Direct measurement of strontium 90 in groundwater: Geometry optimisation of a photodiode based detector

G. Turkington,[a,1] **K. A. A. Gamage,**[a] **J. Graham.** [b]

[a] *Electronics & Electrical Engineering, University of Glasgow, Engineering Way, Glasgow, United Kingdom*

[b] *National Nuclear Laboratory, Sellafield, Seascale, United Kingdom*

E-mail: g.turkington.1@research.gla.ac.uk

Abstract: This paper examines the feasibility of detecting strontium 90 in groundwater directly with photodiodes and considers the physical parameters which maximise radiation absorption within the detector. Geant4 simulations were used to draw comparisons between silicon, gallium arsenide and cadmium telluride detectors of varying surface area and thickness. Detectors were compared in their ability to absorb point and scattered sources of radiation. The results indicate that a detector, of 10 mm^2 surface area, and 1 mm thickness offered the highest detection efficiency in a contaminated groundwater simulation. 1 mm thick and 100 mm^2 detectors cadmium telluride and gallium arsenide detectors were modelled in a groundwater borehole scenario. Each material offered similar detection efficiency, but the greater backscattering effect in cadmium telluride resulted in a greater peak at lower energies compared to that observed in gallium arsenide.

Keywords: Detector modelling and simulations I, Solid state detectors, Interaction of radiation with matter, etector design and construction technologies and materials

[1]Corresponding author.

Contents

1 Introduction

Leaks and spills of waste at nuclear decommissioning sites have introduced radionuclides, such as ^{90}Sr, into environment resulting in contamination of the groundwater table. ^{90}Sr has a half-life of 28.8 years and decays at 0.546 MeV. Its daughter, Yttrium 90, decays at 2.278 MeV with a significantly shorter half-life of 64 hours [1]. As ^{90}Sr is found in the same column of the periodic table as calcium, it shares some chemical properties and tends to accumulate around calcium rich tissues, such as bone structures, when ingested by organisms. Due to its half life and radiotoxicity ^{90}Sr is associated with irradiation of bone marrow and leukaemia [2, 3]. As such, ^{90}Sr requires routine monitoring for many decades at some nuclear sites. Current techniques necessitate the manual sampling of groundwater from boreholes and analysis is performed in off-site laboratories. Samples are radiochemically treated and activity is measured by liquid scintillation, Cherenkov or proportional gas counting methods [4]. These processes are time consuming, produce secondary waste and are expensive. By detecting radiation directly and in situ, the need for sample collection can be eliminated which may reduce the worker dose and lifetime monitoring costs [5]. This paper proposes the optimal physical characteristics for a photodiode detector for in situ ^{90}Sr measurement in groundwater.

Photodiodes are semiconductor materials which can be used to detect radiation. Under a reverse bias voltage, electron-hole pairs are separated upon interaction with ionising radiation. The separated holes and electrons are swept up by the applied voltage and the resulting current is in proportion to the energy of the incident radiation. Photodiodes may be formed by placing a layer of undoped semiconductor between positively, p, and negatively, n, doped layers, forming a p-i-n junction, or the deposition of metal contacts onto the semiconductor material, forming an ohmic or Schottky diode. The undoped intrinsic layer is the sensitive region and responsible for charge generation as ionising radiation is absorbed within the detector. Originally developed as photon detectors, in recent years photodiodes have been used to detect ionising radiation [6, 7]. When ionising radiation separates electron-hole pairs in the active region the resulting charge must be collected by the applied reverse bias voltage. The charge collection efficiency (CCE) is an important factor in determining the energy resolution of the detector [7]. CCE is limited in semiconductor materials by defects present in the semiconductor crystal. Defects may arise as a result of fabrication

techniques and can reduce the size of the active region, trap charge, or otherwise inhibit the collection of charge in the detector. Difficulties encountered in producing high quality and defect free CdTe [8] and GaAs [9] crystals have held back their adoption as materials for radiation detectors. One of the major defects associated with GaAs detectors is known as EL2 defect which inhibits the charge transport properties of GaAs [10]. Meanwhile, CdTe suffers from a polarization effect after the detector has been operating under bias for a period of time, again degrading charge transport in the detector [7]. The presence of defects must be considered when choosing a semiconductor material and whether they can be reduced with improved fabrication techniques or otherwise.

