatively thin when compared to a standard coaxial design, minimising the contribution from higher energy decays (and therefore improving low energy efficiency). The cryostat (model
7500SL-RDC-6-ULB) is specifically designed to minimise background by using low-background components and offsets so that more active parts can be effectively shielded. A preamplifier (model 2002C SL) processes the initial signal data. All detector dimensions are taken from the manufacturer provided documents, and these are summarised in Table 1.

The preamplifier output is sent to a LYNX™ digital signal processing unit from Canberra, which controls all further amplification, pole-zero correction and digitisation of the pulse. Data is collected in ‘list mode’, which time-stamps each event in the detector and writes these to a text file [5]. This method does produce larger data files than necessary, however an efficient post-processor is available that allows for greater flexibility in the analysis process [6].

The detector is surrounded with a 105 mm thick graded lead shield, with liners of tin and copper to reduce fluorescence from lead x-rays. This creates a low background environment for the BEGe detector, with an ambient background of 2 counts per second (cps). A simulated image of the detector (including the lead cave, but excluding some internal detail of the detector due to graphics limitations) is shown below in Fig. 1.

### Measurement and analysis

Apparent peak efficiencies were calculated using Eq. 1, where \( N \) is the total number of counts in the peak, \( t \) is the detection time, \( A \) the nuclides activity and \( b \) the photon’s branching ratio [7]. \( C_i \) is a correction factor due to dead time, radioisotope decay and coincidence summing corrections, however in the data analysed dead time is already accounted for. As the count times are far shorter than the half-lives of the isotopes, this contribution can also be neglected, and coincidence summing errors were minimised with the use of low activity sources.

\[
\varepsilon = \frac{N}{tAb} C_i
\]  

The isotopic abundances are calculated using Eq. 2, where \( N_0 \) is the number of atoms present, and \( \lambda \) is the decay constant.

\[
N_0 = \frac{N}{\lambda t b \varepsilon} \quad (2)
\]

Some isotopic abundances cannot be measured directly due to detection limits, however they can be inferred from the populations of their daughter isotopes. For sources that are in secular equilibrium, \( \lambda_{\text{parent}} \ll \lambda_{\text{daughter}} \), and the activity of the daughter \( (A_2) \) can be expressed as the product of the parents decay constant and the initial parent population (Eq. 3).

\[
A_2(t) \approx \lambda_{\text{parent}} N_0 \quad (3)
\]

Sources were chosen to cover the 30–3,000 keV energy range that is of use for environmental analysis. Several single \( \gamma \) emitters were used, as well as a NIST (National Institute of Standards and Technology) traceable complex \( \gamma \) source. The isotopes that comprised these were \(^{241}\text{Am} \quad (59.54 \text{ keV})\), \(^{109}\text{Cd} \quad (88.03 \text{ keV})\), \(^{57}\text{Co} \quad (122.06 \text{ keV})\), \(^{139}\text{Ce} \quad (165.86 \text{ keV})\), \(^{113}\text{Sn} \quad (391.68 \text{ keV})\), \(^{137}\text{Cs} \quad (661.67 \text{ keV})\), \(^{54}\text{Mn} \quad (834.84 \text{ keV})\), \(^{88}\text{Y} \quad (898.04 \text{ and 1,836.06 keV})\),

### Table 1  A summary the BEGe detector dimensions from Canberra

<table>
<thead>
<tr>
<th>Component</th>
<th>Dimension (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ge crystal diameter</td>
<td>71.0</td>
</tr>
<tr>
<td>Ge crystal length</td>
<td>26.5</td>
</tr>
<tr>
<td>Aluminium endcap thickness</td>
<td>1.5</td>
</tr>
<tr>
<td>Endcap window thickness</td>
<td>0.5</td>
</tr>
<tr>
<td>Top dead layer</td>
<td>0.0004</td>
</tr>
<tr>
<td>Side dead layer (each side)</td>
<td>0.6</td>
</tr>
<tr>
<td>Crystal distance from outside</td>
<td>5.0</td>
</tr>
</tbody>
</table>
The NORM source contained 0.323 kg of loosely packed granite stones (each approximately 20 mm in diameter) contained within a small bottle.

**GEANT4 simulation**

The simulations have been carried out using the GEANT4 Monte Carlo toolkit developed at CERN (version 9.04p01) [8], which enables the accurate simulation the passage of particles through matter. The definition of the experimental setup (including all shapes, objects and materials) is of particular importance for the accuracy of the simulation, as low energy photons are highly susceptible to small errors in the geometry.

**Implementation of the model**

Low energy electromagnetic physics models (valid from 250 eV to 100 GeV) are used to model photon interactions with the detector throughout the simulations. Radioactive decays from light (A < 137) isotopes are disintegrated using the radioactive decay module built into GEANT4, while heavier isotopes are created manually by firing photons isotropically. These are defined in a $\gamma$ library developed specifically for this purpose, and use branching ratios ($\text{if } > 10^{-5} \text{ per decay}$) from the NuDat2 database [9]. This method reduces the computing time required to simulate a $^{241}$Am source by a factor of 300, is limited to $\gamma$ decays, and does not take into account any angular or temporal correlations between emissions. It does, however provide a useful approximation of the sources, as the simulations accuracy is dominated by possible geometry errors.

The source matrix is reproduced by generating the decay at a random coordinate within the source geometry. Summing effects due to the source activities are simulated by calculating the probability of a detector seeing from 1 to 20 events within the detectors characteristic decay time, and using this probability to generate extra decays in a corresponding number of events.

The resolution of the detector was measured from 32.2 to 2,505.7 keV using several sources, and then approximated with a function of the form $y = a + b \ln(cx + d)$. The energy deposited in the simulated detector at the end of each event was then ‘smears’ using a Gaussian spreading function. This used the function determined from the experimental data to calculate a resolution for that specific energy, and reproduce the detectors resolution in the simulated data.

The description of the detector geometry consists of the crystal itself, the outer and inner electrodes, the aluminium canister and carbon epoxy window. The cold finger and vacuum spaces within the detector were also included, and to complete the model, the tin and copper lined lead shielding and source housings were recreated. NIST compounds were used where available, and all components were reproduced as accurately as possible according to manufacturer specifications (Fig. 2).

**Tuning the model**

Possible sources of error include unknowns in the detector (such as crystal positioning and alignment), and the determination of the crystals dead layers, which may vary across the detector surface [10]. Errors in the uniformity of the source matrix may also cause unwanted effects, and the thicknesses of attenuators within the detector are only approximately known. Simulations can be carried out with

![Fig. 2 A comparison between experimental and simulated spectra for a $^{60}$Co source. This data was taken at the detector top, and collected for 5 min. The corresponding simulation takes around 40 min using an Intel Core i5 processor (2.67 GHz). For a full colour image please see the online version of this article](image-url)
these dimensions, however they are unlikely to be accurate, especially in the low energy region that is most sensitive to geometry changes.

To minimise these errors, secondary measurements were made of all components to verify their dimensions. Unfortunately, internal dimensions of the detector cannot be measured in this way, and so have to be ‘tuned’ to match the experimental data [11]. The most critical of these involve any materials that are between the crystal and the source, and the exact positioning and shape of the crystal. The shape of the crystal is well known, as it is a simple cylinder with non-bulletised edges, however the exact dimensions of the rear electrode are not.

The dead layer in the front face is very thin (≈0.4 μm), and predominantly affects the low energy part of the spectrum. The side dead layers are somewhat larger (≈600 μm), and of importance as the current work also considers unusual geometries and side mounted sources (such as Marinelli beakers).

Results and discussion

A comparison between experimental and simulated data for a complex gamma source is shown in Figs. 3 and 4. Data was collected for ~20 min, and experiments were carried out with multiple sources (in several geometries) to validate the model.

When tuning the model, a range of rear electrode parameters were tested (assuming it was cylindrical in form). A larger rear contact would act to reduce the efficiency of the higher energy decays slightly, while not affecting the lower energy part of the spectrum. No significant change could be found, however, and the variation produced was insignificant when compared to the errors from the positioning and definition of the sources (typically ±5 %). This is assumed to be because it is not a ‘critical’ component (i.e. between the source and the crystal), and therefore has a minimal effect on the detector response. The distance from the carbon epoxy window to the crystal

Fig. 3 Simulated (red) and experimental (black) data for a complex γ source. Data was taken 5 mm from the top of the detector and collected for 20 min. The top image shows the energy spectra collected and simulated, while the bottom shows a peak efficiency plot for the same geometry. For a full colour image please see the online version of this article.
Fig. 4 Simulated (red) and experimental (black) data for the same source as in Fig. 3, mounted onto the side of the detector casing. The top image shows the energy spectra collected and simulated, while the bottom shows a peak efficiency plot for the same geometry. For a full colour image please see the online version of this article.

Fig. 5 Peak efficiency for an encapsulated granite source, for randomly (and evenly) distributed decays as a function of energy. The chemical composition of granite was taken from Ref. [14], and errors are due mainly to the uncertainties in this composition.
was revised down to 4.7 mm, and the dead layers remained unchanged from the manufacturer’s specifications.

The simulated and experimental peak efficiencies were compared using a statistical Z-test [12], which yields a maximum error between experiment and simulation of 3% with a 95% confidence level for the standard geometry. The only significant discrepancies are at low energy, and are due to the sensitivity of the detector response to small geometrical errors. The side mounted geometry has far greater attenuation between the source and crystal, reducing the detectors sensitivity to low energy decays. The simulated data matches this extremely well, with a maximum error across the energy range in the side mounted geometry of less than 1%.

Once these levels of accuracy were achieved, a NORM (Naturally Occurring Radioactive Material) sample of granite was simulated to determine the efficiency for a range of energies. The size, shape, chemical composition and density of both the granite and its container were reproduced, and photons from 30 to 3,000 keV were fired from within the source. The results of this efficiency calibration, which includes self-attenuation of the source, are shown in Fig. 5.

The sample was measured for 24 h, and analysed using the TSpectrum class in ROOT [13]. The isotopes identified and their abundances are recorded in Table 2.

Most radionuclides were identified using multiple peaks, and only 40K, 210Pb, 224Ra, 226Ra, and 228Th were seen as single emitters (increasing the errors in these abundances substantially). The sample (Fig. 6) appears to be in secular equilibrium, as most radionuclides in each decay chain

Table 2 Isotopic abundances from a granite NORM sample measured for 24 h, using a GEANT4 based efficiency characterisation

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Decay chain</th>
<th>Abundance (PPM)</th>
<th>Activity (Bq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>228Ac</td>
<td>232Th</td>
<td>7.61E-013 ± 0.3E-01</td>
<td>20.4 ± 0.4</td>
</tr>
<tr>
<td>212Bi</td>
<td>232Th</td>
<td>1.2E-013 ± 0.2E-013</td>
<td>22 ± 4</td>
</tr>
<tr>
<td>214Bi</td>
<td>238U</td>
<td>8.88E-014 ± 0.3E-015</td>
<td>47 ± 3</td>
</tr>
<tr>
<td>40K</td>
<td>–</td>
<td>4.2E+000 ± 0.6E+000</td>
<td>358 ± 49</td>
</tr>
<tr>
<td>210Pb</td>
<td>238U</td>
<td>4.5E-008 ± 0.7E-008</td>
<td>41 ± 6</td>
</tr>
<tr>
<td>212Pb</td>
<td>232Th</td>
<td>1.2E-012 ± 0.3E-012</td>
<td>19 ± 3</td>
</tr>
<tr>
<td>214Pb</td>
<td>238U</td>
<td>1.08E-013 ± 0.1E-014</td>
<td>42.4 ± 1.6</td>
</tr>
<tr>
<td>224Ra</td>
<td>232Th</td>
<td>1.280E-011 ± 0.2E-013</td>
<td>25 ± 4</td>
</tr>
<tr>
<td>226Ra</td>
<td>238U</td>
<td>3.7E-006 ± 0.5E-006</td>
<td>44 ± 6</td>
</tr>
<tr>
<td>228Th</td>
<td>232Th</td>
<td>2.9E-009 ± 0.8E-009</td>
<td>28 ± 8</td>
</tr>
<tr>
<td>234Th</td>
<td>238U</td>
<td>1.720E-010 ± 0.6E-012</td>
<td>47 ± 2</td>
</tr>
<tr>
<td>208Tl</td>
<td>232Th</td>
<td>2.23E-015 ± 0.3E-016</td>
<td>7.9 ± 0.6</td>
</tr>
<tr>
<td>235U</td>
<td>235U</td>
<td>1.3E-001 ± 0.5E-001</td>
<td>3.3 ± 1.2</td>
</tr>
<tr>
<td>238U*</td>
<td>238U</td>
<td>1.2E+001 ± 0.5E+001</td>
<td>47 ± 2</td>
</tr>
</tbody>
</table>

*a An isotope that was not be measured directly, but inferred from the daughter decay populations

Fig. 6 Energy spectra for the granite NORM source. Data was collected for 24 h at detector top
have around the same activity (the only exception is $^{208}\text{Tl}$, however this is due to the $^{232}\text{Th}$ decay chain splitting at $^{212}\text{Bi}$, with only 36% of the decays going to $^{208}\text{Tl}$). The population of $^{238}\text{U}$ was therefore inferred from the daughter decay of $^{234}\text{Th}$, giving a $^{235}\text{U}/^{238}\text{U}$ abundance of $1.07 \pm 0.36\%$, which is in fair agreement with the natural abundance of 0.72%. The abundances also agree with a prior analysis using proprietary software, and the simulation allows much more flexibility when determining the efficiency of unusual source geometries.

Conclusions

Broad-energy HPGe spectra for single and complex sources have been simulated using the GEANT4 Monte Carlo toolkit, with the geometry parameters optimised to obtain detection efficiencies within 3% of those determined from the experimental data, with a 95% confidence level. Multiple geometries have been explored, characterising the response of the detector for sources in standard and non-standard configurations, with no loss of accuracy. Small deviations were found at low energies (<100 keV) in the standard (detector top) source geometry, and are due to the limited accuracy of the detector reproduction.

A granite NORM sample has also been analysed using a GEANT4 based efficiency, allowing abundance estimates to be calculated for a range of radionuclides in the sample. Where multiple peaks could be identified for a single radionuclide, the abundances calculated across the energy range showed no sign of systematic error (that would arise if the efficiency calibration had been wrong), and the abundances also agree with a prior analysis using proprietary software. The accuracy of the abundance calculations is only limited by the counting time available, and the GEANT4 simulation also has the advantages of being license free, and highly flexible. This model can now be used to characterise the detector response for a variety of complex geometries and source configurations.

Acknowledgments

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References

Monte-Carlo based background reduction and shielding optimisation for a large hyper-pure germanium detector

R. Britton · J. L. Burnett · A. V. Davies · P. H. Regan

Abstract The performance of a radiation shielding system for a hyper-pure germanium detector has been characterised for Terrestrial radiation sources, Cosmic muons, X-ray fluorescence and the Compton scattering of source photons. Several methods to reduce the background seen are quantified, including increasing the inner radius of the Pb cave, and increasing the thickness of the shielding. Substantial improvements in the reduction of fluorescence X-rays are found to be achievable by modifying the liner thicknesses used. Increasing the Sn liner from 1.5 to 2.5 mm will increase the shielding of Pb X-rays from 95 to 99.5%. Reducing the Cu liner from 1.0 to 0.5 mm maintains a 99.5% level of shielding for Sn/Cd X-rays, however it greatly reduces the amount of Compton scattering of source photons into the detector (a process that is shown to cause an order of magnitude more events in the background than X-ray fluorescence). Cosmic muons were found to increase the amount of background radiation seen, both through direct interaction and the production of secondary radiation. The Cosmic muon contribution, however, was found to produce a much smaller effect than that caused by Terrestrial radiation and Compton scattered photons/fluorescence from the source. The total level of background radiation entering the detector chamber was found to decrease up to the full 200 mm of Pb shielding simulated.

Keywords HPGe detector · GEANT4 · Background reduction · Radiation shielding · X-ray fluorescence · Compton scattering · Cosmic muons

Introduction

The sensitivity of γ (gamma) spectroscopy relies upon the absolute efficiency of the detector, and the ability to identify decays of interest above the natural background. This consists of Terrestrial sources, including those within the detector, cryostat, shielding and surrounding materials, and secondary radiation produced by the interactions between the source radiation and the detector/shielding. Fast neutrons and muons also cause background counts in the detector, and are produced by Cosmic-ray interactions within the upper atmosphere.

The focus of this work is the optimisation of a radiation shield for use with future hyper-pure germanium (HPGe) detectors, including a large (model BE5030) broad-energy HPGe (BEGe) detector. This will minimise the background, improving the minimum detectable activities (MDAs) of the detector. Monte-Carlo simulations are used to achieve this, and specifically the GEANT4 toolkit [1] developed at CERN. All simulations are based upon similar, low-energy optimised codes discussed in Refs. [2, 3].

Theory

The MDA of a detector is inversely proportional to the detector’s efficiency, and only proportional to the square root of the number of background counts across the peak [4]. Simply increasing the size of the detector will therefore provide a greater MDA benefit than shielding up to a point,
however this is not a cost effective solution. Large detectors also suffer from increased summing effects, and peak to count (P/C) ratios level off around 50–66 % relative efficiency [5]. Bigger detectors also see much more background radiation, reducing the MDA benefit gained. Effective shielding is therefore critical to extract the optimum performance from radiation detectors.

Background sources

Terrestrial radiation

This primarily consists of the $^{238}$U, $^{235}$U, $^{232}$Th decay chains, and $^{40}$K. All of these may be present in the detector materials, cryostat, shielding and the building materials that house the laboratory. $^{222}$Rn (from the $^{238}$U chain) and $^{228}$Rn (from the $^{232}$Th chain) are usually present in the air, however these can be reduced by simply using the N$_2$ boiloff from the dewar to create a slight overpressure in the lead cave, preventing the majority of Radon isotopes from entering[6]. Standard lead shielding is also contaminated with $^{210}$Pb (typically up to 500 Bq/kg), although low background (aged) lead can reduce this to 25 Bq/kg or less.

Fluorescence and Compton scattering

Atoms in the shielding can become ionised or excited by impinging radiations, and then de-excite emitting a characteristic X-ray. For lead (which makes up the majority of the shielding) these are around 72–87 keV, and are typically shielded with a thin liner of low Z material, such as tin or cadmium. Tin and cadmium also emit characteristic X-rays from 23–29 keV, which can be shielded with an even lower Z material, such as copper.

Gamma (γ) radiation from the source will also interact with these liners and the lead shielding. The cross-section for Compton scattering varies linearly with the Z of the material, whereas the cross-section for the photo electric effect increases with Z$^{2.5}$ [7]. It is more likely for a photon of moderate energy to Compton scatter in the liners than in the lead due to the dominance of the photo electric effect in high Z materials. Low Z materials (such as the liners) will therefore substantially increase the background seen in a detector, despite suppressing any fluorescence.

Cosmic radiation

Cosmic ray interactions within the upper atmosphere generate secondary radiation, known as Cosmic showers. These include relativistic muons and fast neutrons, both of which may require several hundred metres of overburden to shield [8, 9]. Specialist plastics can be effective for thermalising and absorbing the neutron flux, such as polyethylene (PE) and borated polyethylene (PE:B) respectively. A typical neutron spectra is detailed in Table 1.