Photodiodes are fabricated from numerous materials, each with unique properties. Silicon is the most commonly used, mature and widely available semiconductor. It has a low atomic number, 14, and a narrow bandgap, 1.12 eV and density og 2.329 gcm^{-3}. GaAs has a higher effective atomic number, 32, and a bandgap of 1.42 eV which makes it viable at room temperature [11] and a higher density, 5.32 gcm^{-3}. In recent years GaAs detectors been developed as gamma and X-ray detectors with applications in medical physics and astronomy [6, 12–15]. The intrinsic layer in these devices range from just 10 μm to 500 μm. CdTe has an even higher effective atomic number, 50, than GaAs, and an even wider bandgap, 1.5 eV, and a density of 5.85 gcm^{-3}. In principle, these properties suggest CdTe may be even more effective at absorbing ionising radiation while also capable of room temperature operation [7, 9, 16, 17].

This paper aims to design photodiodes which will be deployed into contaminated groundwater to determine the activity of ^{90}Sr directly. The detection efficiency must be maximised, defined as the ratio between the number of particles which deposit energy in the detector and are detected and the number of particles emitted [18]. The surface area, thickness and material composition of the detector are factors which will determine the detection efficiency. In this case the detector must be capable of fully absorbing the beta particles released during ^{90}Sr decay, up to 0.546 MeV, and ^{90}Y decay, up to 2.278 MeV. If the particles in question are of sufficient energy, they will simply pass right through the detector, while only depositing a fraction of their initial energy. While this can be recorded as a "hit" in the detector, and can be used to determine the activity of a source, the information will be insufficient for capturing a spectrum of emission. ^{90}Sr is the predominant source for beta radiation at Sellafield and as such its distribution across the site is akin to distribution of total beta activity [19]. However the presence of other radionuclides, such as ^{137}Cs, may obfuscate the determination of ^{90}Sr activity. By taking a gross measurement of activity in the groundwater and determining detector response to individual radionuclides linear regression may be performed to non-destructively determine the presence and relative activities of radionuclides [20].

2 Monte Carlo Simulations

Geant4 is a Monte Carlo simulation code developed to model the propogation of radiation through matter [21]. Written in C++, Geant4 code has been used to simulate radiation-matter interactions in high-energy particle physics, medical and nuclear applications. The software traces radiation, step by step, as it traverses through matter. The probability of interactions with matter are described by probability distributions, known as cross-sections. Random number generation at the beginning of each step determines the outcome of the interaction. Data about the detector's response to radiation can then be collated and processed. The cross-sections are determined by the "physics list" which

details the set of processes involved in the simulation. In this project a Geant4 model has been developed to simulate how beta radiation is absorbed in photodiodes in a groundwater borehole environment, and whether they are suitable for use as in situ beta detectors. This simulation used the FTFP BERT list which covers electron interactions up to 100 TeV, maximum step length of 0.65 mm, energy threshold of 900 keV, and a low energy limit of 1 eV.

2.1 Absorption of beta particles

The thickness of the detector is one of the factors which will determine how effectively it absorbs incident beta particles. Detectors of increasing thickness were modelled with 100 mm^2 surface areas. A beam of 1×10^7 0.546 MeV electrons was fired into the centre of the detector, from the surface, and the energy deposition of each particle was recorded. The results are displayed in Fig. 1. As anticipated, Si fully absorbed significantly fewer particles than GaAs and CdTe at lower thicknesses due to its density. The detector only begins to fully absorb a small fraction, 1.766 % ±0.004, of incident particles when the thickness is approximately 200 μm. CdTe outperformed GaAs in this range, however at 300 μm GaAs absorbed a higher percentage of particles completely. As the thickness of the detectors further increases, Si begins to absorb a greater proportion of particles and outperforms GaAs and CdTe at 800 μm. While GaAs and CdTe show diminishing returns as detector thickness is increased beyond 400 μm, Si does not exhibit the same behaviour in this range, and is capable of fully absorbing 71.696% ±0.027 of incident beta particles at 800 μm thickness. Meanwhile CdTe only fully absorbed 50.590% ±0.023of incident particles and GaAs 58.903% ±0.024.

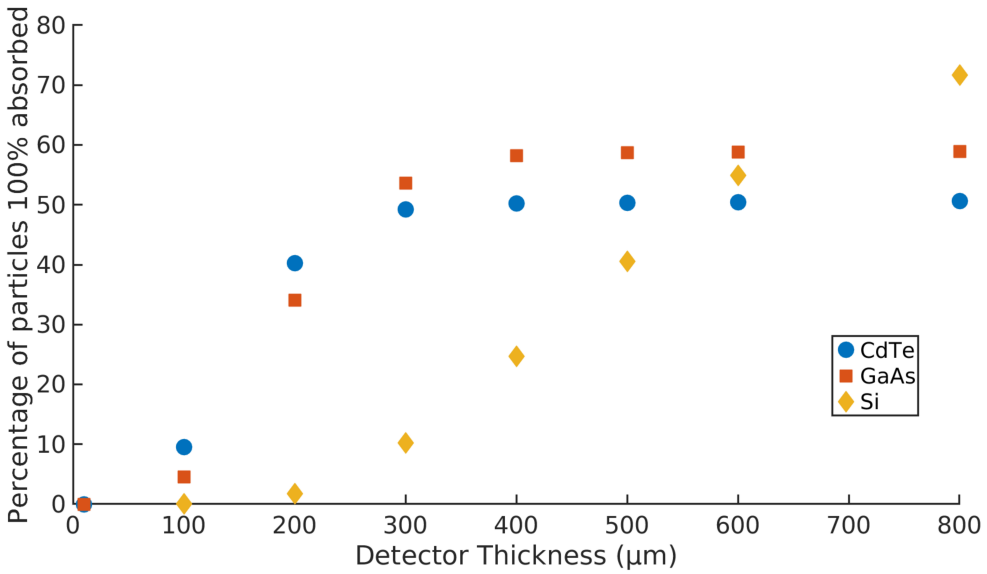


Figure 1. A comparison of complete 0.546 MeV beta particle absorption in GaAs (squares), CdTe (circles), and Si (diamonds) detectors of varying thickness. Errors are plotted, but not visible due to marker size.

In principle higher desnity materials should absorb more radiation, however the simulation results can be explained by another factor related to the atomic number. As electrons interact with a surface they are prone to a phenomenon known as backscattering [18]. Electrons undergo erratic

and sizeable deflections as they propagate through matter. Particles incident on the surface of the detector may deflect through such large angles that they simply rebound and leave the detector while only depositing a fraction of their energy. This phenomenon is more pronounced in high atomic number materials and this offers an explanation of the behaviour observed in Fig. 1. A lower proportion of particles are backscattered by silicon, but as the material is inefficient at absorbing radiation of this type, the benefit is only seen in large volumes of material. CdTe is in theory the best absorber of radiation, but in this experiment its full benefit cannot be extracted. It outperforms GaAs at low thickness, but as a higher fraction of particles are backscattered, its absorption plateaus at a lower percentage of incident particles. To minimise the effects of backscattering and compare stopping power of each material, a particle beam was generated from the centre, rather than at the surface of 800 μm thick detectors. In this simulation, GaAs fully absorbed 95.96 % ±0.03% of particles, CdTe 96.41 % ±0.03% , and Si just 36.25 % ±0.02%.

When ^{90}Sr decays via beta emission, it transmutes to ^{90}Y which itself decays by beta emission at 2.278 MeV. As a result, ^{90}Y contributes to the gross beta activity of the groundwater and therefore its presence must be quantified. Fig. 2 compares the ability of 100 mm^2 devices of varying thickness to fully absorb 1×10^7 beta particles at 2.278 MeV. While 400 μm seemed to be the point of diminishing returns for GaAs with 0.546 MeV particles, for 2.278 MeV only 0.430% ±0.002% are absorbed. CdTe, meanwhile, fully absorbs 1.480% ±0.004%. Silicon detectors are essentially ineffective for particles of this energy, absorbing 0.0008 % ±0.0002%. ^{90}Y quantification will require a detector thicker than 400 μm, with CdTe offering superior beta particle absorption than GaAs.