Muons from Cosmic showers arrive at sea level with a mean energy of 4 GeV [10], and as they are charged particles, typically lose energy via ionisation of the the matter that they pass through. Studies using a Cosmic veto [11] have found that up to 75.2 % of muon induced background events seen in a surface detector can be vetoed using coincidence techniques. A representative muon spectra is detailed below, in Table 2.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>An example neutron spectra, from Ref. [12]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy (MeV)</td>
<td>Neutron flux (10$^{-8}$cm$^{-2}$s$^{-1}$)</td>
</tr>
<tr>
<td>&lt;5 × 10$^{-9}$</td>
<td>1.07 ± 0.05</td>
</tr>
<tr>
<td>50 × 10$^{-9}$−10$^{-3}$</td>
<td>1.99 ± 0.05</td>
</tr>
<tr>
<td>10$^{-3}$ − 2.5</td>
<td>0.53 ± 0.08</td>
</tr>
<tr>
<td>2.5 − 5</td>
<td>0.18 ± 0.04</td>
</tr>
<tr>
<td>5 − 10</td>
<td>0.04 ± 0.01</td>
</tr>
<tr>
<td>10 − 15</td>
<td>(0.7 ± 0.2) × 10$^{-3}$</td>
</tr>
<tr>
<td>15 − 25</td>
<td>(0.1 ± 0.3) × 10$^{-6}$</td>
</tr>
</tbody>
</table>

It is clear that the spectra is dominated by thermal and slow neutrons of energies <1 keV, however there is still a significant contribution from higher energy neutrons.

<table>
<thead>
<tr>
<th>Table 2</th>
<th>An example muon spectra, from Ref. [13]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average momentum (GeV/c)</td>
<td>Muon intensity (cm$^{-2}$sr$^{-1}$s$^{-1}$)</td>
</tr>
<tr>
<td>0.2–0.4</td>
<td>3.57 × 10$^{-3}$</td>
</tr>
<tr>
<td>0.4–0.6</td>
<td>3.70 × 10$^{-3}$</td>
</tr>
<tr>
<td>0.6–1</td>
<td>3.41 × 10$^{-3}$</td>
</tr>
<tr>
<td>1–1.5</td>
<td>2.73 × 10$^{-3}$</td>
</tr>
<tr>
<td>1.5–2.5</td>
<td>1.73 × 10$^{-3}$</td>
</tr>
<tr>
<td>2.5–4</td>
<td>7.92 × 10$^{-4}$</td>
</tr>
<tr>
<td>4–6</td>
<td>4.24 × 10$^{-4}$</td>
</tr>
<tr>
<td>6–10</td>
<td>1.84 × 10$^{-4}$</td>
</tr>
<tr>
<td>10–13</td>
<td>1.13 × 10$^{-4}$</td>
</tr>
<tr>
<td>13–17</td>
<td>6.04 × 10$^{-5}$</td>
</tr>
<tr>
<td>17–25</td>
<td>2.51 × 10$^{-5}$</td>
</tr>
<tr>
<td>25–40</td>
<td>8.01 × 10$^{-6}$</td>
</tr>
<tr>
<td>40–70</td>
<td>1.89 × 10$^{-6}$</td>
</tr>
<tr>
<td>70–128</td>
<td>3.38 × 10$^{-7}$</td>
</tr>
<tr>
<td>128–250</td>
<td>5.19 × 10$^{-8}$</td>
</tr>
<tr>
<td>250–450</td>
<td>7.84 × 10$^{-9}$</td>
</tr>
<tr>
<td>450–1000</td>
<td>6.40 × 10$^{-10}$</td>
</tr>
</tbody>
</table>

Because of their high energy, muons also create large amounts of secondary radiation, especially when interacting with dense materials (such as Pb shielding).
Materials and design

Several designs and materials (Table 3) were tested, and all are based around a ‘standard’ cylindrical configuration of (from the outside in) lead (Pb), aged lead (low-Pb), tin (Sn) or cadmium (Cd), and copper (Cu). The lead makes up the primary shield with a nominal thickness of 102 mm, and the Sn/Cd and Cu liners are 1.0 and 1.5 mm respectively. The combination of these liners have been shown to reduce the X-rays from the lead by up to 98 % [4]. Cadmium (Cd) is often used instead of Sn for it’s higher density, however it has a high neutron capture cross-section and emits a 558 keV photon when absorbing a neutron, which may adversely affect detector performance. An example geometry is shown in Fig. 1.

Plastic neutron moderators and absorbers were also investigated but not fully simulated, as these were found to have been studied extensively in a previous publication by Stewart et al [14] (the results of which were in agreement with preliminary simulations carried out). Neutron moderators and absorbers must minimise the neutron flux and any subsequent photon production near the detector, and absorb thermalised neutrons to reduce neutron capture reactions within the detector or nearby materials. This rules out the use of internal moderators/absorbers, as thicknesses less than ~15 mm produce additional neutrons, and all thicknesses produce additional γ radiation when subjected to a neutron flux [14]. Large internal thicknesses cannot be used as the plastic is a low-Z material, and will therefore create a large continuum from scattered photons. The best performing shields in Ref. [14] (for stopping both neutrons and gammas) had separate neutron moderators and attenuators, used

<table>
<thead>
<tr>
<th>Material</th>
<th>Composition (atom no.ratio)</th>
<th>Density (g/cm³)</th>
<th>γ Production (neutron⁻¹mm⁻¹)</th>
<th>Activity (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>Natural</td>
<td>11.34</td>
<td>0.05</td>
<td>&lt;500</td>
</tr>
<tr>
<td>Pb</td>
<td>Low ²¹⁰Pb content</td>
<td>11.34</td>
<td>0.05</td>
<td>&lt;25</td>
</tr>
<tr>
<td>Cu</td>
<td>Natural</td>
<td>8.96</td>
<td>0.38</td>
<td>–</td>
</tr>
<tr>
<td>Sn</td>
<td>Natural</td>
<td>7.31</td>
<td>0.55</td>
<td>–</td>
</tr>
<tr>
<td>Cd</td>
<td>Natural</td>
<td>8.65</td>
<td>1.39</td>
<td>–</td>
</tr>
<tr>
<td>PE:Bi</td>
<td>CH₂, Bi [1:2:0.14]</td>
<td>3.0</td>
<td>0.22</td>
<td>–</td>
</tr>
<tr>
<td>PE:Li</td>
<td>CH₂, Li [1:2:0.1231]</td>
<td>1.06</td>
<td>0.11</td>
<td>–</td>
</tr>
</tbody>
</table>

Table 3 A summary of the materials considered in this study

Premadex, PE:B and water were eliminated as possible neutron shielding materials due to better performing alternatives. γ production values are for source neutrons interacting with 1 mm of material, and describe the amount of photons that penetrate the shielding per incident neutron.

Results and discussion

Terrestrial radiation

Background radiation in the laboratory is equivalent to a ~ 30 kBq (distributed) source with energies up to 2.5 MeV (assuming a 10 % efficiency for detection, and a 1 count per second, per keV average detection rate). This rate was verified experimentally, and is similar to that reported in [4]. The decays seen consist of various radionuclides over the 30 keV–2.5 MeV energy range, and the rate can change on a daily basis (mainly due to the ²²²Rn concentrations in the air). To simulate the background a ‘worst case’ scenario is reproduced, with 10⁷ 2.5 MeV photons (equivalent to 5–6 mins of a 30 kBq source) fired directly at the shielding. The total number of photons reaching the detector are recorded, and all secondary processes such as ionisation, fluorescence, pair-production etc. are included. The results are shown in Fig. 2, below, and an example spectra is also shown in Fig. 3.

As expected, larger shielding thicknesses provide greater protection from Terrestrial radiation. The amount that penetrated the shielding is reduced to 0.01 % of the initial flux (a factor of 10,000) at ~ 150 mm Pb. This is an order of magnitude greater than for the standard, 102 mm case, and is preferred for larger detectors as they are increasingly likely to pick up high energy gammas that penetrate the shield.
Fluorescence and Compton scattering from internal sources

Quantifying the levels of Compton scattering and fluorescence within the shielding is a difficult process. Source photons that are Compton scattered in the liners/shielding must be discerned from source photons that interact directly with the detector, and the fluorescence must be counted above the continuum and/or peaks from other radionuclides. The former is prohibitively difficult, while the latter suffers from large errors or unreasonably large count times.

These obstacles can be overcome in Monte-Carlo simulations by defining the source in such a way that it does not interact with the detector directly. This is achieved by limiting the initial photon directions to the $2\pi$ of the sources solid angle that is not subtended by the detector (see Fig. 4). Only secondary events are then seen, and an example spectra is shown in Fig. 5.

The number of Compton scattered photons seen in the simulations (minus the Pb X-rays) was normalised to the case where no liners were included. The levels of fluorescence seen were also recorded, however the statistics were too low to calculate the relationship to liner thickness with any accuracy. A separate artificial source was therefore created that only consisted of the X-rays required, and this was fired from outside the liners towards the detector. A summary of these results are shown in Figs. 6 and 7, and further simulations comparing the inner diameter of the Pb cave to the background seen are shown in Fig. 8.

From Figs. 5, 6, 7 and 8, it is clear that the liner’s composition, thickness and the inner radius of the shield have an important effect on the amount of background seen. Cu is the worst performing material due to it’s low $z$ value, increasing the scattered photons seen by 128% ($\sim 1.6 \times 10^4$ events) and reducing the fluorescence seen by 85% ($\sim 1.7 \times 10^3$ events) at typical liner thicknesses (3 mm). Sn and Cd both perform well, reducing the fluorescence by 99.78% and 99.85% respectively at 3 mm. Scattering was increased by 49 and 64%, however Sn and Cd both emit further X-rays in the 23–29 keV range. These can be attenuated by a thin layer of Cu, however this should be minimised due to the additional Compton scattered events this will cause.

If the X-rays are tolerable (or not within the energy range of interest) it is preferable to omit one or both liners completely, otherwise, 2.5 mm Sn/Cd with 0.5 mm Cu is optimal, as the Pb and Sn/Cd X-rays will be attenuated by >99.5%. Greater thicknesses may be detrimental to...
detector performance, as they increase the amount of Compton scattering seen (an effect that causes an order of magnitude more background events than fluorescence) for no appreciable benefit. The additional material will also increase the chance of interactions with Cosmic neutrons and muons [15], and should therefore be minimised. The inner shield diameter should be set as large as is reasonably practical; a radius increase of 20% reduced the number of counts seen from scattered photons by 35%.

210Pb decay in the shield

The lead shielding is layered, with standard lead used for the outside, and aged lead used for the inner shield, which must therefore be thick enough to attenuate the radiation from the outer shield. This was simulated by populating the outer lead shield with 500 Bq/kg 210Pb isotopes, and as expected, no radiation could be seen through aged lead (of at least 5 mm).

Further simulations were also undertaken to evaluate the thickness of the liners, and whether these could attenuate radiation from the inner (aged lead) shield. These results

![An example spectra, showing the fluorescence and Compton scattering from the shield and liners. The simulation included a 0.2 mm Cd liner, and ran for $2 \times 10^7$ events. Insert—a zoomed section showing the Pb (72–87 keV) and Cd (23–29 keV) X-rays in more detail](image)

![The percentage of 72–87 keV fluorescence photons seen as a function of liner thickness (normalised to a setup with no liners) for cadmium, tin and copper. The attenuation of 23–29 keV Sn and Cd X-rays with a Cu liner is also shown. For a full colour image please see the online version of this article](image)

![The percentage of Compton scattered photons seen as a function of liner thickness (normalised to a setup with a bare Pb cave) for cadmium, tin and copper. For a full colour image please see the online version of this article](image)

![The number of secondary events seen as a function of the shielding’s inner radius, normalised to the total number of events seen with a 140 mm inner radius. Liners were set to 1.0 mm Sn and 1.5 mm Cu for these simulations to mimic the current setup and minimise fluorescence. For a full colour image please see the online version of this article](image)
need to be considered within the context of the Section "Fluorescence and Compton scattering from internal sources", as increased liner thicknesses will also increase the amount of Compton scattering and Cosmic interactions seen. These results are shown in Fig. 9.

The 47 keV gamma decay is suppressed by increasing liner thicknesses as expected, with the intensity reduced to 0.001 % for a 2 mm liner. The $\beta$ decay of the $^{210}\text{Bi}$ daughter causes a bremsstrahlung continuum up to the endpoint energy at 1.162 MeV, which can be seen as the scattered photons in Fig. 9. These are not shielded as effectively as they are from a higher energy decay. To reduce the contribution from these events, the internal diameter of the lead should therefore be increased, or the $^{210}\text{Pb}$ concentration minimised.

Neutron absorption in the liners

Neutron radiation from Cosmic events is spread over a range of energies, and varies with both location and altitude. As cadmium has a high neutron interaction cross-section, simulations were performed to investigate whether the liners could attenuate the neutron flux reaching the detector. This would be at the expense of a 558 keV $\gamma$-ray emitted when cadmium absorbs a neutron, however this may be outside the energy range of interest, and so be of no further consequence.

At 1 and 10 MeV, neutron production is seen with greater liner thicknesses, but for energies less than this a reduction in neutron flux is observed (see Fig. 10). Due to it’s higher cross-section for neutron interactions, these effects are exaggerated for cadmium when compared to tin. For ‘real’ neutron spectra from Cosmic sources, the neutrons are mainly of an energy $<1$ keV [12, 16]. Simulations were therefore performed using this spectra, and the results for $\text{Sn}$ and $\text{Cd}$ are shown below in Fig. 11. The $\gamma$-rays produced with increasing thicknesses of $\text{Sn}$ and $\text{Cd}$ are also shown in Fig. 12.
With a realistic neutron spectra, both Sn and Cd liners reduce the neutron flux incident on the detector. Cd reduced the neutron flux much faster than Sn, however this reduction plateaued after a few mm of either material. This is due to the lower energy part of the incident neutron spectrum being absorbed by the liners, but the higher energy neutrons continuing through the material and producing more neutrons (as in Fig. 10). The reduction in background due to neutron absorption may however be offset by the increase in $\gamma$ production, which is up to 3 times greater in Cd than Sn. At 1 mm liner thicknesses, Cd reduced the neutron flux by $\sim 27\%$ and Sn by $\sim 10\%$. The corresponding values for $\gamma$ production are 1.39 and 0.55 $\gamma$-rays penetrating the liner per source neutron for Cd and Sn, respectively.

Muon absorption in the shield

To simulate the effects of a Cosmic muon flux on a typical lead shield, the Cosmic muon spectra from Ref. 13 was used to generate $10^6$ events for the geometry shown in Fig. 13. Substantial amounts of both muons and secondary radiation penetrated through the shielding to the detector, and the effects of increasing the shielding’s lid thickness are shown below in Fig. 14. The effects of increasing the bodys thickness was also investigated, however above 100 mm (the minimum needed to effectively shield Terrestrial radiation), no statistically significant levels of additional radiation were found to enter the chamber. Increasing the thickness of the shielding showed a corresponding increase in the total number of events seen in the chamber. This included muons that penetrated the shielding, but also neutrons, $\beta$-particles, gamma radiation and neutrinos. These are created by the muons interactions within the shielding, and the proportion of radiation seen at each lid thickness is shown in Fig. 14.

The major components that will interact with the detector (muons, $\beta$-particles, neutrons and gammas) maintain roughly the same percentage of the total number of particles seen irrespective of shield thickness, however the total number of events seen increases by $\sim 50\%$ in the first 50 mm of shielding, and a further $10\%$ over the next 150 mm. This suggests that even small shielding thicknesses will increase the Cosmic muon derived background substantially in a detector, however further increases beyond the first 50 mm only increase the background seen by a relatively small amount.

Minimising the external background radiation

The simulations suggest that while increasing shield thicknesses reduce the impact of radiation from Terrestrial and Cosmic neutron sources, the background will be increased by Cosmic muon sources. By using a realistic Cosmic muon spectra and an estimate of the Terrestrial background rate, an optimum shield thickness can be estimated. This result is shown in Fig. 15, which presents the relationship between shielding thickness and the total background that can be expected in a laboratory setting.
As the Terrestrial background rate is much greater than the Cosmic muon and neutron flux (by $\times 2$ orders of magnitude) the effects of Cosmic radiation do not become significant until the shielding reaches a thickness of 80 mm, where the Cosmic muon derived background approaches 25% of the total seen. Even at the full 200 mm simulated however, the reduction in Terrestrial background outweighs the increase in Cosmic muon derived radiation. One other subtlety involves the type of radiation produced, as Cosmic muons cause additional neutrons to enter the chamber as well as the gamma and beta radiation that is typically seen from a Terrestrial source. This is a small fraction of the radiation produced however ($\sim 0.2\%$), and therefore the small increases seen at large shielding thicknesses will not produce a measurably greater neutron flux.

**Conclusion and recommendations**

Plastic neutron moderators and absorbers were initially investigated, but not fully simulated due to the comprehensive nature of previous publications (the results of which were found to be in agreement with preliminary simulations of neutron absorption). The best performing shields (for stopping both neutrons and gammas) have separate neutron moderators and attenuators, used Lithium doped PE (PE:Li), Bismuth doped PE (PE:Bi) and Pb in a ratio 3:8:4. It is therefore recommended that future shielding designs use this specification where the shielding of neutrons is critical, as it is known to be both commercially available and the most effective solution. Internal neutron moderators and absorbers are not recommended as these greatly increase the amount of neutron and $\gamma$ radiation seen in the detector.

From the simulations, it is clear that increasing the inner radius and thickness of the shielding are effective measures for reducing the background that a detector will see. Increasing the shield thickness from 100 to 150 mm will reduce the amount of Terrestrial radiation seen by an order of magnitude, while a 20% increase in inner radius reduces the background seen by $\sim 35\%$.

To reduce fluorescence X-rays, liners of lower Z materials are needed, although these increase the overall level of background seen due to their relatively high Compton scattering cross-sections. The liners are not effective shields for radiation from $^{210}\text{Pb}$ decays, as the higher energy component is not attenuated by the relatively thin layers. Liners of the thicknesses needed to achieve this would substantially increase the number of background events due to the reduced internal diameter of the cave, increased Compton scattering and increased interactions with Cosmic radiation.

Currently, Cu and Sn is used in a 3:2 ratio (typically 1.5 and 1.0 mm thicknesses respectively), however using a thicker liner of Sn or Cd would be of great benefit, as these are far more effective than Cu for shielding Pb X-rays, and only 0.5 mm of Cu is required to remove 99.5% of Sn/Cd X-rays. Minimising the thickness of the liners (especially Cu) in the shielding also significantly reduces the level of Compton scattering seen (an effect that causes $\sim$ an order of magnitude more background events than the X-rays themselves).

Liner materials (Cd and Sn) were also tested for their neutron absorption properties, with cadmium outperforming tin for a realistic Cosmic neutron spectra. The net effect for both materials is neutron attenuation, reducing the flux by up to 28% at 3 mm. Additional $\gamma$ radiation is produced as the liners are typically thin components, and cadmium also emits a 558 keV $\gamma$-ray from the $(n, \gamma)$ interaction. Depending on the experimental requirements, Sn and Cd both have their respective advantages and disadvantages, and will provide some added benefit for the reduction of the Cosmic neutron flux incident on the detector.