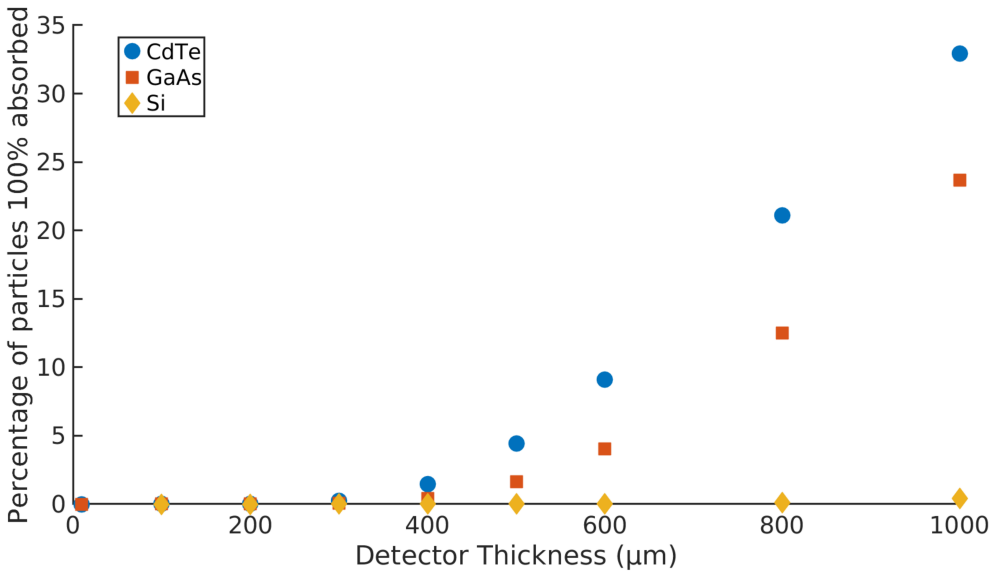


Figure 2. Fully absorbed 2.278 MeV beta particles in GaAs (squares), CdTe (circles) and Si (diamonds). Errors are plotted, but not visible due to marker size.

Beta particles take an erratic path through matter, therefore the width of the detector will influence its ability to absorb incident particles from a point source. To investigate the significance of this parameter detectors were modelled with a thickness of 400 μm and varying surface area

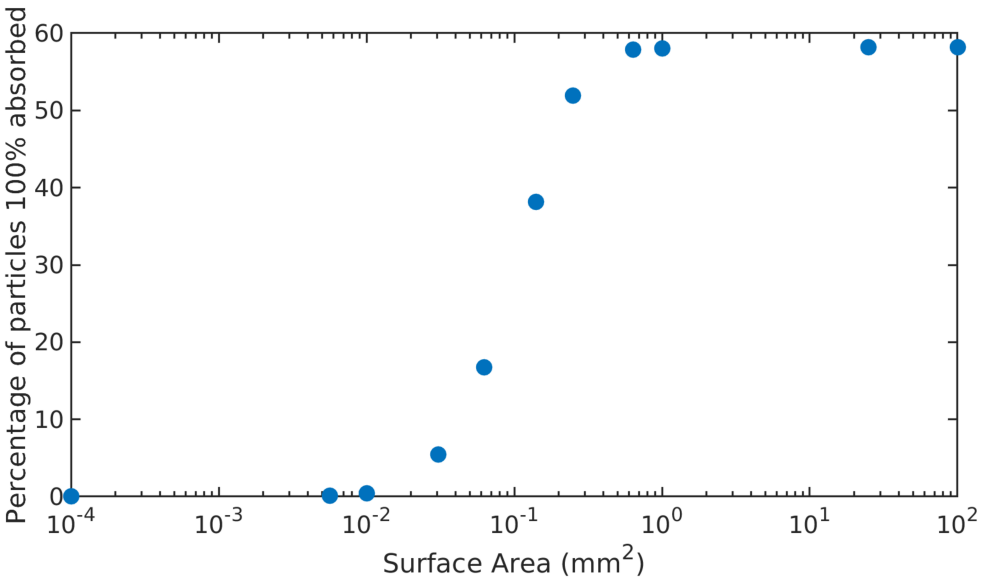


Figure 3. Complete 0.546 MeV beta particle absorption in 400 μm thick GaAs detectors of varying surface area.

from 10^{-4} mm^2 to 100 mm^2. The results of their interaction with 1×10^7 electrons at 0.546 MeV can be seen in Fig. 3. While the surface area is below 10^{-2} mm^2 the detector fails to record the full energy of incident particles at all. In the range between 10^{-2} mm^2 to 6.4 mm^2 the device becomes viable. Again, diminishing returns are seen when the surface area is further increased as the complete absorption of particles levels off out at approximately 58%.

2.2 Contaminated groundwater simulation

The simulations presented so far have examined point sources of radiation, however this research concerns the assay of contaminated groundwater. This scenario is modelled by simulating a 5 cm diameter and 2.5 cm deep cylinder of groundwater, populated with 10 $\times 10^7$ isotropic decaying ^{90}Sr ions. This simulation examined ^{90}Sr decay only. Four detectors of 400 μm thickness and varying surface area were investigated for their detection efficiency. The detectors were positioned at the surface of the water and the spectra collected after the run of particles is displayed in Fig. 4. With a 1 mm^2 surface area, the detector only managed to record 163 hits leading to an incomplete spectrum; the maximum energy observed was 0.268 MeV. The 100 mm^2 detector counted a total of 13015 particles, with the counts dropping below 5 after 0.458 MeV. The total counts observed scaled linearly with surface area. The vast majority of decay products did not reach the detector as they are either emitted in the wrong direction, or absorbed before they reach the device. This simulation highlights the importance of maximizing the number of particles striking the detector when counting from a scattered source.

To examine the sensitivity to thickness, four 100 mm^2 GaAs detectors ranging from 0.4 mm to 2 mm were compared. The detectors were positioned at the surface of the water and the spectra they collected after the run of 10^8 ^{90}Sr and ^{90}Y ions had decayed is displayed in Figure 5. In this scenario, the 0.4 mm thick detector counted 196,515 particles but only counted 115 particles in the

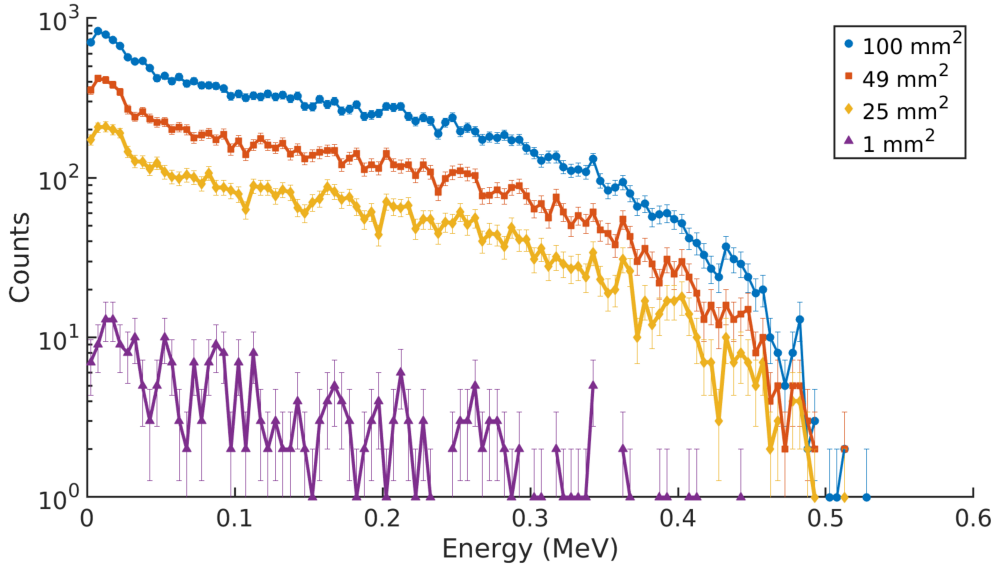


Figure 4. ^{90}Sr spectra observed in 400 μm thick GaAs detectors with increasing surface areas.

range from 1.725 MeV to 2.300 MeV, and therefore was inefficient at capturing the full spectrum of radiation. The thickest device detected 215,032 particles and counted 1,873 particles in the range from 1.725 MeV to 2.300 MeV. A peak can be seen at 0.302 MeV for the 0.4 mm thick device, likely a result of more energetic particles only depositing a fraction of their energy in the material. This peak is not observed in thicker detectors, but each spectrum features a peak approximately at 0.023 MeV as a result of backscattering. At greater thicknesses the spectra are distinguished by their absorption of higher energy particles, from 1.150 to 2.3 MeV. As the thickness is increased from 0.75 mm to 1 mm, a 20.58 % increase in counts is observed, but as the thickness is then doubled to 2.0 mm only an 8.37 % increase in counts is seen.

The full decay of 10^8 ^{90}Sr ions, was simulated for 100 mm^2 CdTe and GaAs detectors of 1 mm thickness to compare the spectra collected by each material. The spectra from these simulations are shown in Fig. 6. The GaAs detector counted 218,188 particles, and the CdTe detector 217,965. In this scenario the detectors therefore share virtually identical detection efficiency. The materials are distinguished by the shape of their spectra. A breakdown of the counts recorded in each quarter of the energy spectrum is displayed in Table 1. The CdTe detector features a higher and broader peak in the low energy range, centred at 0.013 MeV, fewer counts in the mid range, and greater counts in the high energy range. This can be attributed to the greater backscattering effect in CdTe, as low to mid range particles are more likely to backscatter while just depositing a fraction of their energy. Higher energy particles are more likely to be absorbed in CdTe, however this effect is limited as fewer particles are emitted in this range. The figure also plots the spectrum of emitted electrons and the spectrum observed in an ideal detector of identical dimensions. Each particle incident on this detector is completely absorbed. The typical activity concentrations for ^{90}Sr found at a site such as Sellafield, vary from the World Health Organisation water guideline limit of 10 BqL^{-1} to 59,000 BqL^{-1} . The simulations results presented here would correspond 1 or fewer counts observed at the water guideline limit after a monitoring period of 1 hour, and 4752 ± 69 counts at a location