As expected, there is no way to substantially reduce the Cosmic muon flux (and associated secondaries) over the relatively short material thicknesses involved in a typical Pb shield. Any thickness of shielding that is practical in a laboratory will actually increase the background seen, however this effect is most crucial over the first 50 mm of material, where the background derived from Cosmic muons increases by $\sim 50\%$. Above this, the level of background radiation entering the detector chamber is only increased by a further 10%, and produces a much smaller effect than that caused by Terrestrial radiation and Compton scattered photons/fluorescence from the source.

As a result of the differing contributions, the overall background radiation levels entering the detector chamber...
continue to fall up to the 200 mm of Pb shielding simulated. The main contribution is from Terrestrial radiation up until 100 mm, after which the Cosmic muon derived background contributes a substantial amount. The majority of background (both Terrestrial and Cosmic muon derived) is in the form of gamma radiation, however a significant neutron flux is also present (mainly from Cosmic neutrons, with some contribution from neutrons created by impinging Cosmic muons). This cannot be effectively shielded in 200 mm, however external neutron moderators and absorbers can reduce this contribution.

Depending on the materials and budget available, it can be concluded that increasing the Pb shielding thickness up to 150 mm will substantially decrease the levels of Terrestrial background seen. Beyond this thickness, additional background reduction is possible but at ever decreasing value due to the Cosmic muon contribution. Including an inner Pb shield with a low $^{210}$Pb content will also reduce the background contribution from $^{210}$Pb decays. If combined with a Sn liner of 2–3 mm, Pb fluorescence will also be minimised in an important energy region (72–87 keV), as well as additional Compton scattering of source photons by the liner material. The internal diameter of the shielding should be maximised and flushed with N$_2$, as this will substantially decrease both the primary and secondary radiation seen from all the above mentioned sources.

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References
Characterisation of cascade summing effects in gamma spectroscopy using Monte Carlo simulations

R. Britton · J. L. Burnett · A. V. Davies · P. H. Regan

Abstract A GEANT4 based Monte Carlo simulation has been successfully utilised to calculate peak efficiency characterisations and cascade summing (true coincidence summing) corrections in two source geometries commonly used for environmental monitoring. The cascade summing corrections are compared with values generated using an existing (validated) system, and found to be in excellent agreement for all radionuclides simulated. The calculated correction factors and peak efficiencies were also tested by analysing well defined sources used in the operation of the International Monitoring System, which undertakes radionuclide monitoring for verification of the Comprehensive Nuclear-Test-Ban Treaty. All abundances of the radionuclides measured matched the values that were previously determined using proprietary software. Using GEANT4 in this way, cascade summing corrections can now be extended to complex detector models and source matrices, such as Compton Suppression systems.

Keywords GEANT4 · True coincidence summing · Cascade summing · Gamma spectroscopy · Environmental monitoring

Introduction

High purity Germanium (HPGe) detectors are routinely used for environmental analysis, and provide an unrivalled energy resolution and efficiency for identifying different radionuclides. The useful range for such analysis is from 30 keV to 3 MeV, however many radionuclide activities need to be corrected for ‘cascade’ or ‘true coincidence’ summing [1].

Cascade summing occurs where an unstable nuclei emits multiple photons or X-rays in coincidence/cascade. If multiple photons deposit their energy in the detector crystal, a single output pulse will be generated as the charge collection times are typically much greater than the corresponding nuclear lifetimes. These events will therefore be interpreted as a full energy peak with the energy equal to the sum of the cascade photons. This is known as ‘summing out’, and reduces the apparent efficiency of the detector to that nuclide. It is also possible for multiple photons to sum to an equivalent energy of the photon of interest, increasing the apparent efficiency of the detector, and is known as ‘summing in’. Such correction factors can be estimated empirically [2], however they will change for each new geometry and source counted. It is therefore more efficient to calculate the correction factors numerically [3–5], or use Monte Carlo simulations to determine the appropriate correction factors [6, 7]. The latter also allows for additional complexity in the source matrix that may be difficult to achieve numerically.

The aim of this work is to utilise and extend an existing high-accuracy Monte Carlo model [8] of a HPGe detector to improve the efficiency calculations for radionuclides that are sensitive to cascade summing effects. Simulations using the full GEANT4 gamma library are compared to those using an efficient (user-defined) decay library, which allows for greater simulation speed at the expense of the number and complexity of decay channels [8]. The cascade summing correction factors can then be determined, and are validated with experimental data and an alternative calculation method.
Experimental methods

GEANT4 simulation

The simulations have been carried out using the GEANT4 Monte Carlo toolkit developed at CERN (version 9.5) [9], and are based upon similar models detailed in [8]. These are tuned to the detector used, and the detector response is accurate to within 3% at the top of the detector, and 1% in side-mounted source geometries. The definition of all shapes, objects and materials is of particular importance for the accuracy of the simulation, as low energy photons are highly susceptible to small errors in the geometry.

The description of the detector geometry consists of the BEGe (a Broad Energy HPGe detector), associated shielding and the source. The BEGe contains the detector crystal, outer and inner electrodes, the aluminium canister and the carbon epoxy window. The cold finger and vacuum spaces within the detector are also reproduced, as were the dead layers within the crystal. To complete the model, the tin and copper lined lead shielding and source housings were also recreated. NIST compounds were used where available, and all components were reproduced as accurately as possible according to manufacturer specifications.

Cascade summing is simulated using the GEANT4 radioactive decay data libraries. A radioactive isotope is disintegrated per event, with the decay radiation selected based upon the relative branching ratios for that nucleus. Multiple decay modes are possible, including situations where multiple photons are emitted in cascade. Radiation from these decays is propagated throughout the simulated volume, and any energy deposited within the active detector crystal recorded. These are summed on an event by event basis, and so reproduce the cascade summing seen in the laboratory. Simulations can be completed up to 300 times faster however, with a user-defined gamma library [8]. This approximates each source as a single particle emitter, and generates one photon per event. Branching ratios (if greater than $10^{-9}$) are taken from the NuDat2 database [10], and energy deposited in the detector is calculated as before. The speed increase allows for greater simulation accuracy and complexity where coincidence summing is not a serious concern, as the reduction in the number and complexity of decay channels prevents cascade summing from being simulated in this mode.

In both modes, complex sources can be created by selecting multiple radioactive nuclides and giving each a relative weighting. Summed events that are due to the activity of the source are also simulated as this is based upon a separate programming loop that can generate multiple decays per event.

Experimental setup

The BEGe detector [11] (model BE3825) and associated electronics are supplied by Canberra UK (Harwell, Oxfordshire). The BEGe has a short, planar shape to maximise efficiency for <1 MeV photons. The detector also utilises a carbon epoxy window to minimise the attenuation of low energy photons. This reduces the response to photons below 10 keV when compared to a Beryllium window (which can allow photons of energy ≥2 keV), however it is far more robust. The crystal is a cylinder of 71 mm diameter, and 26.5 mm thickness. A low background cryostat (model 7500SL-RDC-6-ULB) is used, and a preamplifier (model 2002C SL) processes the initial signal data.

The preamplifier output is sent to a LYNX™ digital signal processing unit from Canberra, which controls all further amplification, pole-zero correction and digitisation of the pulse. Data is collected in ‘list mode’, which timestamps each event in the detector and writes these to a text file [12].

The BEGe detector is placed within a cylindrical 105 mm thick graded lead shield, with tin and copper liners to reduce fluorescence from lead X-rays. This reduces the background to ~2 counts per second (cps), and a simulated image of the setup is shown in Fig. 1.

![Fig. 1 A simulated image of the experimental setup. The primary detector is shown in the centre (the grey cylinder), with an example source above it (the yellow cylinder). The surrounding lead shield and liners are also shown, with grey corresponding to lead, silver to tin and red to copper. (Color figure online)](image-url)
Measurement and analysis

Apparent peak efficiencies and isotopic abundances were calculated as in [8]. All data was analysed using the post-processor detailed in [13].

Calibration sources were chosen to cover the 10–3,000 keV energy range, and included NIST (National Institute of Standards and Technology) traceable complex γ sources. The isotopes that comprised these were $^{241}$Am (59.54 keV), $^{109}$Cd (88.03 keV), $^{57}$Co (122.06 keV), $^{139}$Ce (165.86 keV), $^{113}$Sn (391.68 keV), $^{137}$Cs (661.67 keV), $^{54}$Mn (834.84 keV), $^{88}$Y (898.04 and 1,836.06 keV), $^{65}$Zn (1,115.54 keV), and $^{60}$Co (1,173.23 and 1,332.49 keV).

Two complex γ sources were used to validate the cascade summing corrections; a cylindrical compressed air filter [type half-RASA (Radionuclide Aerosol Sampler/Analyser)], and a 15-layer reference filter pack (type CINDERELLA). Both are used for the operation of the IMS (International Monitoring System), which undertakes radionuclide monitoring for verification of the CTBT (Comprehensive Nuclear-Test-Ban Treaty). The half-RASA source measured ~72 mm diameter by 18 mm thickness, and is from IMS station JPP38. This was exposed to some of the environmental radiation from the Fukushima incident (March 2011), and as such contains a high proportion of $^{134}$Cs (a nuclide that requires substantial cascade summing corrections). The second source is a CINDERELLA type, and measures ~83 mm diameter by 8 mm thickness. The CINDERELLA source contains multiple radionuclides, the activities of which were accurately characterised at NPL (National Physical Laboratory). The performance and behaviour of these source types is well understood, and they therefore provide an excellent test of both the GEANT4 efficiency characterisation and the cascade summing corrections.

Calculating the correction factors

Cascade summing is dependant on both the decay of the radionuclides, and the geometry of the detection system. The correction factors are therefore different for each variation in source matrix and position, and only comparable in specific geometries. The cascade summing sources have been previously analysed, and the concentrations of the radionuclides present are well known. The correction factors and efficiency characterisations used in the previous analysis were calculated using the Geometry Composer module in Canberra’s GENIE™ 2000 software (version 3.6). This has been previously validated, and will be used as the basis for comparison with the GEANT4 calculated values (as well as the NPL activities for the CINDERELLA source).

Simulations were performed using the full GEANT4 decay libraries, and repeated using the user-defined gamma libraries. The total number of events was calculated such that the total number of photons from each source was the same, however in the latter case these were all emitted individually, with no coincidence data (see Fig. 2). The full energy photopeaks can then be counted in each simulation, and the ratio of the peak areas used to determine the correction factors appropriate for the specific detector, source, and experimental geometry used.

Results and discussion

Efficiency of the sources

To allow a comparison between the radionuclide abundances in both samples previous and current analyses, the peak efficiency of the detection system must be determined. As the GEANT4 model of the detector has been validated and is accurate to within 3 %, simulations were run using photons of energy from 10 to 2,500 keV. Both sources were reproduced according to their size, chemical composition and density, and the photons fired from randomised coordinates within this volume. The determined efficiencies are shown in Fig. 3. All errors are quoted at the 1 sigma significance level.

Calculated correction factors

An example simulated spectra using the full GEANT4 libraries is shown in Fig. 4. Corresponding simulations were also performed using data-files that contained no coincidence information, and the calculated correction factors for the main γ decays in the half-RASA source are summarised in Table 1. A full list of the calculated
For the half-RASA source, the results show excellent agreement between the calculated correction factors and those determined using Canberra’s GENIE™ 2000 software, with a average deviation of 3 % and a maximum of 6 %. Similar agreement is also found in the CINDERELLA source.

Table 1 A summary of the correction factors calculated for the half-RASA source

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy (keV)</th>
<th>Correction factor</th>
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<td>$^{134}$Cs</td>
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Some correction factors were not calculated in the Canberra software due to those peaks not being present in the preliminary analysis. Errors in the calculated correction factors are ±6 %, and were dominated by the variability in the source geometry/composition.

Table 2 A summary of the correction factors calculated for the CINDERELLA source, with ‘G4’ representing GEANT4 values, and G2K representing GENIE™ 2000 derived values

<table>
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<th>Isotope</th>
<th>Energy (keV)</th>
<th>Correction factor</th>
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<td>$^{140}$La</td>
<td>1136</td>
<td>0.744</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>1398.57</td>
<td>0.677</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>109.42</td>
<td>0.543</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>131.12</td>
<td>0.480</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>241.93</td>
<td>0.603</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>266.54</td>
<td>0.598</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>328.76</td>
<td>0.709</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>432.49</td>
<td>0.753</td>
</tr>
</tbody>
</table>

Some correction factors were not calculated in the Canberra software due to those peaks not being present in the preliminary analysis. Errors in the calculated correction factors are ±6 %, and were dominated by the variability in the source geometry/composition.
geometry. For $^{140}$Ba, $^{134}$Cs, $^{140}$La, $^{147}$Nd and $^{103}$Ru the two calculation methodologies had an average deviation of 4 %, and a maximum of 8 %. Reasonable agreement was found for $^{99}$Mo, $^{135}$I, and $^{132}$Te with an average deviation of 8 % between the GEANT4 and GENIE $^{TM}$ 2000 correction factors.

Overall, the two approaches yield values that are in close agreement for a variety of radionuclides. As they use completely different methods, this confirms both the validity of the GEANT4 model and the approach used to determine the correction factors. It is worth noting that the errors in this calculation are fairly large, as true coincidence summing is highly sensitive to the geometry of both the detector and the source (as well as their relative positions).

### Analysis of realistic sources

Analysis was completed for both the half-RASA и CINDERELLA sources. Figure 5 shows the complex gamma spectrum obtained from the half-RASA source, while Fig. 6 shows the spectrum for the CINDERELLA source. For both samples, radioisotopes were identified using their characteristic energies and a weighted mean taken if the isotope emitted multiple photons. The radioisotopes detected in the half-RASA source and their (cascade corrected) abundances are summarised in Table 3, alongside the accepted values for this source (corrected for radioactive decay between the previous analysis and the latest measurement). The same comparison between the CINDERELLA source and the NPL calibration values is shown in Table 4.

In the half-RASA data, it is clear that there is a large background continuum from higher energy decays. This reduces the sensitivity slightly, however mBq detection levels are still attainable. The CINDERELLA spectra was far more complex, and some energy peaks could not be determined solely using Monte-Carlo methods (GEANT4). The ‘accepted activity’ is from a previous study, and is time corrected to account for the radioactive decay between the collection of the two datasets.

### Table 3 A summary of the radioisotopes detected in the CTBT half-RASA source, and their abundances

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Accepted activity (Bq/m$^3$)</th>
<th>Calculated activity (Bq/m$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{7}$Be</td>
<td>$4.10 \pm 0.25 \times 10^{-3}$</td>
<td>$3.8 \pm 0.4 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>$1.05 \pm 0.02 \times 10^{-4}$</td>
<td>$1.04 \pm 0.05 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>$1.44 \pm 0.04 \times 10^{-4}$</td>
<td>$1.39 \pm 0.07 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{210}$Pb</td>
<td>$4.6 \pm 0.5 \times 10^{-4}$</td>
<td>$4.4 \pm 0.4 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

The ‘calculated activity’ is from the present study, and was determined solely using Monte-Carlo methods (GEANT4). The ‘accepted activity’ is from a previous study, and is time corrected to account for the radioactive decay between the collection of the two datasets.

### Table 4 A summary of the radioisotopes detected in the CTBT CINDERELLA source, and their abundances

<table>
<thead>
<tr>
<th>Isotope</th>
<th>NPL calibration value (Bq/m$^3$)</th>
<th>Calculated activity (Bq/m$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{140}$Ba</td>
<td>$4.77 \pm 0.10 \times 10^{-3}$</td>
<td>$4.43 \pm 0.26 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{141}$Ce</td>
<td>$3.13 \pm 0.06 \times 10^{-3}$</td>
<td>$3.05 \pm 0.30 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{144}$Ce</td>
<td>$4.97 \pm 0.15 \times 10^{-4}$</td>
<td>$4.84 \pm 0.47 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>$1.383 \pm 0.027 \times 10^{-3}$</td>
<td>$1.28 \pm 0.16 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>$2.22 \pm 0.07 \times 10^{-5}$</td>
<td>$2.23 \pm 0.31 \times 10^{-5}$</td>
</tr>
<tr>
<td>$^{140}$La</td>
<td>$0.87 \pm 0.03 \times 10^{-3}$</td>
<td>$0.84 \pm 0.15 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{99}$Mo</td>
<td>$3.81 \pm 0.12 \times 10^{-4}$</td>
<td>$4.24 \pm 0.38 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{95}$Nb</td>
<td>$8.23 \pm 0.61 \times 10^{-4}$</td>
<td>$7.4 \pm 0.4 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{147}$Nd</td>
<td>$1.48 \pm 0.10 \times 10^{-3}$</td>
<td>$1.52 \pm 0.07 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{103}$Ru</td>
<td>$7.72 \pm 0.17 \times 10^{-4}$</td>
<td>$7.48 \pm 0.41 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{132}$Te</td>
<td>$4.67 \pm 0.14 \times 10^{-4}$</td>
<td>$4.89 \pm 0.26 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{90}$Zr</td>
<td>$2.476 \pm 0.048 \times 10^{-3}$</td>
<td>$2.53 \pm 0.10 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

The calculated activity was again determined solely via Monte Carlo methods. This is compared to the (time-corrected) activity values determined during the initial calibration at NPL. Nuclide activities were also corrected to account for the feeding of different isotopes from other decays (such as $^{140}$Ba, which decays into $^{140}$La).
resolved (increasing the errors in the activities of these radioisotopes). For both sources, the abundances determined are in good agreement with the previously reported values, suggesting that the sample efficiency and cascade correction factors are accurate. Where radioisotopes were identified using multiple peaks, good agreement was also seen in the calculated abundances across the energy range, which would not be the case if some systematic error had been present in the analysis process.

Conclusion

A GEANT4 based Monte Carlo simulation has been successfully utilised to generate peak efficiency characterisations and multiple cascade summing corrections in two source geometries commonly used for environmental monitoring. The cascade summing corrections are compared with values generated using an existing (validated) system, and found to be in excellent agreement for many radionuclides (within 10% of the GENIE™ 2000 values). The calculated correction factors and peak efficiencies were also tested using half-RASA and CINDERELLA type sources, in which the abundances of the radioisotopes detected matched those previously determined.

The process of generating the efficiencies relies upon an accurate simulation of the detector system and source, and as such requires a working knowledge of GEANT4 and C++. The software from Canberra requires no such expertise, and is inherently more user-friendly. The Monte Carlo method also requires more time to complete, unless an accurate simulation is already available (in this case the methods take a similar amount of time). A major benefit of using the GEANT4 method, however, is the complexity possible in both the detector model and the source matrix. This may be required for applications involving multiple detector set-ups, such as Compton Suppression systems. As well as being applied to laboratory measurements, cascade summing corrections may also be used in simulations. This would involve generating the correction factors in GEANT4, and then applying these to further simulations utilising the user-defined decay library. The user can then take advantage of the speed gained while using an efficient decay library, and the accuracy achievable when using the full GEANT4 dataset for complex efficiency characterisations.

Acknowledgments The authors thank the University of Surrey, the AWE Technical Outreach Program and EPSRC for funding this research.