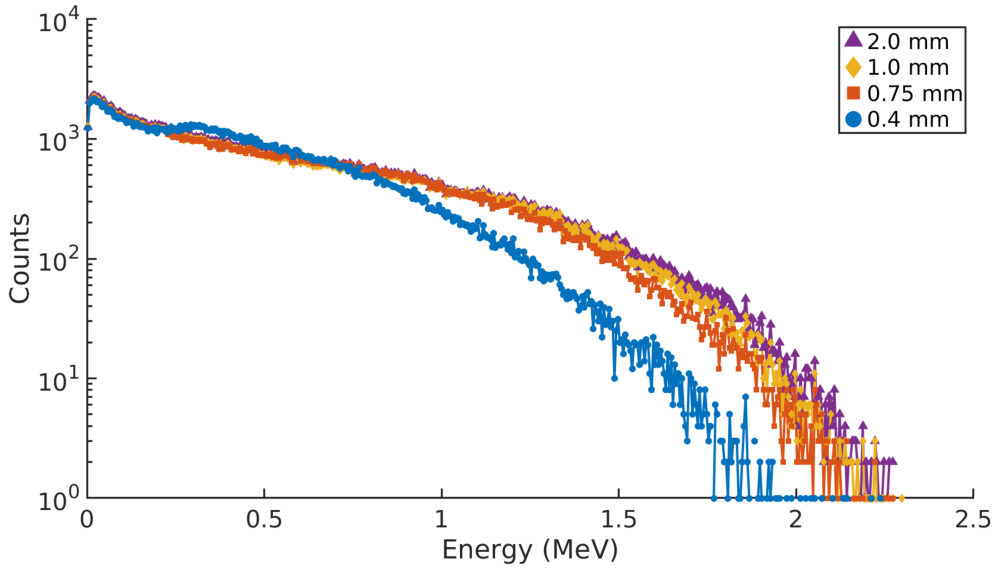


Figure 5. ^{90}Sr and ^{90}Y spectra observed in GaAs detectors with 100 mm^2 surface areas and varying thickness. Energy is resolved into 5 keV bins. Errors are omitted for visual clarity.

of peak activity. There are factors which may diminish the precision of this measurement such as noise introduced by electronic components, such as amplifiers, and background. An estimate of 1% uncertainty for these factors could increase the uncertainty to ± 82, whereas an estimate of 10 % would see it rise to ± 484. This suggests that the detector may be better suited as a device for monitoring areas of high activity.

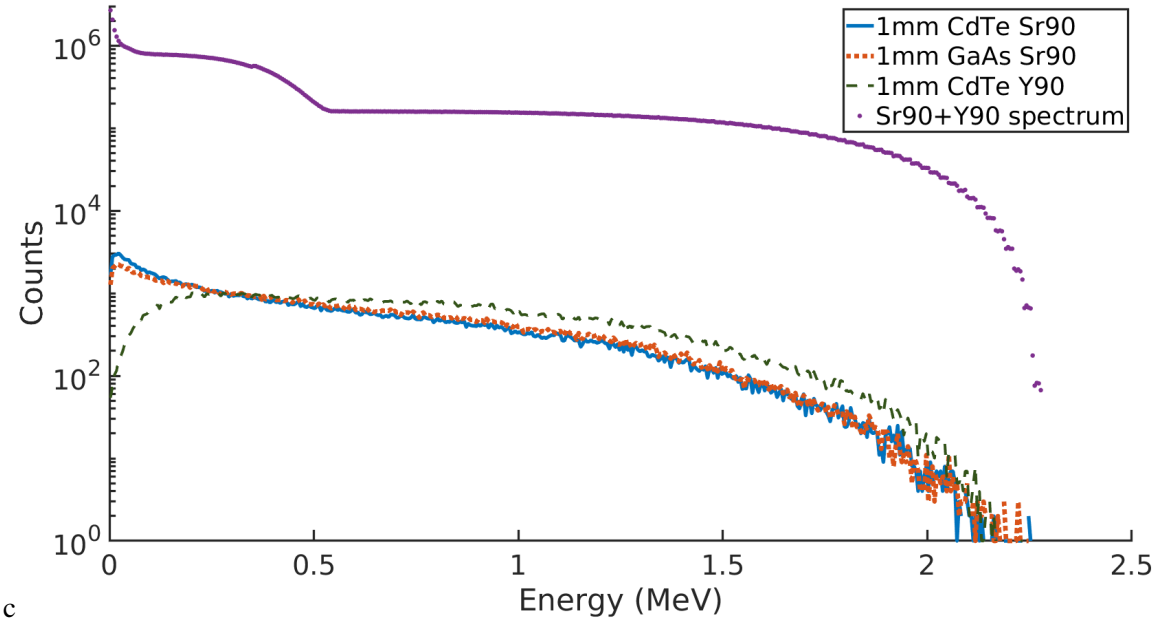
Table 1. Counts recorded in different energy ranges of the ^{90}Sr + ^{90}Y spectra observed in figure 6.

Material	0 - 0.575 MeV	0.575 - 1.150 MeV	1.150 - 1.725 MeV	1.725 - 2.300 MeV
GaAs	143,647 ± 379	55,448 ± 235	17,947 ± 134	1146 ± 34
CdTe	151,689 ± 389	49,258 ± 221	15,791 ± 126	1227 ± 35

3 Conclusions

The optimal design of a photodiode detector for the in situ measurement of ^{90}Sr has been investigated by modelling sensors and their absorption of beta radiation in a groundwater borehole scenario.

The complete absorption 0.546 MeV and 2.280 MeV beta particles in Si, GaAs and CdTe photodiodes was simulated to determine the most efficient absorber. The atomic number of a material both determines the propensity for that radiation to backscatter. The result is that as the thickness of GaAs and CdTe sensors increases, the complete absorption of 0.546 MeV beta particles plateaus. This occurs at approximately 300 μm and 400 μm for CdTe and GaAs sensors respectively. As backscattering is not as prevalent in Si, it absorbed more radiation completely at 800 μm. Sensors 1 mm thick made from CdTe, GaAs and Si absorbed 32.92 %, 23.66 % and 0.413 % of 2.28 MeV particles completely. This eliminated Si as a candidate material in this application.



c

Figure 6. ^{90}Sr and ^{90}Y spectra observed in GaAs (red and dashed) and CdTe (blue and solid) detectors with 100 mm^2 surface areas and 1 mm thickness. Energy is resolved into 5 keV bins. The spectra observed in an ideal detector is plotted (green and dash-dot) along with the spectrum of particles emitted (purple and dots).

Detector geometry was optimised by modelling a cylinder of ^{90}Sr contaminated groundwater. The number of counts observed in the detector scaled linearly with surface area, and the optimal sensor modelled featured a surface area of 100 mm^2. It was found that the thickness of the detector should exceed 400 μm for absorption of high energy particles (1.150 to 2.30 MeV) released during ^{90}Y decay. However, doubling the thickness of the sensor from 1.0 mm to 2.0 mm only yielded an 8.37 % increase in counts for particles in this range. The benefit of this must be weighed against the defects associated with higher volume detectors. The thickness of compound semiconductors can be limited by defects which restrict the movement of charge carriers, reducing charge collection efficiency [7, 10].

The merits of CdTe and GaAs detectors were compared by modelling 1 mm thick and 100 mm^2 surface area detectors. Spectra from 10^8 decaying ^{90}Sr ions and the resulting ^{90}Y daughters were collected. Each material offered similar detection efficiency, but the shape of the spectra collected varied slightly. While CdTe offered marginally better absorption in the energy range from 1.725 - 2.3 MeV, the backscattering which resulted upon contact with lower energy particles produced a larger peak at low energies compared to that observed in the GaAs detector. The GaAs sensor counted 8,456 more particles above 0.575 MeV, exclusively the result of ^{90}Y decay. Ultimately this may identify GaAs as the prime material when counting from a mixed source.

Acknowledgments

The authors would like to acknowledge the Nuclear Decommissioning Authority and the University of Glasgow for funding support.

References

[1] V. Chiste, "Table de Radionucleides." http://www.lnhb.fr/nuclides/Sr-90_tables.pdf, May, 2005.

[2] L. Y. Krestinina, F. G. Davis, S. Schonfeld, D. L. Preston, M. Degteva, S. Epifanova et al., *Leukaemia incidence in the Techa River Cohort: 1953–2007*, *British Journal of Cancer* **109** (Nov., 2013) 2886–2893.