References

Improving the effectiveness of a low-energy Compton suppression system

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A novel method for collecting and processing coincidence data from a Compton Suppressed Low Energy Photon Spectrometer (LEPS) is presented, greatly simplifying the current setup and extending the suppression abilities of the system. Offline analysis is used, eliminating the need to discard coincidence data when vetoing coincident events with fast-timing electronics. Additional coincident events are identified that are usually missed, and which represent interactions in the active NaI(Tl) shield prior to an interaction in the LEPS detector. By suppressing these events, the Compton Suppression factor was improved by 144\% for the 661.66 keV decay line in a $^{137}$Cs source. The geometry used for this particular Compton suppression system is highly sensitive to these effects, however similar event profiles are expected in all coincidence systems.

1. Introduction

Gamma (\(\gamma\)) spectroscopy allows the non-destructive identification of radionuclides in environmental samples, however the sensitivity of these systems is dependent on many factors. This includes the resolution and efficiency of the detector, and the amount of background radiation seen \cite{1}. The Compton continuum (which arises due to the incomplete energy deposition of a Compton scattered \(\gamma\) decay in the crystal) also obscures lower energy decays, reducing the observed Peak to Count (P/C) ratio for these transitions \cite{2}. Many radionuclides of interest fall into this category, and methods employed to improve the low-energy performance of \(\gamma\)-spectroscopy systems are discussed below.

Low-Energy HPGe (LEGe) crystals allow for greater relative efficiency in the low-energy region where the Compton continuum dominates the spectra. The crystals are typically quite small, reducing the contribution from higher energy decays. The electrical contacts on the crystal are also optimised to reduce the detector capacitance (and therefore the pre-amplifier noise), improving the low energy resolution \cite{3}. These improvements, however, come at a cost; due to the size of the LEGe crystal the efficiency is typically lower than that of an equivalent coaxial or broad energy design, and many photons Compton scatter out of the crystal. To suppress these events, additional NaI(Tl) detectors are used to ‘capture’ the escaping photons. As the timescales of these atomic processes are infinitesimally smaller than the corresponding timescales for charge collection and pulse formation in the detector and associated electronics, such events are seen in coincidence between the LEGe and the NaI(Tl) crystals.

Events in coincidence can be discriminated using fast-timing electronics, which are typically employed \cite{4} to set up a ‘delay window’. If an event is detected in the LEGe, and another event is seen in the NaI(Tl) detector (within the predefined time window), the original event in the LEGe is discarded. This limits the system, as the delay window must be characterised before any data collection, and cannot be modified during a run. All coincidence and temporal information is also lost.

The work presented in this report describes the use of a LEGe based Compton suppression system in conjunction with List-mode acquisition software \cite{5} and a customised post-processor \cite{6}, to substantially improve the suppression factors gained in comparison to a ‘standard’ electronics based Compton veto system.

2. Materials and methods

2.1. Experimental setup

The LEGe detector (model GL0510) and associated electronics are supplied by Canberra UK (Harwell, Oxfordshire). The detector is optimised for low energy photons, and is set up to cover a range of 10–700 keV (above this value the efficiency is $< 0.5\%$). The detector also has a carbon epoxy window to minimise the attenuation of low energy photons, which allows $\sim 82\%$ transmission at 10 keV \cite{3}. The crystal is fairly small (an active diameter of 25.5 mm, and a thickness
of 10.5 mm), minimising the contribution from higher energy decays. An ultra-low background cryostat (model 7915-30-ULB) is used, and a preamplifier (model I-TRP) processes the initial signal data.

The preamplifier output is sent to a LYNX™ digital signal processing unit from Canberra, which controls all further amplification, pole-zero correction and digitisation of the pulse. Data is collected in ‘list mode’, which time-stamps each event in the detector and writes these to a text file [5].

The NaI(Tl) shield detectors are from Scionix (Utrecht, Netherlands), and comprise of a cylindrical annulus with a removable plug that is positioned above the primary detector. This is not the most effective geometry for a Compton suppression system, as there may be a large number of false coincidences due to γ radiation emitted in cascade, chance coincidences due to the activity of the source, and to a lesser extent, background radiation. Suppression will also be reduced when compared to lower energy decays, as dead layers in the setup (including crystal dead layers, detector casings, etc.) greatly reduce the detection efficiency for low energy photons. Note that while the LEGe has a thin entrance window at the top, Compton scattered photons will typically exit the detector through the Aluminium casing, and will therefore be attenuated before entering the NaI(Tl) veto detector (through another thin layer of Steel).

The cylinder has five Photo-multiplier Tubes (PMTs) for efficient charge collection, and the plug one. The outputs are combined from all of the PMTs, and passed to a preamplifier (model 2005). Again, the preamplifier output is sent to a LYNX™ unit, where the data is collected in ‘list-mode’. A synchronisation cable also runs between the two LYNX™ units to allow synchronisation of the clocks used to record the events in each detector.

The LEGe and NaI(Tl) combination is placed within a 80 mm thick lead shield, with tin and copper liners to reduce fluorescence from lead and tin x-rays. This reduces the background to ~2 counts per second (cps). A simulated image of the setup (including the lead cave, but excluding some internal detail of the detectors due to graphics limitations) is shown below in Fig. 1.

2.2. Compton Suppression

Several methods are available for quantifying the levels of suppression achieved [2], however the main ones used in this document will be the Peak to Count ratio (P/C), Peak to Total ratio (P/T) and the Compton Suppression Factor (CSF).

- **Peak to Count (P/C) ratio** – This is defined as the ratio of the counts in the highest photopeak channel to the counts in a typical channel of the Compton continuum. This is usually taken to be a flat, representative portion just to the left of the Compton edge.

- **Peak to Total (P/T) ratio** – The peak-to-total ratio (P/T) is expressed as the ratio of the counts in the full-energy peak to the total counts in the spectrum.

- **Compton Suppression Factor (CSF)** – This is the ratio of P/C for unsuppressed and suppressed spectra, which also takes into account the reduction in photopeak efficiency as well as the suppression of the continuum:

\[
CSF = \frac{(P/C)_{\text{suppressed}}}{(P/C)_{\text{unsuppressed}}}
\]  

2.3. Measurement and analysis

Calibration sources were chosen to cover the 37–662 keV energy range to fully characterise the energy response and efficiency of the LEGe crystal. Several single γ emitters were used, as well as a NIST (National Institute of Standards and Technology) traceable complex γ source. The isotopes that comprised these were 241Am (59.54 keV), 109Cd (88.03 keV), 57Co (122.06 keV), 133Ce (165.86 keV), 133Ba (302.85 keV, 356.01 keV), 115Sn (391.68 keV), and 137Cs (661.67 keV). Apparent peak efficiencies and isotopic abundances were calculated as in Ref. [7].

Analysis was completed using the post-processor detailed in Ref. [6] to sort the data, and ROOT [8] for matrix manipulation and peak searching/fitting. Coincidences were identified by searching for all events in the NaI(Tl) detector with a time delay (τ) between −10μs and +10μs (with t=0 defined as the interaction time in the LEGe). This is achieved by reading the LEGe output file, and comparing the timestamp of each event with those in the NaI(Tl) output file. To speed up this process, the data is presorted by timestamp, and the software ‘remembers’ how far into the NaI(Tl) file it has read. This allows a full comparison to take place while only ever comparing a small subset of the data files. These coincidences were then recorded into a 3D matrix with the energy deposited in the LEGe, energy deposited in the NaI(Tl) detector, and the time delay on each axis (Eγ, Eγ, τ). This allows a delay spectrum to be created, with peaks identifying characteristic delays where there are a large number of coincident events. As they are in a matrix, time and energy gates can also be set to extract information from the dataset.
3. Results and discussion

3.1. Full Coincidence Search

Data was acquired for 1 h using a complex $\gamma$ source comprised mainly of $^{241}$Am, $^{109}$Cd, $^{57}$Co, and $^{137}$Cs. The energy spectra for the LGe with this source are shown below, in Fig. 2. The full delay window for this source (between $-10 \mu$s and $+10 \mu$s) is shown in Fig. 3.

In the time coincidence spectrum, there are four distinct regions: a well defined coincidence peak at $-2 \mu$s, a rising peak at $-5 \mu$s, the random coincidence background before the $-2 \mu$s peak, and a slightly increased coincidence background after the $-2 \mu$s peak. The random background coincidences are due to chance coincidences from the source and additionally from background radiation. The raised background is due to the time-walk of low energy interactions [9], which causes coincidences to appear later than they would at higher energies. The coincidence peak at $-\mu$s is the standard signal used for Compton suppression, and is the result of a photon scattering out of the LGe into the NaI(Tl) detector (LEGe→NaI(Tl)). As the NaI(Tl) scintillation process is relatively fast (and this interaction happens after the interaction in the Germanium crystal), the coincidence peak is well defined. The rising peak at $-5 \mu$s is far less well defined, which is due to the order of the interaction process. This peak represents events that have entered the NaI(Tl) detector first, and then interacted with the LGe (NaI(Tl)→LEGe). The ‘rising’ shape is caused by the rise time of a pulse in the LGe, which was set to $2.2 \mu$s (and is consistent with the rise time in the coincidence time distribution).

3.2. Single delay gate

In a traditional Compton suppression system, the time delay for rejecting pulses is set electronically, and this is performed during the data collection so that coincident events are discarded. This setup requires careful calibration, as the gate is critical to the coincidences collected. In the current experimental setup, it is trivial to collect the data due to the List-mode acquisition software [5], and all analysis (including setting and resetting delay gates) is performed after the experiment, with no loss of coincidence data.

For the complex $\gamma$ source, the delay gate was set from 1 $\mu$s to 3.0 $\mu$s to cover the main coincidence peak. The spectra extracted are shown in Fig. 4.

The coincidence spectra clearly shows Compton scattered photons from the various decay lines ($^{241}$Am at 59.5 keV, $^{57}$Co at 122.1 and 136.5 keV, $^{137}$Cs at 661.7 keV) in the complex source. Almost no full photopeak counts are seen in coincidence (which would only occur due to false coincidences). By suppressing these events, the suppression will match that of a typical electronic system, and the $P/C$ and $P/T$ ratios are substantially improved over the raw spectra.

3.3. Multiple delay gates

As there is another peak at $-5 \mu$s, additional suppression can be achieved if these events add to the background seen in the original spectra. The coincidences were extracted from the peak at $-6.2 \mu$s to $-3.2 \mu$s, and the resulting energy spectra are shown below, in Fig. 5.

Again, almost no full photopeak counts are seen in coincidence, however those events that are seen in coincidence are characteristic of the continuum from a higher energy decay or the natural background. As these events interact with the NaI(Tl) shield before the HPGe crystal, possible sources include Compton scattered photons (that interact with the NaI(Tl)) shield and then backscatter into the LGe), and Cosmic radiation such as high energy muons, fast and thermalised neutrons. A seven day background was taken to estimate the amount of coincidences caused by these events, and while some were seen, background radiation could only account for $\sim 0.8\%$ of the events in the 2 $\mu$s peak, and $\sim 0.5\%$ of the $-5 \mu$s peak. The extracted spectra when suppressing both of

\begin{align*}
\text{Fig. 2.} & & \text{The energy spectrum recorded in the LGe during one hour of data acquisition. For a full colour image please see the online version of this paper.} \\
\text{Fig. 3.} & & \text{The time distribution of the coincidences seen in the NaI(Tl) detector, when observing an event in the LGe at } t=0. \text{ For a full colour image please see the online version of this paper.} \\
\text{Fig. 4.} & & \text{The raw, coincidence and anticoincidence spectra for the complex } \gamma \text{ source, with the delay gate set on the 1.5$\mu$s to 3.0$\mu$s coincidence peak. Note, that due to the little amount of coincidence seen above 300 keV, the anticoincidence spectra overlaps with the raw spectra. For a full colour image please see the online version of this paper.} \\
\end{align*}
these coincidence peaks are shown in Fig. 6. All suppression ratios are detailed in Table 1.

With both coincidence peaks suppressed, the P/C, P/T, and CSF ratios are slightly improved for a range of nuclei in the low energy regime (typically by 2%–10%). The suppression ratios for a higher energy decay ($^{137}$Cs) are improved by up to 144%. This is because the continuum counts at this energy are fairly low, and therefore the background reduction is far more dramatic than at lower energies. In the complex $\gamma$ source used, there are higher energy decays. Compton suppression of these, however, cannot account for the reduction in the high energy background (when suppressing the −6.2 $\mu$s to −3.2 $\mu$s time gate), as these events would interact with the HPGe crystal first and produce the time delay signal seen at ≈2 $\mu$s.

3.4. Monte-Carlo simulations

To corroborate the findings in Table 1, and confirm the origins of the −5 $\mu$s peak, Monte-Carlo simulations were performed to determine the proportion of coincident events that would be expected to interact in the LEGe first, and vice versa. The simulations were carried out using the GEANT4 Monte Carlo toolkit developed at CERN (version 9.5.p1) [10], which enables the accurate simulation of the passage of particles through matter.

The detector geometry was reproduced as accurately as possible, including the LEGe, both NaI(Tl) detectors and the shielding. The LEGe contained the detector crystal, outer and inner electrodes, the aluminium canister and the carbon epoxy window. The NaI(Tl) detectors included the crystal, detector casings and Aluminium cylinders (to approximate the PMTs). To complete the model, the tin and copper lined lead shielding and source housings were also recreated.

The simulations are based upon a collection of many ‘events’. During each ‘event’, the probability of the primary detector ‘seeing’ multiple decays (within a typical charge collection time) is calculated, and one or more radioactive nuclei are then disintegrated according to known branching ratios [11]. Radiation transport during an ‘event’ is split into a number of steps, with the length of each step within the simulated geometry determined automatically according to the physics processes involved. The energy deposited in sensitive volumes (the detector crystals) and the volume it is deposited is recorded at each step during an event, allowing the order of the interaction process to be determined in the subsequent analysis.

The source was recreated in the simulation, and $10^6$ events were run. Coincident events in the GEANT4 simulations (when energy was deposited in LEGe and NaI(Tl) crystals during the same event) showed that ∼63% of coincidences were due to a LEGe→NaI(Tl) interaction path, with the remaining 37% interacting via NaI(Tl)→LEGe. This compares well to the value of 66% for the LEGe→NaI(Tl) path, and 34% via the NaI(Tl)→LEGe path seen in the data.

4. Conclusion

List mode acquisition has been used to run a full Compton suppression system with a low energy, hyperpure Germanium detector. This was achieved with no loss of coincidence data, and a greatly simplified experimental setup. Coincidence information has been successfully written to a 3D interaction matrix, allowing the efficient gating and extraction of events.

Analysis of the time delay spectrum also identified a coincidence peak before the interaction in the HPGe crystal (in addition to the standard Compton suppression peak). The energy spectra of these coincidences are also substantially different to that seen in the standard delay gate. A major source of these coincidences has been identified as photons that are scattered out of the active NaI(Tl) shield (and into the LEGe), and GEANT4 simulations of the detector setup agree with the proportion of coincident events seen in each delay peak. The geometry used for this particular Compton suppression system is highly sensitive to these effects, however similar event profiles are expected in all coincidence systems. The greatest benefit when using multiple time windows is seen with low activity (and therefore high relative background) sources.
Excellent suppression was found in the ‘standard’ Compton suppression configuration (with a single time gate), and a slight improvement on this was obtained at low energies using multiple time gates. A 144% improvement in the Compton Suppression Factor was achieved for the $^{137}$Cs peak when utilising multiple time gates to suppress coincident events.

Compton suppression may be more effective with larger detectors (for example, when using coaxial or broad energy designs), as the dead layers in the setup (including the detector casings) greatly reduce the detection efficiency for low energy Compton scattered photons. Incoming photons however, are also less likely to Compton scatter out of a larger detector, and the increased volume will cause more events of the type NaI(Tl)→LEGe to be seen. Multiple delay windows should therefore be considered in any coincidence system, as there may be substantial additional suppression to be gained.

Acknowledgements

Thank you to the University of Surrey, the AWE Technical Outreach Program and EPSRC for funding this research.

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Monte Carlo characterisation of a Compton suppressed broad-energy HPGe detector

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Abstract GEANT4 Monte Carlo simulations have been successfully utilised to characterise a Compton suppressed broad-energy HPGe detector. The detector setup has been fully recreated in the simulation, which has been optimised to consistently reproduce the detector response. The peak efficiencies for both the primary BEGe detector and NaI(Tl) guard detectors agree with the simulated values for multiple test sources within 3 %. Compton suppression has also been simulated, with good agreement seen between the simulated and actual CSF values (<10 %) for multiple radionuclides. A secondary reference source was also simulated, which contained up to 30 radionuclides in a different geometry to that of the previous source. This showed excellent agreement with experimental data in both unsuppressed and suppressed modes of operation.

Keywords GEANT4 · Monte Carlo · Gamma spectroscopy · Compton suppression

Introduction

High purity germanium (HPGe) detectors are routinely used for environmental analysis, and provide an unrivalled energy resolution and efficiency for identifying different radionuclides. The useful range for such analysis is from 30 keV–3 MeV, however many applications require increased sensitivity in the low energy (20 keV–1 MeV) region to detect specific radionuclides. These often have small branching ratios and the signals from such radionuclides are therefore easily swamped by the Compton continuum from higher energy decays [1, 2].

Broad energy HPGe (BEGe) crystals have several advantages over standard co-axial and low-energy designs in this range, including improved low-energy efficiency and charge collection at higher energies [3]. These improvements come at a cost; due to the size of the BEGe crystal the efficiency is typically lower than that of an equivalent co-axial HPGe crystal at higher energies, and many of these photons Compton scatter out of the crystal, only partially depositing their energy. It is therefore desirable to veto Compton scattered events, and to accurately characterise the detector response across the required energy range. Additional NaI(Tl) detectors and Monte Carlo simulations [4, 5] are used to achieve this, with the methods and results presented in the following report.

Materials and methods

Experimental setup

The BEGe detector (model BE3825) and associated electronics are supplied by Canberra UK (Harwell, Oxfordshire). The detector is optimised for maximum efficiency below 1 MeV, while still retaining the ability to collect γ radiation from higher energy decays. This is achieved by having a large area but relatively thin crystal (this particular model has an effective area of 38 cm² and is 25 mm thick). A carbon epoxy window also maximises photon and X-ray transmission (carbon reduces the response to photons below 10 keV when compared to a Beryllium window, however it is far more robust). An ultra-low background
cryostat (model 7915-30-ULB) is used, and a preamplifier (model 2002C) processes the initial signal data.

The preamplifier output is sent to a LYNX™ digital signal processing unit from Canberra, which controls all further amplification, pole-zero correction and digitisation of the pulse. Data is collected in ‘list mode’, which timestamps each event in the detector and writes these to a text file [6].

The NaI(Tl) shield detectors are from Scionix (Utrecht, Netherlands), and comprise of a cylindrical annulus with a removable plug that is positioned above the primary detector. Due to this geometry, the efficiency for suppressing Compton scattered events is very high, however there may be a large number of additional coincidence signals due to γ radiation emitted in cascade, and chance coincidences due to both the activity of the source and background radiation. Note that while the BEGe has a thin entrance window at the top, Compton scattered photons will typically exit the detector through the Aluminium casing, and may therefore be attenuated before entering the NaI(Tl) veto detector (through another thin layer of steel).