[3] S. Musilli, N. Nicolas, Z. El Ali, P. Orellana-Moreno, C. Grand, K. Tack et al., *DNA damage induced by Strontium-90 exposure at low concentrations in mesenchymal stromal cells: The functional consequences*, *Scientific Reports* **7** (Jan., 2017) 41580.

[4] N. Vajda and C.-K. Kim, *Determination of radiostrontium isotopes: A review of analytical methodology*, *Applied Radiation and Isotopes* **68** (Dec., 2010) 2306–2326.

[5] G. Turkington, K. A. A. Gamage and J. Graham, *Beta detection of strontium-90 and the potential for direct in situ beta detection for nuclear decommissioning applications*, *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **911** (Dec., 2018) 55–65.

[6] A. Šagátová, B. Zaťko, V. Nečas, F. Dubecký, T. Ly Anh, K. Sedlačková et al., *From single GaAs detector to sensor for radiation imaging camera*, *Applied Surface Science* **461** (Dec., 2018) 3–9.

[7] S. Del Sordo, L. Abbene, E. Caroli, A. M. Mancini, A. Zappettini and P. Ubertini, *Progress in the Development of CdTe and CdZnTe Semiconductor Radiation Detectors for Astrophysical and Medical Applications*, *Sensors* **9** (May, 2009) 3491–3526.

[8] M. Sammartini, M. Gandola and G. Bertuccio, *Advanced Theoretical Models for Charge Collection in CdTe Radiation Detectors: A comparison based on experimental data*, in *2017 IEEE Nuclear Science Symposium and Medical Imaging Conference (NSS/MIC)*, pp. 1–3, Oct., 2017. DOI.

[9] G. C. Sun, H. Samic, J. C. Bourgoin, D. Chambellan, O. Gal and P. Pillot, *A comparison between GaAs and CdTe for X-ray imaging*, *IEEE Transactions on Nuclear Science* **51** (Oct., 2004) 2400–2404.

[10] M. Veale, S. Bell, D. Duarte, M. French, A. Schneider, P. Seller et al., *Chromium compensated gallium arsenide detectors for X-ray and γ-ray spectroscopic imaging*, *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **752** (July, 2014) 6–14.

[11] G. Bertuccio, *Prospect for energy resolving X-ray imaging with compound semiconductor pixel detectors*, *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **546** (July, 2005) 232–241.

[12] C. Erd, A. Owens, G. Brammertz, M. Bavdaz, A. Peacock, V. Lämsä et al., *Hard X-ray test and evaluation of a prototype 32×32 pixel gallium–arsenide array*, *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **487** (July, 2002) 78–89.

[13] X. Wu, T. Peltola, T. Arsenovich, A. Gädda, J. Härkönen, A. Junkes et al., *Processing and characterization of epitaxial GaAs radiation detectors*, *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **796** (Oct., 2015) 51–55.

[14] G. Lioliou, M. D. C. Whitaker and A. M. Barnett, *High temperature GaAs X-ray detectors*, *Journal of Applied Physics* **122** (2017) 244506.

[15] S. V. Chernykh, A. V. Chernykh, S. I. Didenko, F. M. Baryshnikov, N. Burtebayev, G. I. Britvich et al., *GaAs detectors with an ultra-thin Schottky contact for spectrometry of charged particles*, *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **845** (Feb., 2017) 52–55.

[16] S. H. Shin, G. T. Niizawa, J. G. Pasko, G. L. Bostrup, F. J. Ryan, M. Khoshnevisan et al., *P-I-N CdTe Gamma Ray Detectors by Liquid Phase Epitaxy (LPE)*, *IEEE Transactions on Nuclear Science* **32** (Feb., 1985) 487–491.

[17] J. Becker, M. W. Tate, K. S. Shanks, H. T. Philipp, J. T. Weiss, P. Purohit et al., *Characterization of CdTe sensors with Schottky contacts coupled to charge-integrating pixel array detectors for X-ray science*, *Journal of Instrumentation* **11** (Dec., 2016) P12013–P12013.

[18] G. F. Knoll, *Radiation Detection and Measurement*. Wiley, New York, 3rd ed ed., 2000.

[19] "Groundwater monitoring at Sellafield: 2016 data review." https://www.gov.uk/government/publications/groundwater-monitoring-at-sellafield-2014-data-review.

[20] E.-W. Bai, K.-s. Chan, W. Eichinger and P. Kump, *Detection of radionuclides from weak and poorly resolved spectra using Lasso and subsampling techniques*, *Radiation Measurements* **46** (Oct., 2011) 1138–1146.

[21] "Geant4." http://www.geant4.org/geant4/.