The cylinder has five photo-multiplier tubes (PMT’s), and the plug has a separate PMT for efficient charge collection. The outputs are combined from all of the PMT’s, and passed to a preamplifier (model 2005). Again, the preamplifier output is sent to a LYNX™ unit, where the data is collected in ‘list-mode’. A synchronisation cable also runs between the two LYNX™ units to allow synchronisation of the clocks used to record the events in each detector.

The BEGe and NaI(Tl) combination is placed within a 80 mm thick lead shield, with tin and copper liners to reduce fluorescence from lead and tin X-rays [7]. This reduces the background to ~2 counts per second (cps). A simulated image of the setup (including the lead cave, but excluding some internal detail of the detectors due to graphics limitations) is shown below in Fig. 1.

Measurement and analysis

Apparent peak efficiencies and isotopic abundances were calculated as in reference [8]. All data was analysed using the post-processor detailed in reference [9].

Calibration sources were chosen to cover the 10–2000 keV energy range to fully characterise the efficiency of the BEGe crystal. Several single γ emitters were used, as well as a National Institute of Standards and Technology (NIST) traceable complex γ source. The isotopes that comprised these were $^{241}$Am (59.54 keV), $^{109}$Cd (88.03 keV), $^{57}$Co (122.06 keV), $^{139}$Ce (165.86 keV), $^{113}$Sn (391.68 keV), $^{137}$Cs (661.67 keV), $^{54}$Mn (834.84 keV), $^{88}$Y (898.04 and 1836.06 keV), $^{60}$Zn (1115.54 keV), and $^{60}$Co (1173.23 and 1332.49 keV).

Analysis was completed using the post-processor to sort the data, and ROOT [10] for matrix manipulation and peak searching/fitting. Coincidences were identified by searching for all events in the NaI(Tl) detector with a time delay (t) between $-10$ and +10 μs (with $t = 0$ defined as the interaction time in the BEGe). These coincidences were then recorded into a 3D matrix with the energy deposited in the BEGe, the energy deposited in the NaI(Tl) detector, and the time delay on each axis ($E_{1,2}, t$). This allows a delay spectrum to be created, with peaks identifying characteristic delays where there are a large number of coincident events. As they are in a matrix, time and energy gates can also be set to extract information from the dataset.

Compton suppression

Several methods are available for quantifying the levels of suppression achieved [2], however the main ones used in this document will be the peak to count ratio (P/C), and the Compton suppression factor (CSF).

- P/C ratio This is defined as the ratio of the counts in the highest photopeak channel to the counts in a typical channel of the Compton continuum. This is usually taken to be a flat, representative portion just to the left of the Compton edge.
• **CSF** This is the ratio of P/C for suppressed and unsuppressed spectra, which also takes into account the reduction in photopeak efficiency as well as the suppression of the continuum;

\[
CSF = \frac{(P/C)_{\text{suppressed}}}{(P/C)_{\text{unsuppressed}}}
\]  

(1)

**GEANT4 simulation**

The simulations have been carried out using the GEANT4 Monte Carlo toolkit developed at CERN (version 9.5.p02) \([11]\), which enables the accurate simulation of the passage of particles through matter. The definition of the experimental setup (including all shapes, objects and materials) is of particular importance for the accuracy of the simulation, as low energy photons are highly susceptible to small errors in the geometry. The GEANT4 simulations are based upon similar models detailed in reference \([8]\), with the geometry changed to accommodate the different detector setup.

The description of the detector geometry consists of the BEGe, both NaI(Tl) detectors and the shielding. The BEGe contains the detector crystal, outer and inner electrodes, the aluminium canister and the carbon epoxy window. The cold finger and vacuum spaces within the detector are also included, as are the dead layers within the crystal. The NaI(Tl) detectors were recreated with a NaI(Tl) crystal, the detector casing and Aluminium cylinders (to approximate the PMT’s). This approximation has been shown to provide accurate results \([9]\), and will not affect the vetoing ability of the detectors. To complete the model, the tin and copper lined lead shielding and source housings were also recreated. NIST compounds were used where available, and all components were reproduced as accurately as possible according to manufacturer specifications.

The simulations are based upon a collection of many ‘events’. During each ‘event’, the probability of the primary detector ‘seeing’ multiple decays (within a typical charge collection time) is calculated, and one or more radioactive nuclei are then disintegrated according to known branching ratios \([12]\). Each event may also result in multiple photons due to nuclei emitting photons in cascade, and secondary radiation such as fluorescence, auger electrons and photon induced X-ray emission (PIXE). Radiation transport during an ‘event’ is split into a number of steps, with the length of each step within the simulated geometry determined automatically according to the physics processes involved. The energy deposited in sensitive volumes (the detector crystals) and the volume it is deposited in is recorded at each step during an event, and written to a file. Due to this setup, the energy deposited in all detectors for each event can be determined, and appropriately binned to create realistic spectra. If energy is deposited in multiple sensitive volumes during a single event, these are treated as coincident (radiation transport through the geometry happens on a much faster timescale than charge collection in the crystals). The recording of energy deposition at ‘step’ level also allows the order of the interaction process to be determined.

**Results and discussion**

**Delay window analysis**

A delay spectrum was created by searching the data for coincidences between \(-10\) and \(+10\) µs (see Fig. 2). As in reference \([13]\), multiple coincidence peaks were found when analysing this data, indicating the presence of multiple event types occurring with characteristic delays. When extracting the coincidence spectra for each detector, the time window was further refined to only include events seen within the main coincidence peak.

The peak in the delay spectrum at \(\sim 0\) µs is a convolution of a rising sawtooth shape peak, and a well defined, Gaussian shaped peak. The Gaussian peak is the standard signal used for Compton suppression, and is the result of a photon scattering out of the BEGe into the NaI(Tl) detector (BEGe \(\rightarrow\) NaI(Tl)) \([13]\). As the NaI(Tl) scintillation process is relatively fast (and this interaction happens after the interaction in the Germanium crystal), the coincidence peak is well defined. The rising sawtooth shaped peak is far less well defined, which is due to the order of the interaction process. This peak represents events that have entered the NaI(Tl) detector first, and then interacted with the BEGe (NaI(Tl) \(\rightarrow\) BEGe). The ‘rising’ shape is caused by the rise time of a pulse in the BEGe, which is consistent with the rise time seen in the coincidence time distribution.
Other possible sources of these events include high energy terrestrial background radiation, and cosmic radiation such as high-energy muons. The previous study [13] has shown that for this geometry, background events make up <1.0 % of the events in the convoluted coincidence peak, and that ~66 % of the events are due to a photon interacting via the BEGe → NaI(Tl) path, and 34 % via the NaI(Tl) → BEGe path. Although this was for a different primary detector (a low-energy HPGe crystal), these values compare well with those seen in the current simulations (68 % due to a BEGe → NaI(Tl) interaction path, and 32 % via a NaI(Tl) → BEGe interaction path).

Efficiency comparisons

Peak efficiencies were calculated for multiple sources, and are compared to the simulated values in Figs. 3, 4.

![Graph 3](image1.png)

**Fig. 3** The actual and simulated peak efficiencies for the BEGe detector. A variety of sources were each counted for 1 h to minimise statistical error. For a full colour image please see the online version of this article.

![Graph 4](image2.png)

**Fig. 4** The actual and simulated peak efficiencies for the NaI(Tl) detector. A variety of sources were each counted for 1 h to minimise statistical error. For a full colour image please see the online version of this article.

### Table 1 A summary of the suppression ratios calculated for the BEGe with the delay gate set on the ~0 μs peak

<table>
<thead>
<tr>
<th>Nuclide (energy in keV)</th>
<th>CSF actual</th>
<th>CSF simulated</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}$Am (59.5)</td>
<td>1.26</td>
<td>1.14</td>
<td>0.91</td>
</tr>
<tr>
<td>$^{109}$Cd (88.0)</td>
<td>2.61</td>
<td>2.47</td>
<td>0.95</td>
</tr>
<tr>
<td>$^{57}$Co (122.1)</td>
<td>2.88</td>
<td>3.17</td>
<td>1.00</td>
</tr>
<tr>
<td>$^{57}$Co (136.5)</td>
<td>1.99</td>
<td>2.12</td>
<td>1.06</td>
</tr>
<tr>
<td>$^{137}$Cs (661.7)</td>
<td>2.61</td>
<td>2.79</td>
<td>1.07</td>
</tr>
<tr>
<td>$^{60}$Co (1173.2)</td>
<td>1.68</td>
<td>1.67</td>
<td>1.00</td>
</tr>
<tr>
<td>$^{60}$Co (1332.5)</td>
<td>1.06</td>
<td>1.07</td>
<td>1.01</td>
</tr>
</tbody>
</table>

These are compared to the simulated values from the GEANT4 model, and a ratio of the simulated to actual values is given in the final column. All values agree to within 10 %, and the errors range from 4 to 12 % (at 1 sigma). These are dominated by the contribution from the number of counts in the continuum, which may contain as few as 20 counts per channel.

Excellent agreement was found across the energy range for the BEGe detector, with the detector response accurate to within 3 % for the efficiency of each peak.

In the case of the NaI(Tl) guard detector, only a limited number of efficiency points could be used as many peaks were unresolvable (the detectors poor resolution was due to both the crystal material and the difficulty in gain matching multiple PMT’s). The efficiencies for the NaI(Tl) detector are accurate to 3 % at 661.66 keV and over, however the GEANT4 model overestimates the NaI response at low energies. This is due to the gain setting for the NaI(Tl) detector, which notably reduced the signal rate below 100 keV.

For $^{60}$Co, significant amounts of cascade summing were seen due to the geometry of the NaI(Tl) detector. To correct for this, a cascade summing correction factor was calculated using GEANT4 as in [14], which gave a value of 0.23 ± 0.01. When applied to the data, the efficiency for the $^{60}$Co source matched that of the GEANT4 calculated peak efficiencies. This provides further evidence that the model accurately reproduces the detector response, as cascade correction factors are highly sensitive to the detector geometry.

**Compton suppression**

Both detectors are accurately (individually) characterised, however they must perform together to achieve Compton suppression. A source containing $^{241}$Am, $^{109}$Cd, $^{57}$Co, $^{137}$Cs, $^{133}$Cs and $^{60}$Co was used to calculate CSF’s for the Compton suppression system. The same factors were calculated for the simulated results, and these are compared in Table 1. Good agreement was found between the CSF values for actual and simulated data, and the unsuppressed and suppressed spectra for the γ source used are shown in Figs. 5, 6 respectively.
Both the unsuppressed (Fig. 5) and suppressed (Fig. 6) spectra show excellent agreement between the simulated and actual data for this source. The continuum is slightly underproduced in the suppressed (simulated) spectra, and this is due to the over-suppression of the continuum in the simulations. This is primarily caused by the activity of the source ($\sim 3$ kBq); as the source activity increases, the dead time of the acquisition system can cause it to miss coincident events [4]. When calculating the peak to count ratios, a large variation was seen in the continuum where the number of counts per channel was low ($\sim 20$). To mitigate this, continuum counts were averaged over several bins, which greatly reduced the variation seen. All CSF values agree within 10%.

**Simulation of a realistic source**

To verify the accuracy of the GEANT4 simulation, a reference source was also simulated. This was comprised of a thin 12 mm diameter filter placed upon a 25 mm diameter steel disc. The isotopic composition was known to within 5%, and recreated within the GEANT4 simulation. The source included up to 30 distinct isotopes, which were primarily fission fragments with a combined activity of $\sim 6$ kBq. The unsuppressed and suppressed spectra are shown in Figs. 7, 8.

Excellent agreement was found between the simulated and actual results, however accidental summing between multiple events (due to the activity of the source) was not included in these simulations. This is because by default, GEANT4 decays radioactive isotopes until a stable isotope is reached, which therefore includes all subsequent emissions in a decay chain. You can limit this by specifying an end to a decay chain (typically the isotope that the user wishes to decay), however this cannot be applied to the complex sources here due to the way primary events are created in the simulations. The events from each nuclide were therefore simulated separately (where this limit could
be applied), and brought together in post-processing to produce the final dataset.

The original anti-coincidence spectra (Fig. 8) initially showed poor agreement with the data from the Compton suppression system. This was due to both the low (<150 keV) and high (>1.6 MeV) energy coincident events being missed by the NaI(Tl) crystal due to the gain settings of the electronics. The lower energy events were missed due to difficulties in gain matching multiple PMT’s, while the higher energy events were missed as they were simply outside of the electronics acquisition range. Both of these event types were still classed as coincident in the GEANT4 simulations, causing an over-suppression of coincident events. A close up of the peaks are shown in the inset. For a full colour image please see the online version of this article.

The lack of accidental coincidences in these simulations will increasingly affect the accuracy of the model as the count rate increases (and sum events between uncorrelated emissions become more likely), however this is a small and often negligible effect in environmental samples. As with the previous source, the continuum was slightly over-suppressed in the simulation, which is likely due to a combination of the dead time in the detectors and inefficiencies in the acquisition electronics (the coincidence acquisition system also has a small (<2 %) inherent dead time). Given these limitations, simulations to predict the performance of a coincidence system should be treated as a best case scenario, as several factors may artificially limit the suppression achieved. It is worth noting that with standard electronics (running an online veto), the dead time can be greatly reduced as the incoming pulse need not be shaped, but simply used as a logic pulse to veto coincident events.
Conclusion

GEANT4 Monte Carlo simulations have been successfully utilised to characterise a Compton suppressed broad-energy HPGe detector. The detector setup has been fully recreated in the simulation, which has been optimised to consistently reproduce the detector response. The peak efficiencies for both the BEGe and NaI(Tl) detectors agree with the simulated values for multiple test sources within 3%, and cascade correction factors calculated using the model are consistent with the experimental data.

Compton suppression has also been simulated, with good agreement seen between the simulated and actual CSF values (<10%) for multiple radionuclides. Simulations were also performed of a complex γ reference source, which contained up to 30 radionuclides with a total activity of 6 kBq. Excellent agreement was found for the detector response in both the suppressed and unsuppressed modes.

The model has been shown to be highly accurate, and can reproduce many aspects of the spectrum seen when performing environmental analysis. This includes peak efficiencies, the Compton continuum, true-coincidence (cascade) summing, and Compton suppression for a variety of source matrices and geometries. The accuracy of the model may be reduced when source activities increase, however this should not be an issue in the low activity regime that is typically measured. It has also been found that the effects of the systems electronics must be taken into account to fully reproduce the coincidence behaviour of the detectors. This model can now be used to fully characterise the detector response for any environmental source.

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References

Maximising the sensitivity of a γ spectrometer for low-energy, low-activity radionuclides using Monte Carlo simulations

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Monte-Carlo simulations have been utilised to determine the optimum material and thickness for a γ spectrometer to be used for the assay of radionuclides that emit radiation in the 50–300 keV energy range. Both HPGe and LaBr₃(Ce) materials were initially considered for use, however the additional background radiation and lack of resolution in the latter drove the selection of HPGe for further optimisation. Multiple thicknesses were considered for the HPGe detector, with the aim of improving the sensitivity of the system by maximising the efficiency for low energy emissions, and reducing the probability of interaction with (and therefore the continuum from) higher energy photons. The minimum amount of material needed to achieve this was found to be 15 mm for a source that is dominated by high energy (>2.614 MeV) photons, and 20–30 mm for a typical reference source (with photons of energy 59.54 keV–2.614 MeV).

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1. Introduction

Gamma (γ) spectroscopy is utilised for the non-destructive assay of radioactive materials in a variety of applications, including (but not limited to) environmental analysis (Kapsimalis, 2009; Habib et al., 2013), fundamental physics research (Alharbi et al., 2013; Mason et al., 2013) and the monitoring of the Comprehensive Nuclear-Test-Ban Treaty (CTBT) (CTBTO, 2011). It is important to maximise the efficiency of such systems, as this subsequently increases the sensitivity of the detectors, allowing either smaller amounts of material to be reliably identified, or count times to be decreased, increasing measurement throughput.

The CTBT network consists of seismic and atmospheric monitoring stations across the globe, which monitor for 85 key radionuclides that may be indicative of nuclear weapons tests and/or reactor incidents, including 140Ba, 95Zr, 99Mo, 141Ce, 144Nd, 131I, 134Cs and 137Cs (CTBTO, 2000). Note that these are often very small signals amongst the natural background present in the atmosphere, which is dominated by isotopes from the natural Uranium (238U) and Thorium (232Th) decay chains, and cosmogenic radionuclides produced by Cosmic radiation (e.g. 7Be). In particular, 241Am (59.54 keV), 144Ce (133.52 keV), 95mTc (140.51 keV), 141Ce (145.44 keV), 235U (143.76 keV, 185.72 keV), and 95mNb (235.68 keV) are difficult to accurately detect and quantify due to the Compton continuum from higher energy γ-rays, which may obscure lower energy (and low activity) signals. One possibility to increase the detectors sensitivity in this range is to reduce the thickness of the detector material, which detrimentally affects detection efficiency for higher energy photons far more acutely than those of a lower energy.

This document evaluates a number of designs for a proposed environmental γ spectroscopy system, optimising the material and thickness of the detector crystal to achieve the highest sensitivities for those radionuclides that may be obscured by higher energy photons.

2. GEANT4 simulations

All simulations were carried out using the GEANT4 Monte Carlo toolkit developed at CERN (version 9.6.p02) (Agostinelli, 2003). These were based upon previous simulations (Britton et al., 2012a,b), that were validated with experimental data, however the geometry has been modified to create each respective detector configuration evaluated in this work.

The description of the detector geometry consists of the primary crystal, the aluminium canister and the carbon epoxy window (if...
3. Material choice

For γ spectroscopy both scintillation and semi-conductor type crystals can be used, with the most common of these being NaI(Tl) and HPGe respectively. Scintillation crystals use the process of prompt fluorescence to convert the kinetic energy of an incoming radiation (Compton electrons in case of γ-rays) into fluorescence photons, which are then collected by a photomultiplier that acts to amplify this signal, allowing it to be measured using suitable electronics. Semi-conductor based detectors rely on the creation of electron–hole pairs in the material, which are accelerated and collected by applying a large electrical bias.

As a result of these differences, semi-conductor based materials typically have a far better energy resolution than scintillators due to the relatively small amounts of energy required to create an information carrier. In scintillators, this can be of the order \( \sim 100 \text{ eV} \) (Knoll, 2010), and therefore the inherent statistical uncertainty in the number of information carriers limits the resolution of the detector. The most common scintillation based crystals used include NaI(Tl), LaBr\(_3\)(Ce), and BGO, while the semi-conductor based materials are dominated by HPGe and Silicon. Most scintillators can be made in larger volumes than semi-conductor detectors, and have good efficiency across a large energy range. Semi-conductor crystals are generally far more expensive than scintillation crystals (per unit volume), and in the case of HPGe have to be substantially cooled to reduce the excitation of electrons across the band gap, which renders HPGe unusable at room temperatures.

For the assay of environmental samples, there are several requirements for a detection system. The detector crystals must be able to be produced in large sizes, with a high Z material to maximise the efficiency of the system. The material must be stable and have low levels of radioactive contamination, and the resolution must be good enough (\( \leq 3\% \)) to separate multiple peaks within a complex γ spectrum. The detector does not have to be particularly fast (one of the main advantages of LaBr\(_3\)(Ce), which has a light decay time of 35 ns (Rulísek, 2007)), due to the low count rates experienced.

These requirements rule out NaI(Tl) and BGO, which have resolutions of \( \sim 7\% \) and \( \sim 16\% \) respectively, and Silicon detectors are primarily of interest in the 5–50 keV region due to their low Z value and the sizes of crystal available. This leaves HPGe and LaBr\(_3\)(Ce), which have resolutions of \( \sim 0.2\% \) and \( \sim 3\% \) respectively. LaBr\(_3\)(Ce) does have a substantial internal background from \(^{138}\text{La} \) (788.7 keV γ from β decay, and a 1435.8 keV γ from e\(^-\) capture), \(^{227}\text{Ac} \) (which decays via a series of five α decays to \(^{207}\text{Pb} \)), and Barium K X-rays from 31 to 38 keV (Hartwell and Gehrke, 2005; Milbrath et al., 2005), however it can be used at room temperatures, allowing a much simpler system than HPGe (which requires cooling apparatus often several times the size of the detector itself).

Simulations were developed to evaluate the full photopake efficiency of both HPGe and LaBr\(_3\)(Ce) materials with respect to both material thickness and photon energy. Up to 400 simulations were performed for each material, and the results are presented as efficiency maps in Figs. 2 and 3. Note that the efficiency is shown as a contour (thin lines across the plot), and as a graded colour map to allow some interpolation of the contours.

Figs. 2 and 3 provide minimum sizes to detect photons of a variety of energies. Both materials are effective absorbers, however LaBr\(_3\)(Ce) outperforms HPGe due to its larger Z number. Despite the combined advantages of additional efficiency and room temperature operation, the detector performance does not offset the reduced resolution and increased background levels seen in a
LaBr₃(Ce) crystal. HPGe will therefore be selected as the primary crystal, but the authors would like to note that in a γ–γ system where coincidences are utilised (reducing the importance of the resolution and additional background), LaBr₃(Ce) would be the crystal of choice.

4. Crystal design

Due to the cooling requirements of HPGe, the crystal itself must be in contact with a heat sink (often a Copper based ‘cold-finger’). Usually (in a co-axial design) this is emplaced within a cutout in the crystal, with the electrodes on the outer and inner surfaces of the crystal respectively. Depending on the electrode design, this can result in a relatively thick (<700 μm) dead layer either on the outside or the inside of the crystal, which will attenuate low-energy photons. The electrodes also have to be carefully designed, as poor charge collection will reduce the resolution and performance of the detector. For this particular system, a large-area planar crystal is required to maximise efficiency for low-energy photons, while minimising interactions with higher energy γ’s. The outer dead layers must also be minimised, with only a small volume of the crystal lost to the heat sink.

The closest commercially available detectors that follow these design principles are the Broad-energy range from Canberra UK (Harwell, Oxfordshire), which can operate from 3 keV to 3 MeV (Canberra UK, 2014). Ortec (Oak Ridge, Tennessee) also produce a planar HPGe detector (the SLP and GLP range), however these are designed specifically for low energy (<300 keV) detection (ORTEC, 2014). Planar crystals differ from the traditional co-axial design as they use a small electrode implanted into the back of a thin crystal. They also have a very thin external dead layer (<1 μm), increasing sensitivity to low energy photons. As the Canberra design has been shown to work (from a charge-collection and engineering point of view), and has a greater energy range (useful when requiring greater flexibility from a detection system), this will be used as a basis for the following simulations.

5. Minimum Detectable Activity (MDA)

The Minimum Detectable Activity (MDA) of a detector is an energy specific measure of the activity required to identify a radiation source with an amount of statistical certainty (normally 95%).

A widely used form of MDA is the Currie equation (Knoll, 2010). This evaluates the level of counts needed from a source to ensure a false-negative and false-positive rate of no greater than 5%. This statistical level can be changed by modifying the number of standard deviations from the mean that the equation is based on (5% uses a standard deviation of 1.64). The MDA can then be calculated in the following way:

\[
N_B = 4.65 \sqrt{N_B} + 2.71
\]  

(1)

\[N_B\] is the number of counts required for statistical certainty, and \(N_B\) the number of background counts. To convert this value to a minimum detectable activity (α), additional factors for the branching ratios (b), detection efficiency (ε), and the counting time (t) must be considered:

\[
\alpha = \frac{N_B}{bt}
\]  

(2)

Note that the smaller α is, the better. The MDA performance of a detector is therefore proportional to the detectors efficiency, and inversely proportional to the square root of the number of background counts across the peak.
When calculating the MDA for each radionuclide, the background is defined as any energy deposition that is not part of the full energy photopeak for that particular γ-emission. In a sample with multiple radionuclides, this includes the full energy photopeaks and Compton continuum from other photons. To accurately calculate the MDA, it is therefore important to have a representative background.

To simulate the background for each detector thickness, two reference sources were created using GEANT4. The first represents a worst case scenario for the continuum measured, where the background is dominated by photons from the highest energy γ emitter routinely measured, 208Tl (2.614 MeV). As the proportion of low and medium energy photons Compton scatter out of the crystal is greater at higher energies, the continuum that results from such emissions will be maximised. The second source contained multiple radionuclides that may be seen in CTBT reference samples, including γ emitters such as 40K, 106Zr, 133Cs, 137Cs, 144La, 140Ba, 208Tl, 210Pb, and 226Ra. An average activity of 3 kBq was calculated from a number of such samples in the laboratory, and then used to estimate the number of radioactive disintegrations necessary to simulate a 7 day acquisition period.

The source geometry was defined as a compressed air filter (a geometry commonly used for CTBT measurements, 70 mm diameter and 26 mm thickness). When simulating photons of specific energies, photons were emitted in a randomised isotropic direction from this geometry, otherwise the ‘G4RadioactiveDecay’ module was used to fully recreate each radioactive decay in the simulation.

### 7. Optimising the thickness of the crystal

Simulations were initially carried out using both the 208Tl and CTBT background sources, with the background spectra generated for every thickness of detector simulated (which ranged from 1 mm to 100 mm). Further GEANT4 simulations were then carried out to establish the full photopeak efficiency for each γ-ray of interest (note that these fired γ radiation with no coincidence information, and so cascade summing effects are not included in these calculations). By combining the efficiency, branching ratio and background information with the length of acquisition, an MDA can then be calculated for each nuclide of interest and detector thickness. Simulated background spectra for the CTBT source are shown in Fig. 4.

Increasing the thickness of the HPGe detector clearly improves the overall efficiency of the system, however there is a limit to the improvement that is achievable, with little efficiency gain seen above a detector depth of 50 mm for this particular source. The increased efficiency at larger thicknesses drives up the total number of events seen, and therefore the MDA for lower energy radionuclides. Efficiencies were generated for each radionuclide and material thickness, and are shown in Fig. 5.

By combining the efficiencies calculated and the background seen, the MDA’s can then be calculated for the radionuclides of interest (Figs. 6 and 7).

From the MDA’s calculated for a range of nuclei, it is clear that there is no or very little detrimental effect on the nuclides of interest when increasing the crystal size. For the pure 208Tl source (which represents the worse case scenario due to the high energy γ decays), a minima in the calculated MDA is seen at ~ 15 mm for a range of nuclei, however the subsequent increase seen in MDA with increasing crystal thickness is minimal. For the CTBT reference source (which is much more typical of a reference sample), no minima is found in the calculated MDA’s, with the sensitivity achievable levelling off past 20–30 mm.

The overall ‘background’ with each source increased as expected, however only to a limiting point where the efficiency is then dominated by the radius of the crystal, not the depth. Also, at lower energies (where the continuum level is most crucial for the nuclides of interest) the ‘background’ levelled off after 30–40 mm of HPGe. This is due to a combination of effects that act to stabilise the continuum seen. With increased crystal thickness, higher energy events are more likely to interact with the detector, increasing the Compton continuum as these photons scatter out of the crystal. The proportion of low and medium energy photons Compton scattered out of the crystal, however, will fall due to the increased detector material, therefore reducing the continuum.

The MDA’s achieved for each permutation of detector are minimised for the nuclides of interest with a 30 mm HPGe crystal, and there is no obvious benefit (or detriment) to increasing the crystal depth further. The only nuclide that showed a different behaviour with the CTBT source is 241Am, where the energy of the γ-emission is far lower than that of the other nuclides evaluated. Again, however, there was very little detrimental effect to increasing the crystal depth.
For higher energy decays, increased crystal depth has obvious benefits. For this particular system however, it is recommended that the crystal depth does not exceed 30 mm, as no additional sensitivity can be gained, and larger crystals will suffer from increased interaction from conventional background sources, such as the cosmic muon and neutron flux.

8. Conclusion

Monte-Carlo simulations have been utilised to determine the optimum material and thickness for a $\gamma$ spectrometer to be used in environmental monitoring. HPGe and LaBr$_3$(Ce) were initially considered for use, however the additional background radiation and lack of resolution in the latter drove the selection of HPGe for further optimisation.

Multiple thicknesses were considered for the HPGe detector, with the aim of maximising the sensitivity of the system for radionuclides in the 50–300 keV energy range. By restricting the thickness of the HPGe crystal to 30 mm (currently the largest thickness routinely produced for the crystal design considered), the system has been optimised to both reduce the continuum from higher energy photons, and maximise the efficiency of the detector. While no obvious detrimental effects were observed when increasing the crystal depth past this size, it is recommended to minimise the amount of HPGe used as an increased volume of material will suffer from a greater interaction rate with the cosmic and terrestrial background.

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References

Monte-Carlo optimisation of a Compton suppression system for use with a broad-energy HPGe detector

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A B S T R A C T

Monte-Carlo simulations are used to evaluate and optimise multiple components of a Compton Suppression System based upon a Broad-energy HPGe primary detector. Several materials for the secondary crystal are evaluated, including NaI(Tl), BGO and LaBr3(Ce). BGO was found to be the most effective across the required energy range, with the sizes of the proposed veto detector then optimised to extract the maximum performance for a given volume of material. Suppression factors are calculated for a range of nuclides (both single and cascade emitters) with improvements of 2 for the Compton Suppression Factors, and 10 for the continuum reduction when compared to the Compton suppression system currently in use. This equates to a reduction in the continuum by up to a factor of ~240 for radionuclides such as 60Co, which is crucial for the detection of low-energy, low-activity γ emitters typically swamped by such a continuum.

1. Introduction

High-resolution gamma spectroscopy is often utilised for the non-destructive assay of radioactive materials. It has many applications, which include food testing [1], neutron activation analysis [2,3], fundamental physics research [4,5] and environmental analysis [6,7].

High-purity Germanium (HPGe) crystals are often selected as the primary detector for these applications due to their excellent resolution, high-Z value and the large crystal sizes available, however there are several constraints on the performance of such systems. The background seen (which has both terrestrial and cosmic components [8]) can obscure spectral detail, and the charge collection time is slower than that of a typical scintillation type detector, possibly limiting HPGe’s use in high-count rate applications. Another major component of the spectra which reduces the detectors sensitivity is the Compton continuum. This arises due to partial energy depositions from incident γ radiation Compton scattering out of the crystal [9]. Detector crystals with higher Z numbers (or a larger crystal volume) will reduce the amount of scattered photons that escape the crystal, however as Germanium satisfies both of these conditions already, alternative techniques must be considered to reduce the continuum seen.

Anti-Coincidence Compton suppression is one such technique, which uses a secondary detector to capture escaping γ radiation. If the detectors are time-synchronised, it is possible to identify coincident events which can then be vetoed from the final dataset. This report discusses a variety of possible designs for a Compton Suppression system that is specifically optimised for 20 keV–1.5 MeV, low-activity environmental samples. Monte-Carlo simulations, and in particular, the GEANT4 toolkit [10] are used to achieve this, and compare multiple geometries and materials for the secondary veto detector.

2. Compton suppression

There are three main methods of Compton Suppression; Anti-coincidence, sum coincidence, and pair spectroscopy [11]. Pair spectroscopy focusses on the observation of two 511 keV annihilation photons from a positron decay. This produces very clean spectra, however the efficiency is low and this type of measurement is obviously restricted to nuclei which emit high energy γ’s. Sum coincidence assumes that the secondary detector captures the escaping γ completely, and therefore rebuilds the original event by summing the energies seen in each crystal. This is often used in large γ arrays, with highly segmented HPGe crystals and summing over many different detectors. Anti-coincidence is the standard method of Compton Suppression, and is the most feasible for detectors that perform environmental monitoring. This is due
to the compactness of the system; it can be packaged such that the
detectors can be contained within a low-background lead shield,
with the primary detector still available to count samples in close
geometries to maximise efficiencies.

2.1. Quantifying the levels of suppression

Several methods are available for quantifying the levels of
suppression achieved, however the main ones used in this docu-
ment will be the Peak to Count ratio (P/C), Peak to Total ratio (P/T),
and the peak and total Compton Suppression Factors (CSF).

- P/C ratio: This is defined as the ratio of the counts in the highest
  photopeak channel to the counts in a typical channel of the
  Compton continuum. This is usually taken to be a flat, rep-  
  resentative portion just to the left of the Compton edge.
- Peak to total (P/T) ratio: The peak-to-total ratio (P/T) is
  expressed as the ratio of the counts in the full-energy peak to
  the total counts in the spectrum.
- Peak CSF: This is the ratio of P/C for suppressed and unsuppressed
  spectra, which also takes into account the reduction in photopeak
  efficiency as well as the suppression of the continuum.

\[
\text{CSF}_\text{P} = \frac{P/C_{\text{suppressed}}}{P/C_{\text{un-suppressed}}}, \tag{1}
\]

- Total CSF: This is the ratio of P/T for suppressed and unsup-
  pressed spectra.

\[
\text{CSF}_\text{T} = \frac{P/T_{\text{suppressed}}}{P/T_{\text{un-suppressed}}} \tag{2}
\]

2.2. Current designs

The Compton Suppression system currently used [12] has a
Broad-Energy HPGe (BEGe) primary detector, which is surrounded
by a NaI(Tl) cylindrical annulus. An additional removable NaI(Tl)
plug is positioned above the primary detector, allowing sources to
be inserted and removed from the system. Preamplifier outputs are
sent to LYNX™ digital signal processing units from Canberra
(one for each detector), which controls all further amplification,
pole-zero correction and digitisation of the pulse. Data is collected in
‘list mode’, which time-stamps each event in the detector and
writes these to a text file [13]. A synchronisation cable also runs
between the two LYNX™ units to allow synchronisation of the
clocks used to record the events.

The BEGe and NaI(Tl) combination is placed within a 80 mm
thick lead shield, with tin and copper liners to reduce fluorescence
from lead and tin x-rays. This reduces the background to ~2
counts per second across the 20 keV–2.5 MeV energy range. A
simulated image of the setup is shown below in Fig. 1.

Due to this geometry, the efficiency for suppressing Compton
Scattered events is very high, however there may be a large
number of false coincidences due to γ radiation emitted in cascade,
chance coincidences due to the activity of the source, and to a
lesser extent, background radiation. Note that while the BEGe has
a thin entrance window at the top, Compton scattered photons
will typically exit the detector through the Aluminium casing, and
may therefore be attenuated before entering the NaI(Tl) veto
detector (through another thin layer of Steel).

2.3. Overview of materials considered

The secondary detector crystals must be able to be produced in
large volumes (> 100 cm³), allow relatively fast signal formation
(as the volumes will be large, the count rate will be high), and the
Z values and material densities must also be as high as possible to
reduce the amount of material needed to absorb scattered γ
radiation. As all detectors must fit inside the lead shield, plastic
scintillators were ruled out (due to their low Z-values, and
large resulting sizes). To minimise cost and maximise efficiency,
NaI(Tl), BGO, and LaBr₃(Ce) scintillators were selected for further
evaluation.

To test each material effectiveness as a detector, simulations
were created in which the number of interactions could be
recorded as a function of both impinging γ energy and material
thickness. By combining the results of each simulation set, the
materials effectiveness as a detector can be depicted below, in
Figs. 2–4.

BGO is clearly the most effective material for interacting with
γ’s across the entire energy range. To interact with 50% of
impinging γ’s at 1 MeV, the amount of material required would
be ~30 mm of NaI(Tl), ~21 mm of LaBr₃(Ce), or ~13 mm of BGO.
For the current design (which contains 80 mm of NaI(Tl)), the
same performance (for γ’s of energy 1 MeV) could be obtained
with ~52 mm of LaBr₃(Ce), or ~38 mm of BGO. Alternatively, the
performance of the NaI(Tl) shield (again for 1 MeV photons) could
be improved by up to 14% if LaBr₃(Ce) was used, or 25% if BGO
was used.
Fig. 2. The efficiency of an interaction (not necessarily a full energy deposition) for a photon within a NaI(Tl) crystal for a range of photon energies and material thicknesses (absorption lengths). While low energy photons (\(< 100 \text{ keV}\)) are absorbed with high efficiency in only a few mm of material, 1 MeV photons require \( \sim 145 \text{ mm of NaI(Tl)} \) to interact with a probability \( > 95\% \). For a full colour image see the online version of this paper.

Fig. 3. The efficiency of an interaction (not necessarily a full energy deposition) for a photon within a LaBr\(_3\)(Ce) crystal for a range of photon energies and material thicknesses (absorption lengths). LaBr\(_3\)(Ce) has a greater stopping power than NaI(Tl), and a minimum of 100 mm of material would be required to stop 1 MeV \( \gamma \)'s with a 95\% probability. For a full colour image see the online version of this paper.

Fig. 4. The efficiency of an interaction (not necessarily a full energy deposition) for a photon within a BGO crystal for a range of photon energies and material thicknesses (absorption lengths). Medium energy photons (\(< 500 \text{ keV}\)) are absorbed with far higher efficiency than for NaI(Tl) or LaBr\(_3\)(Ce), and 1 MeV photons require up to \( \sim 65 \text{ mm of BGO} \) to interact with a probability \( > 95\% \). For a full colour image see the online version of this paper.
Despite NaI(Tl) performing the worst of the three materials, it is the cheapest and most widely available. Resolution for the three materials ranges from 3–4% for LaBr$_3$(Ce), 6–8% for NaI(Tl), and ~15% for BGO, however LaBr$_3$(Ce) is currently restricted in size to 75 × 75 mm crystals. LaBr$_3$(Ce) also has much faster timing properties, with a typical decay time of 35 ns [14], compared to 230 ns for a NaI(Tl) crystal, and 300 ns for a BGO crystal [9].

If the secondary detector is used purely as a veto, then BGO is the best option, as this minimises the material needed for the veto, and maximises interaction efficiency. If the secondary crystal is also required to output a usable spectra, then (if 75 mm crystals are efficient enough) LaBr$_3$(Ce) would be a better choice due to its impressive energy resolution, timing characteristics, and stopping power. Where crystals are required to be more efficient than 75 mm of LaBr$_3$(Ce), the best compromise would use NaI(Tl) to veto/record coincident events. For this work, the suppression system is to be used solely in veto mode, with no spectral information required. BGO will therefore be selected as the principle material for the secondary crystal.

2.4. Overview of the design considered

Several possible designs were considered, and infeasible, problematic or undesirable designs were ruled out. These included conical secondary detectors, as there is no need to fit many Compton Suppressed HPGe detectors in close geometry around a single focal point. Such a requirement is useful for large arrays of detectors, but in the domain of single HPGe crystals it would only serve to artificially limit the efficiency of the secondary detector. Any designs that required a source to be collimated, or that prevented a source from being placed in direct contact with the primary detector were also eliminated, as these would severely limit the efficiency of the system (which is the most critical factor when counting environmental radiation). Any designs that were too large to fit into a standard Pb cave (of inner diameter 230 mm) were also eliminated, as this is required to substantially lower the background seen in the detector system. The largest environmental source that must fit within the system is a compressed air filter, which can measure up to 70 mm diameter × 26 mm thickness.

Re-designing the primary detector was considered, which is an especially attractive prospect when considering the possible advantages of creating a veto system with no dead layers between primary and secondary crystals. This idea was rejected however, as not only is the design and manufacture of such a system fraught with engineering challenges, but the removal of the copper crystal holder and cold finger (which make up the majority of the dead material within the immediate vicinity of the HPGe detector) would require the entire apparatus to be cryogenically cooled for the operation of a HPGe detector. The system envisaged will instead use a commercially available primary detector that has been designed to minimise dead layers and photon attenuation within the system.

Three main components of a veto detector are considered for this project, including the main cylinder around the primary detector, a base plate for forward scattered γ radiation [15], and a lid for backscattered radiation. All designs are based upon BEGe 5030 primary detector from Canberra UK (Harwell), which has a 110 mm outer casing. This detector has been selected as a result of a previous study that has shown a 30 mm thick, planar crystal to be the most effective size for maximising the sensitivity of the primary detector to environmental radiation [16]. Each component will be evaluated for a variety of material sizes and offsets, with the most effective design defined as having the greatest suppression ratios up to 1.5 MeV.

3. GEANT4

The simulations have been carried out using the GEANT4 Monte Carlo toolkit developed at CERN (version 9.5.p02) [10], which enables the accurate simulation of the passage of particles through matter. The definition of the experimental setup (including all shapes, objects and materials) is of particular importance for the accuracy of the simulation, as low energy photons are highly susceptible to small errors in the geometry. The GEANT4 simulations are based upon similar models detailed in reference [12], with the geometry changed to accommodate the different detector setups. These simulations were shown to be accurate to 3% for the individual detector responses, and 10% for the CSF values.

The description of the detector geometry consists of the BEGe, secondary detectors and the shielding. The BEGe contains the detector crystal, outer and inner electrodes, the aluminium canister and the carbon epoxy window. The cold finger and vacuum spaces within the detector are also included, as were the dead layers within the crystal. The secondary detectors were recreated with a crystal, the detector casings and Aluminium cylinders (to approximate the PMT’s). This approximation has been shown to provide accurate results [17], and will not affect the vetoing ability of the detectors. To complete the model, the tin and copper lined lead shielding and source housings were also recreated. NIST compounds were used where available, and all components were reproduced as accurately as possible according to manufacturer specifications.

The simulations are based upon a collection of many ‘events’. During each ‘event’, the probability of the primary detector ‘seeing’ multiple decays (within a typical charge collection time) is calculated, and one or more radioactive nuclei are then disintegrated according to known branching ratios [18]. Each event may also result in multiple photons due to nuclei emitting photons in cascade, and secondary radiation such as fluorescence, Auger electrons and photon induced x-ray emission. Radiation transport during an ‘event’ is split into a number of steps, with the length of each step within the simulated geometry determined automatically according to the physics processes involved. The energy deposited in sensitive volumes (the detector crystals) and the volume it is deposited in is recorded at each step during an event, and written to a file. Due to this setup, the energy deposited in all detectors for each event can be determined, and appropriately binned to create realistic spectra. If energy is deposited in multiple sensitive volumes during a single event, these are treated as coincident (radiation transport through the geometry happens on a much faster timescale than charge collection in the crystals). The recording of energy deposition at ‘step’ level also allows the order of the interaction process to be determined.

4. Results and discussion

For the majority of the simulations, multiple sources were considered. These include $^{241}$Am, $^{137}$Cs, $^{55}$Mn, and $^{60}$Co. All were simulated in the geometry of a compressed air filter, as this represents the largest, most dispersed source that would be routinely used. Due to the finite size of the crystal, geometrically large sources will increase the amount of Compton Scattering seen.

4.1. A typical event

Simulations were performed for a range of energies from 20 keV to 1.5 MeV. These fired photons directly at the primary detector crystal from a coordinate 10 mm above the detector. Each
A series of plots showing the distribution of photons that interact with the primary detector at a variety of energies. Low-energy photons are mostly backscattered, while higher energy photons tend to either penetrate the detector crystal completely, or interact and scatter in a forward direction. For the final (2 MeV plot) the current Compton Suppression system (including the annulus and plug) is overlaid for reference. The x and y coordinates represent the physical dimensions of the detector and resulting photon distribution. For a full colour image see the online version of this paper.
event was tracked to build up a map of where the photons typically scatter at each energy. The results of these are shown below, in Fig. 5.

In Fig. 5, the plots show two main interaction points for γ's emitted towards the crystal. These are the crystal itself, and the copper cold finger below the crystal (especially for high-energy photons). The final plot (with the current system overlaid) shows that there is significant amount of scattered events missed by the current system. While some of these scattered photons will be due to events that interact with (and scatter from) the cold finger, there is clearly a benefit to placing additional vetoing material in the area below the main detector.

4.2. Current performance

The current system uses multiple large volume NaI(Tl) crystals to veto coincident events. Due to the geometry of the system, it also heavily suppresses radiation emitted from the source in cascade. This is desirable in some situations, however it does reduce the statistics available for identifying radionuclides. The primary detector is also smaller than the BEGe 5030, however this has been replaced in these simulations to make the results comparable.

Simulations were run with the full set of sources listed above, and the suppression factors achieved are detailed below in Table 1, along with the peak efficiency in the unsuppressed and suppressed modes at each point. As these simulations reproduce the radioactive decays of each nuclide using the GEANT4 decay libraries, cascade summing is also included in the efficiency values calculated. Suppression factors were also calculated for a range of incident γ energies, and are detailed in Fig. 6. An example spectra for both the unsuppressed and suppressed modes of operation is shown in Fig. 7.

It is clear from Table 1 that Compton Suppression is only beneficial for certain radionuclides. Any that decay via a cascade of γ emission (134Cs, 54Mn) are suppressed as multiple coincident γ emissions cannot be discriminated from a Compton Scattered event. Detector dead layers, crystal holders, casings and endcaps all attenuate events that do escape, limiting the amount of suppression achievable.

The large peak CSF values calculated for low energy emissions (Fig. 6) are due to the relative reduction of the Compton edges for each energy of incident photon. γ emissions that are particularly low in energy (for example 241Am) are unlikely to scatter out of the crystal, and the small amount of radiation that does is extremely limited in geometry. As low energy emissions do not penetrate far into the crystal, the vast majority of scattered events escape backwards, away from the primary crystal and system dead layers. This allows extremely effective suppression of these photons with a detector above the primary crystal.

The values in Table 1 and Fig. 6 represent a baseline for the performance of the proposed Compton Suppression systems. Note that these simulated values represent the peak performance of the Compton Suppression system, as additional dead time and acquisition electronics effects [19,12] can reduce the performance seen in the laboratory.

4.3. Proposed configuration

The most effective configuration for Compton Suppression is an all enclosing geometry, where scattered photons (at all angles) are intercepted by the veto detector. As the system will be primarily tasked with environmental studies (where sources have low activities) there is no detrimental effect to surrounding the source with secondary crystals (as in the current design). If the secondary crystal were modular in design, then parts of the veto system (for example those crystals surrounding the source), could be turned off to minimise the veto of nuclei that decay via a cascade of γ radiation.

The proposed system will use the minimum amount of material to veto the maximum amount of events. As the height of the cylindrical crystal is increased, there will be diminishing returns as the cross-sectional area decreases for each increment in height. It is therefore desirable to minimise the height of the main secondary crystal, and then include lid and base detectors of appropriate thicknesses.

4.3.1. Main body

The chamber required for sources is 100 mm high, and the width of the detector (110 mm) across. The top of the secondary crystal will therefore be set to this height, and the base of the crystal in line with the base of the detector casing (making the total length ~240 mm). The thickness of the BGO cylinder can then be varied to establish the optimum amount of material. The thicknesses evaluated range from 30 mm to 70 mm, as 30 mm gives the minimum level of performance needed (from Fig. 4), and 70 mm is the maximum possible size within the shielding. These results are presented in Fig. 8.

Increasing the thickness of the BGO veto detector improves the CSF values as expected. The additional material particularly improves low to medium energy suppression, as scattered events that deposit a small amount of energy in the HPGe detector typically scatter with a medium to high energy remaining. The energy range from 10–1000 keV, which is the major beneficiary of an increased cylindrical size, is critical for observing many important radionuclides. Typical spectra for a 54Co source in a system with a variety of cylinder thicknesses are shown in Fig. 9.

The benefit of the main secondary crystal is realised across the entire energy range, as this intercepts the majority of scattered events from the HPGe primary crystal. Due to the cross-sectional area it subtends for scattered events from the primary crystal, this should always be the first component of a Compton Suppression shield to be implemented. While the optimum thickness is around 50–60 mm, improvements in the suppression ratios are seen up until the full 70 mm thickness for the energies simulated.

4.3.2. Lid

To veto γ’s that have scattered from the primary detector back towards the source, a ‘lid’ can be placed on the system. In the simulations of this configuration, the ‘lid’, main secondary crystal body, and the primary detector are all present in the geometry, and the performance of the system with a variety of ‘lid’ thicknesses is compared to the performance of a 70 mm thick annulus (with no lid present). This allows a realistic estimation of the suppression ability of

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Energy (keV)</th>
<th>Unsuppressed efficiency (%)</th>
<th>Suppressed efficiency (%)</th>
<th>Peak CSF</th>
<th>Total CSF</th>
</tr>
</thead>
<tbody>
<tr>
<td>241Am</td>
<td>59.54</td>
<td>24.23</td>
<td>24.23</td>
<td>1.00</td>
<td>1.00</td>
</tr>
<tr>
<td>134Cs</td>
<td>569.26</td>
<td>2.80</td>
<td>0.08</td>
<td>0.62</td>
<td>0.34</td>
</tr>
<tr>
<td>134Cs</td>
<td>684.71</td>
<td>3.33</td>
<td>0.46</td>
<td>2.77</td>
<td>1.53</td>
</tr>
<tr>
<td>134Cs</td>
<td>795.91</td>
<td>2.34</td>
<td>0.29</td>
<td>1.62</td>
<td>1.43</td>
</tr>
<tr>
<td>137Cs</td>
<td>661.66</td>
<td>3.60</td>
<td>3.60</td>
<td>4.28</td>
<td>1.61</td>
</tr>
<tr>
<td>54Mn</td>
<td>834.83</td>
<td>3.22</td>
<td>3.22</td>
<td>4.00</td>
<td>1.82</td>
</tr>
<tr>
<td>60Co</td>
<td>1173.24</td>
<td>2.10</td>
<td>0.54</td>
<td>2.84</td>
<td>1.85</td>
</tr>
<tr>
<td>60Co</td>
<td>1332.50</td>
<td>1.80</td>
<td>0.41</td>
<td>2.99</td>
<td>1.69</td>
</tr>
</tbody>
</table>
Fig. 6. The Peak and Total Compton Suppression Factors calculated for the current Compton Suppression system. These represent the system response for singular \( \gamma \) emissions, and therefore the theoretical potential of the system at each energy in the case where no accidental or cascade coincidences are emitted from the source. For a full colour image see the online version of this paper.

Fig. 7. A simulated spectra for a \(^{60}\text{Co}\) source in the compressed air filter geometry, showing both unsuppressed and suppressed modes of operation for the current \( \text{NaI(Tl)} \) based Compton Suppression system. For a full colour image see the online version of this paper.

Fig. 8. A comparison between the peak and total Compton Suppression Factors for varying thicknesses of a BGO secondary crystal. For a full colour image see the online version of this paper.
the ‘lid’ to be determined, while retaining any effects that the main veto detector will have. The ‘lid’ was placed 100 mm above the primary crystal, with a diameter equal to the outer edge of the main annulus. Compton Suppression Factors for this setup are shown in Fig. 10, and example spectra in Fig. 11.

The lid thickness was varied from 10 to 50 mm, with the majority of the additional suppression achieved seen at the Compton edges, (where photons deposit the maximum amount of energy when scattering, and are therefore likely to be scattered through ~180° towards the ‘lid’). While this equates to a small improvement in the peak and total Compton Suppression Factors, it is a substantial improvement in a specific part of the spectrum. Improved suppression was seen for all ‘lid’ thicknesses, however it was very slight above 30 mm.

4.3.3. Base
High energy γ radiation is more likely to be forward scattered, and therefore additional suppression may be achieved with a ‘base’ detector situated below the primary detector. As with the ‘lid’ simulations, the main crystal was included in the geometry, and used in conjunction with the base detector. Results from these simulations are shown in Figs. 12 and 13.

Similar levels of suppression are achieved with a ‘base’ detector, however it is concentrated in the lower energy region of the spectrum. This is a result of high energy photons depositing a small amount of energy in the primary crystal, and then interacting with the ‘base’ detector after scattering forward, beyond the primary crystal. The ‘peak’ at ~250 keV is a result of photons that backscatter out of materials surrounding the detector, including the aluminium canister and copper crystal holder/cold finger. This can only be minimised by reducing the amount of dead material surrounding the detector. An effective thickness for the ‘base’ detector was found to be 40 mm, with little additional suppression achieved above this (for a 60Co source).

4.3.4. Combination
So far, effective sizes have been determined for the main body (50 mm), the ‘lid’ (30 mm), and the ‘base’ (40 mm) secondary
Fig. 11. A comparison between the suppressed spectra when simulating a $^{60}$Co source with no ‘lid’, and a ‘lid’ between 10 and 50 mm thick. For a full colour image see the online version of this paper.

Fig. 12. A comparison between the peak and total Compton Suppression Factors at a variety of energies and ‘base’ thicknesses. The thickness of the ‘base’ detector was varied between 20 and 60 mm for these runs. For a full colour image see the online version of this paper.

Fig. 13. A comparison between the suppressed spectra when simulating a $^{60}$Co source. Most of the additional suppression achieved with the base detector is at low energies. For a full colour image see the online version of this paper.
detectors, which can now be utilised to evaluate the performance of a combined system (the envisaged design is shown in Fig. 14). The suppression factors achievable are detailed in Fig. 15, with example suppressed spectra in Fig. 16. These are also compared to the current Compton Suppression system, the proposed system with only the main body, a system with the maximum current sizes available (70 mm cylinder, 50 mm lid, 40 mm base) and an oversize system (100 mm cylinder, 60 mm lid, 50 mm base) that would require a new lead cave.

The main cylinder significantly lowers the continuum seen in the primary detector to a value less than that seen in the current NaI(Tl) based system, however it does not perform as well at the Compton edges as it cannot intercept photons that are back-scattered. The ‘lid’ and ‘base’ detectors, however, significantly improve the coverage of the veto detector. This is most obvious at the Compton edges, however additional performance is seen throughout the energy range. While the current system achieves continuum reductions of a factor of \( \sim 30 \)–\( 40 \), a BGO based system (with a 70 mm cylinder, 50 mm lid, and 40 mm base) can reduce the continuum by up to a factor of 240, with additional material in the three components of the secondary detector improving performance further.

The fundamental limit to the suppression ratios achievable arises due to dead layers in the system (crystal dead layers, detector casings, crystal holders etc.), which cannot (currently) be avoided. These layers are detrimental in a variety of ways. Primarily, dead layers reduce the chance of a photon that is Compton scattered from the HPGe detector interacting with the shield detector (it is worth noting that the reverse process, which contributes a significant fraction of coincident events to the total seen [20], is also reduced in efficiency). Source photons interacting directly with the dead material (especially the crystal holder, cold finger, and shield liners) may also scatter into the primary detector, and therefore contribute to the continuum (an obvious manifestation of this is the backscatter peak at \( \sim 250 \) keV).

The Compton Suppression factors achieved with realistic sources (and for each permutation of system design) are detailed in Table 2. Large improvements over the current design are clearly possible, even for nuclei that decay via a cascade of \( \gamma \) emission. If data is collected in both suppressed and unsuppressed modes, then emissions that are detrimentally affected by the suppression system can be analysed from the unsuppressed data, resulting in no loss of information.

5. Conclusion

GEANT4 Monte Carlo simulations have been utilised to develop an optimised, next generation design for a Compton suppression system. A realistic model, based upon an existing system and validated with a variety of detectors, has been developed. This was
then used to identify the optimum size and material of a veto detector.

Several possible materials were investigated, with BGO identified as the most effective material for a suppression shield due to its excellent interaction efficiency. A routine that tracked where photons scatter in the simulation has also been developed, and used to identify key areas where suppression may be achieved/improved, such as the area below the primary detector where high energy γ's tend to scatter.

Simulations were then utilised to estimate the performance of a BGO shield, and optimise the thicknesses and sizes of each shield component. Compton Suppression Factors were improved by a factor of two over the current Compton suppression system, and the continuum reduction for common sources (such as 60Co) is improved by up to an order of magnitude. This equates to a reduction in the continuum of up to 240 times, which is critical for low-energy, low-activity radionuclides that are often swamped by the large continua from higher energy emissions.

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References


Coincidence corrections for a multi-detector gamma spectrometer

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Accidental coincidence

A B S T R A C T

List-mode data acquisition has been utilised in conjunction with a high-efficiency γ-γ coincidence system, allowing both the energetic and temporal information to be retained for each recorded event. Collected data is re-processed multiple times to extract any coincidence information from the γ-spectroscopy system, correct for the time-walk of low-energy events, and remove accidental coincidences from the projected coincidence spectra. The time-walk correction has resulted in a reduction in the width of the coincidence delay gate of 18.4 ± 0.4%, and thus an equivalent removal of ‘background’ coincidences. The correction factors applied to ~5.6% of events up to ~500 keV for a combined 137Cs and 60Co source, and are crucial for accurate coincidence measurements of low-energy events that may otherwise be missed by a standard delay gate. By extracting both the delay gate and a representative ‘background’ region for the coincidences, a coincidence background subtracted spectrum is projected from the coincidence matrix, which effectively removes ~100% of the accidental coincidences (up to 16.6 ± 0.7% of the total coincidence events seen during this work). This accidental-coincidence removal is crucial for accurate characterisation of the events seen in coincidence systems, as without this correction false coincidence signatures may be incorrectly interpreted.

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1. Introduction

Gamma spectroscopy detectors are often employed in singular form, however an increasing number of systems uses multiple detectors, synchronised such that events can be classified temporally as well as energetically. This includes γ-γ systems, which can detect coincident radiation with a granularity that is not possible with a single detector, and use the coincidence information to extract rare events that would normally remain ‘buried’ in the data. The applications for this range from a simple reduction of summing effects in high efficiency systems, to analysis of the time delays between coincident radiation to reveal fundamental nuclear properties [1,2].

Further uses of coincidence systems are Compton and cosmic suppression, where additional detectors are used in a similar fashion to a γ-γ system, however the coincidence information is used to veto (remove) events. Compton suppression systems assume that radiation detected in coincidence is the result of a photon partially depositing its energy in the main crystal, before scattering into the secondary detector [3]. These events only contribute to the Compton continuum (which is essentially noise) in the primary detector, and thus the removal of these events can substantially improve the sensitivity of the detector system [4]. Cosmic suppression works in a similar manner, however the additional detectors are used to veto high-energy cosmic radiation (such as muons) that originate in the upper atmosphere. This radiation again adds noise to a detection system, which may obscure signals from radioisotopes of interest [5,6].

To extract coincident events, complex electronics are often used to identify coincidences ‘online’ (during data acquisition), and record the data. It is increasingly common, however, to use a system that will timestamp all data, and store this for ‘offline’ analysis [7–9]. This is known as ‘list-mode’ acquisition, and has several advantages over the previous method which discards temporal information once it is evaluated and therefore cannot be re-processed to extract further coincidence information.

Coincidences are identified by setting a coincidence window (usually within a few μs of an event registering in a detector) where a coincidence is recorded if signals are seen in any additional detectors. Properly configured (whether in hardware or software) this will contain the coincident events of interest. However it will also contain some random, or false coincidences that are due to the rate of events seen by the detectors. The exact timing of the coincidences seen also has an energy dependence; lower-energy photons will register later in the coincidence window than higher-energy events, and this effect is known as time-walk [10]. This report considers a list-mode acquisition system.
exploiting the ability to re-process raw data to both correct for the time-walk in the detector and substantially reduce the level of accidental coincidences seen.

2. Experimental setup

A $\gamma-\gamma$ coincidence system was utilised for this work, as this method of detection focuses on coincidences (as opposed to anti-coincidences, which are generally far more numerous). A $\gamma-\gamma$ system will therefore be particularly sensitive to the removal of the coincidence background, and the best candidate to demonstrate a substantial improvement in the resulting spectra.

The $\gamma-\gamma$ system consists of two large-area, planar HPGe detectors (model BE6530, from Canberra UK in Harwell, Oxfordshire) situated in a back-to-back configuration (both facing towards a central source). These are then enclosed within a Pb cave to minimise the effects of terrestrial radiation. Each detector signal is passed through a pre-amplifier (model 2005), and to a LYNX™ Digital Signal Processor, which controls all further amplification, pole-zero correction and digitisation of the pulse. A synchronisation cable runs between the LYNX™ units to allow synchronisation of the clocks used to record the events in each detector, and data is collected in list-mode. A schematic of the system is shown below, in Fig. 1.

3. Measurement and analysis

All data was analysed using the post-processor detailed in reference [8]. Calibration sources were chosen to cover the 10–2000 keV energy range to fully characterise each system. Several single $\gamma$ emitters were used, as well as a NIST (National Institute of Standards and Technology) traceable complex $\gamma$ source. The isotopes that comprised these were $^{241}$Am (59.54 keV), $^{109}$Cd (88.03 keV), $^{57}$Co (122.06 keV), $^{139}$Ce (165.86 keV), $^{113}$Sn (391.68 keV), $^{137}$Cs (661.67 keV), $^{54}$Mn (834.84 keV), $^{88}$Y (898.04 and 1836.06 keV), $^{60}$Zn (1115.54 keV), and $^{60}$Co (1173.23 and 1332.49 keV).

Analysis was completed using the post-processor to sort the data, and ROOT [11] for matrix manipulation and peak searching/fitting. Coincidences were identified by searching for all events in the secondary detector with a time delay ($\Delta t$) between 10 $\mu$s and +10 $\mu$s (with $t=0$ defined as the interaction time in the primary detector). These coincidences were then recorded into a 3D matrix with the energy deposited in the primary detector, the energy deposited in the secondary detector, and the time delay on each axis ($E_1, E_2, \Delta t$). This allows a delay spectrum to be created, with peaks identifying characteristic delays where there are a large number of coincident events. As they are in a matrix, time and energy gates can also be set to extract information from the dataset. A typical delay spectrum is shown in Fig. 2.

4. Correcting the time-walk of coincident events

The processing of coincidence signals has a small energy dependence, with low-energy events taking longer to process. This widens the coincidence peak seen in Fig. 2, causing the system to potentially miss coincident events, or include unnecessary ‘background’ coincident events. To correct for this effect, a source with simultaneous $\gamma$ emission is required (any delay in the emission of multiple $\gamma$ decays will artificially affect the position of the coincidence peak), with relatively high-energy decays (to allow characterisation of the time-walk over a large energy range). Both of these requirements are met by $^{60}$Co, which emits two $\gamma$’s (1173.23 and 1332.49 keV) within a picosecond. The time resolution of the LYNX™ units is limited to 0.1 $\mu$s, and a typical coincidence window may span up to ~3 $\mu$s; by comparison the lifetime of $^{60}$Co is negligible.

With a $^{60}$Co source in place, data was acquired until the statistical error in each bin was reduced to less than 1%. Once the coincidences are extracted into an ($E_1, E_2, \Delta t$) matrix, an energy gate is placed around the 1332.49 keV decay in the primary detector (set to two times the full width at half maximum). The coincidence spectra seen in the secondary detector can then be plotted against the time delay to produce Fig. 3.

To correct the time-walk at lower energies, the mean $\Delta t$ is taken in each energy bin, and the resulting plot fitted with a polynomial function. The parameters for these are extracted and the corrections reapplied to the data on an event-by-event basis. The uncorrected and corrected projections of the gated matrix are shown in Figs. 4 and 5, respectively, with the resulting delay spectrum in Fig. 6. A substantial improvement is seen in both the width and the shape of the delay window, allowing a far more effective time gate to be applied when extracting coincidences.
Fig. 2. An example coincidence delay spectrum from the $\gamma - \gamma$ system. Three distinct regions are identifiable: the main coincidence peak at $\sim 0$ $\mu$s, the raised background either side of the peak, and the flat background from $\pm 6$ $\mu$s onwards. The flat background is due to random coincidences between the two detectors, and therefore roughly constant. The raised background is due to time-walk of low-energy events, and therefore dependent on the energy deposited in each detector. Note that this manifests itself either side of the main peak due to the symmetry of the system.

Fig. 3. The $(E_\gamma, \Delta t)$ plot extracted from the matrix when gating on a 1332.49 keV $\gamma$ decay in detector 1. As well as seeing the 1173.23 keV decay in coincidence, energy depositions from 1173.23 keV $\gamma$’s that Compton scatter out of the secondary detector are also seen. At low-energies, there is a trend where $\Delta t$ between coincident events rises, which is the result of time-walk.

Fig. 4. The projection of Fig. 3 fitted with a polynomial function. Note that the points are plotted as crosses, with the height proportional to the error in the mean. Above 1173.23 keV, the errors rapidly increase due to the reduced statistics for coincidences in this region.
This also minimises statistical uncertainty when attempting to remove the ‘coincidence background’.

For a combined $^{137}$Cs and $^{60}$Co high-activity ($>100$ kBq) source, the mean reduction in gate width (and therefore background from accidental coincidences) after the time-walk correction was $18.4 \pm 0.4\%$, with $\sim 5.6\%$ of events being corrected. The time correction was found to be substantial up to $\sim 100$ keV, however non-zero correction factors were present up to $\sim 500$ keV. It was not expected that the time-walk would have an effect at such high energies, however the time resolution of the LYNXTM acquisition module (100 ns) limits further inspection of the coincidence window structure. The improvements in the delay gate allow cleaner coincidence spectra to be created, as reducing the width of the gate directly excludes ‘background’ coincidence events.

5. Characterising and removing the ‘coincidence background’

To extract coincidences from the delay spectrum, a region is selected that covers the peak whilst minimising the amount of random coincidences (see Fig. 7, pink area). The raised region of the coincidence background includes events that should be in the main peak, but are shifted away from this due to the time-walk of low energy signals. This cannot therefore be used as a representative background region, however the flat region in the time delay spectrum can be used (Fig. 7, light blue area). Events are extracted from this area and normalised to the width of the delay window. These events can then be subtracted from those extracted in the main coincidence gate. The blue hatched area represents the resulting coincidences once the coincidence background is subtracted from the main peak.

For the combined $^{137}$Cs and $^{60}$Co high-activity source, coincidences from the delay gate and background areas were extracted from the matrix, and used to create both a ‘normal’ and a ‘background subtracted’ coincidence gate projection. Note that no energy gates were used for this, and so the spectra contain all the coincidences seen in a detector when observing an event in the other. The resulting spectra are plotted in Figs. 8 and 9, with Fig. 8 showing the complete spectra, and Fig. 9 showing a series of zoomed spectra to highlight the differences between the two methods.

This method accounts for $\sim 100\%$ of the accidental coincidences, however it only works if the background rate is high enough, and therefore representative of what is intermixed with the original delay gate. For the $^{137}$Cs and $^{60}$Co source, $16.6 \pm 0.7\%$ of events were removed from the coincidence window, three times more than during

Fig. 5. The resulting projection of Fig. 3 once corrected for low-energy time-walk. This used the parameters from the functions fitted in Fig. 4 to construct the energy dependant time correction, before reprocessing the data and applying these on an event-by-event basis.

Fig. 6. The delay spectrum for list-mode data both before and after the time-walk correction. There is a substantial improvement in both the width and the shape of the coincidence peak.
Fig. 7. A coincidence delay spectrum from the $\gamma-\gamma$ system. The extracted coincidences are in the pink region, the coincidence background in the light blue region, and a representation of the resulting region (once the subtraction is made) within the blue hatched area. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper.)

Fig. 8. The total and coincidence spectra for the $\gamma-\gamma$ system using the combined $^{137}$Cs and $^{60}$Co source. The major features in the coincidence spectra are the backscatter peak ($\sim 250$ keV), Compton edges ($\sim 500, 950, 1150,$ and $2300$ keV), and the two major $\gamma$ decays in $^{60}$Co ($1173.23$ and $1332.49$ keV), which would all be expected in the coincidence spectra. Features that should not be present in the coincidence spectra are the $661.66$ keV $^{137}$Cs peak, and $2505.72$ keV $^{60}$Co sum peak.

Fig. 9. A series of zoomed spectra for the three regions that show major differences between the standard gate and the background-subtracted gate. The left plot focuses on the low-energy region ($^{137}$Cs X-rays), the middle plot on the $661.66$ keV decay from $^{137}$Cs, and the right on the $2505.72$ keV sum peak from $^{60}$Co.
the time-walk correction. The spectra show excellent suppression of false coincidences, which are most apparent as a 661.66 keV $^{137}$Cs peak (this is a singular $\gamma$ emission and should therefore have no coincidence signature). Additional signatures were also removed or reduced; the $^{60}$Co sum peak at 2505.72 keV does not contribute to the corrected spectra (by definition, if both $^{60}$Co emissions are seen in a single crystal, no other true coincident events would be expected), and a substantial amount of Cs X-rays were removed. As X-rays can be caused by impinging $\gamma$ radiation from a variety of energies and sources, there will always be some additional number of X-rays that create a valid coincidence signature, and should not be excluded from the coincidence spectra. No statistical difference could be found in the $^{60}$Co peaks or the additional features (such as the Compton edges), which confirms that the coincidence background subtraction is both valid and effective. Particular care has to be taken when selecting the regions for both the total and background subtraction gates; inhomogeneities throughout the delay window (due to statistical fluctuations/time-walk/delayed coincidences) could cause the subtraction to be over-estimated or under-estimated. The background coincidences also show a similar energy profile to events in each detector, such that higher-energy events contribute to a smaller proportion of the spectra than lower-energy coincidences. This can reduce the statistics available in the higher energy region, causing greater uncertainty when applying the background subtraction.

6. Conclusions

List-mode data acquisition has been utilised in conjunction with a high-efficiency $\gamma-\gamma$ coincidence system, allowing data collection whereby both the energetic and temporal information are retained for each recorded event. As this information is not lost during acquisition, the data can be re-processed multiple times to extract the coincidence information, correct for time-walk of low-energy events, and remove accidental coincidences from the projected coincidence spectra.

The time-walk correction has resulted in a reduction in the width of the coincidence delay gate of 18.4 ± 0.4%, and thus an equivalent removal of accidental coincidences. The correction factors applied to ~5.6% of events up to ~500 keV; however the authors would like to note that while these corrections improved both the shape and the width of the peak, further investigation was limited by the time resolution of the electronics. Future work is planned to investigate the delay gate using improved electronics that can resolve timestamps at the 10 ns level, as opposed to the 100 ns resolution possible in this work. As well as the aforementioned benefits arising from the use of an optimised decay window, the time-stamp correction is crucial for sources with low-energy decays (which may be missed by an uncorrected delay gate), and the collection of data that relies on accurate timing measurements.

By extracting both the delay gate and a representative ‘background’ region for the coincidences, a coincidence background-subtracted spectra can be projected from the coincidence matrix, which effectively accounts for ~100% of the accidental coincidences (these accounted for up to 16.6 ± 0.7% of the events seen during this work). It is important to note that this method only estimates the coincidences present in the spectrum (in contrast to the time-walk correction, which allows the elimination of events directly). This technique is obviously limited to radiation sources that contribute to a constant coincidence background, and sources where a coincidence background region can be extracted that is representative of this. Such sources would include coincidences arising from terrestrial and cosmic background radiation, and radioactive sources in which the half-lives are much greater than the length of the decay window. This correction is essential for accurate characterisation of the events seen in coincidence systems, as otherwise false coincidence signatures may be incorrectly interpreted.

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References

Next Generation Detection Systems for Radioactive Material Analysis

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Compton Suppression techniques have been widely used to reduce the Minimum Detectable Activity of various radionuclides when performing gamma spectroscopy of environmental samples. This is achieved by utilising multiple detectors to reduce the contribution of photons that Compton Scatter out the detector crystal, only partially depositing their energy. Photons that are Compton Scattered out of the primary detector are captured by a surrounding detector, and the corresponding events vetoed from the final dataset using coincidence based fast-timing electronics. The current work presents the use of a LynxTM data acquisition module from Canberra Industries (USA) to collect data in ‘List-Mode’, where each event is time stamped for offline analysis. A post-processor developed to analyse such datasets allows the optimisation of the coincidence delay, and then identifies and suppresses events within this time window. This is the same process used in conventional systems with fast-timing electronics, however, in the work presented, data can be re-analysed using multiple time and energy windows. All data is also preserved and recorded (in traditional systems, coincident events are lost as they are vetoed in real time), and the results are achieved with a greatly simplified experimental setup. Monte-Carlo simulations of Compton Suppression systems have been completed to support the optimisation work, and are also presented here.

I. INTRODUCTION

Current analyses of environmental samples need up to 7 days of counting to achieve the Minimum Detectable Activity (MDA) required for many radionuclides. This project seeks to develop a radiation detection system which will substantially reduce this counting time by increasing the sensitivity of the detector systems used.

The standard tool for the measurement of gamma-rays is the hyper-pure germanium (HPGe) detector (see Fig. 1), which allows discrete, characteristic gamma-ray lines to be identified. The low-energy end of such spectra is extremely important in the detection of gamma-rays emitted from actinide nuclei (such as 239Pu), however this low energy region is complicated by the Compton continuum arising from the higher energy decays in the same sample. The situation is further complicated by the fact that the emission probability for these gamma-rays is challengingly small (with typically only 1-2 gamma-rays emitted per 10,000 239Pu decays). As such, the low-energy gamma emissions from 239Pu can be swamped by the Compton continuum from other radionuclides present in environmental samples.

In the Compton scattering process only a portion of the full gamma-ray energy is deposited in the HPGe detector, and the rest can escape to the immediate surroundings of the detector (resulting in an unwanted background above which the discrete peaks of interest sit). If the Compton scattered photon is measured in a second (guard) detector surrounding the germanium detector, the simultaneous events can be rejected, resulting in a significant suppression of the Compton background [1] (see Fig. 2).

To operate multiple detectors simultaneously, fast-timing electronics are needed which can discriminate coincident events in real time. These are often expensive and require substantial setup time to optimise the coi-
FIG. 2. Compton scattering occurs when the photon interacts with a “free” atomic electron. The resulting photon may escape the detector, resulting in a partial energy deposition (and the Compton continuum). The resulting continuum can be reduced by “catching” the escaped photons, and vetoing these events from the primary detector. The time profile of the coincidences is shown as an inset, with clear coincidences at -5 and 2 μs. The coincident events in the -5 μs peak are shown in blue, and coincident events in the 2 μs peak in green. By suppressing both of these coincidence windows, the red spectra is obtained, which has a greatly reduced background when compared to the black (original) spectra.

II. LIST-MODE ACQUISITION

An alternative method to veto coincident events has been tested, which involves using two Lynx\textsuperscript{TMD} data acquisition modules from Canberra Industries (USA). Customised \texttt{c++} software is used to synchronise the two units, and then collect data for the HPGe and guard detectors respectively. The output is a text file that lists each interaction and a timestamp (with \(\sim\)100 nanosecond resolution) for each detector [2].

To analyse the output, software [3] has been developed using \texttt{c++} and ROOT, which reproduces the ability to veto events based upon their timing, however, with much more flexibility. The software can create 3D coincidence/anticoincidence matrices (typically with detector energy on 2 axes and the time delay on the third). As none of the data has been discarded, multiple time and energy gates can then be set to analyse the data in a variety of ways. The electronics setup is also greatly simplified, however, additional time is required to process large data sets due to the large files produced.

III. MONTE-CARLO OPTIMISATIONS

For the Monte-Carlo toolkit, GEANT4 was selected as it is an open-source, community developed software with a large user base. Preliminary work has focused on the development and validation of several GEANT4 models for a variety of sources, detectors and geometries. These have included both scintillation based detectors (NaI(Tl)), and semiconductor detectors (HPGe). Both models were able to accurately reproduce the detector response (see Fig. 3), and so allow the simulations to progress to more complex systems. These simulations have also allowed the calculation of efficiencies for complex sources, and cascade correction factors for these detectors.

The current work involves the development of a GEANT4 model that accurately simulates the performance of a Low-Energy Germanium (LEGe) detector with an active NaI(Tl) shield (see Fig. 4). This will be validated against an existing system, and then used to aid in the development of a new detector design.

IV. RESULTS

The software written to process the List-mode output speeds up the analysis process by an order of magnitude, and is scalable to large datasets (currently tested up to 60 GB on a Pentium D 2.8 GHz system with 2 GB of RAM).

List-mode analysis allows much greater detail to be extracted when considering coincident events, as no data is discarded during acquisition. Processing the data into 3D matrices allows the user to gate on single or multiple time and energy windows to efficiently extract information (see Fig. 2).

GEANT4 simulations have allowed the characterisation of scintillation and semiconductor based detectors,
creating reliable models from which complex efficiency calculations can be performed. GEANT4 models have also allowed the calculation of cascade summing corrections for multiple detector and source geometries. Further simulations have identified the optimum parameters for shielding high resolution gamma spectroscopy systems.

V. FUTURE WORK

Once validated, the Compton Suppressed LEGe model will be extended to include a BEGe (Broad-Energy Germanium) primary detector. The analysis software will continue to be developed, allowing coincidence processing of more than two detectors. Further investigations into advanced coincidence analysis will be undertaken, including the separation of event types based upon their timing characteristics. Modelling of alternative systems and setups will also be performed, allowing a determination of the optimum design for a high-sensitivity, high-resolution, germanium detector.

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