Beta detection of strontium-90 and the potential for direct in situ beta detection for nuclear decommissioning applications

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Abstract

Strontium-90 is one of the primary beta-emitting radionuclides found at nuclear decommissioning sites. Monitoring its activity in the environment is of utmost importance given its radiotoxicity. Current procedures for the beta detection of strontium-90 are time consuming, produce secondary waste and expensive. There is a demand for real-time in situ radiostrontium monitoring in groundwater at nuclear decommissioning sites. This paper presents a review of existing techniques for strontium-90 monitoring and examines a novel approach through direct beta detection with a gallium arsenide photodiode based detector. A proof of concept detector was modelled in the physics simulation software, Geant4, and evaluated as candidate for in situ detection of beta emitting radionuclides. The simulation results indicate that the detector is physically capable of counting 89.86% of incident 0.546 MeV electrons from a 1 mm range in water. This validation will provide the basis for further development of an in situ beta detector.

Keywords: strontium, beta, radiation, detection, photodiode, gallium-arsenide, groundwater

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1. Introduction

Strontium-90, ^{90}Sr , is a beta emitting radioisotope produced during nuclear fission and has been dispersed into the environment as a result of accidents at nuclear power plants, leaks from nuclear waste storage and as fallout from nuclear weapons testing. Commonly known as a "bone seeker", ^{90}Sr is chemically similar to fellow alkaline metal calcium, and when it is ingested into the body it has the propensity to accumulate in bone structure [1, 2]. Given the long half-life of ^{90}Sr , 28.8 years [3], its presence in the body can lead to prolonged irradiation of skeletal bone structure, increasing the risk of damage to bone marrow, leukaemia and other bone cancers [4, 5]. As a consequence, it is of great importance to monitor its activity in the environment, particularly in groundwater surrounding nuclear facilities.

^{90}Sr is one of the major beta emitting radionuclides found at the Sellafield site in Cumbria, UK. Leaks and spills from corroded Magnox fuel cladding silos and neutralised nitrate containing wastes [4, 6] have introduced radiostrontium into the environment, where it has mixed with groundwater. Currently, counting beta emitting radionuclides is a long and arduous process. Samples must be collected from groundwater boreholes, transported to a laboratory, and processed with hazardous chemicals before the activity can be measured. With nuclear decommissioning set to continue at sites like Sellafield for the next 100 years and more, these procedures present logistical and financial challenges for the nuclear industry. The recent disaster at the Fukushima Daiichi nuclear power plant (FDNPP) has brought increased scrutiny on the proliferation of ^{90}Sr in the environment and highlighted the need for rapid and agile measurement procedures [7, 8].

Traditionally, contaminated groundwater is collected for analysis from monitoring boreholes installed into the groundwater table. Within an aquifer comprised of unconsolidated deposits, a filter pack and slotted PVC screen are typically installed into a borehole at a target depth. These features prevent the influx of aquifer material while allowing water to flow into the borehole. Water samples may then be pumped to the surface and transported to the laboratory where ^{90}Sr activity is determined by radiochemical analysis. This paper presents a review of the current techniques used in beta detection and explores the potential for in situ beta-counting by direct detection with gallium arsenide photodiode based detectors. This scenario is illustrated in Figure 1, where a detector is placed directly into contaminated groundwater in the screened interval of a borehole.

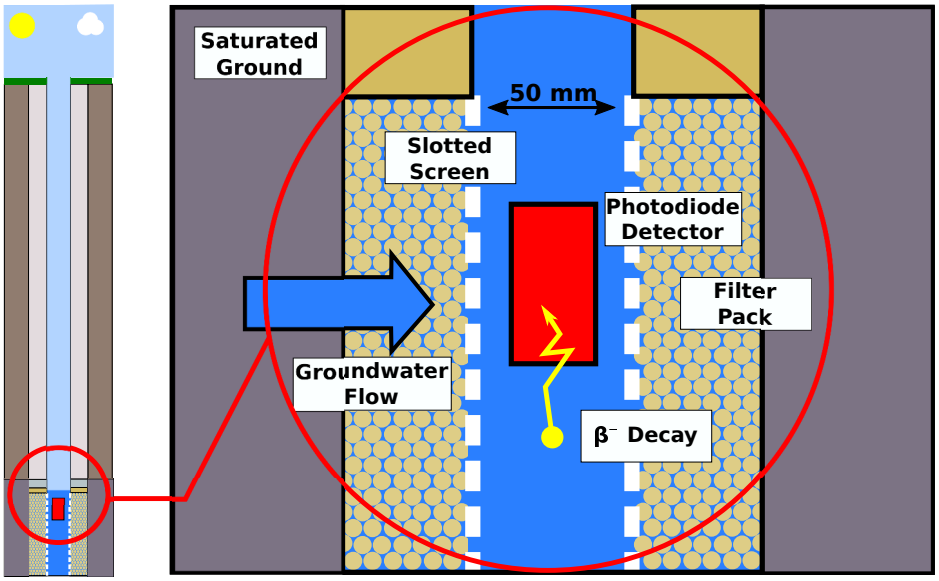

Figure 1: A simplified diagram of a typical groundwater borehole scenario, and the prospective deployment of an in situ photodiode detector.

2. Beta radiation

Beta particles are electrons (e^-) or positrons (e^+) which are emitted during nuclear decay processes. When an unstable nucleus decays via beta emission, a neutron transforms into a proton and excess energy is shared between the emitted beta particle (e^-) and an antineutrino ($\overline{\nu}$). Equation 1 illustrates this through the beta-decay of ^{90}Sr . ^{90}Sr is a pure beta-emitter with a maximum energy of 0.546 MeV [3]. Its daughter nuclei Yttrium-90, ^{90}Y has a shorter half-life of 64 hours and decays itself, via 2.28 MeV beta emission, into stable Zirconium-90, ^{90}Zr. The short half-life of ^{90}Y means that it is often found in secular equilibrium with ^{90}Sr , a property which can be exploited in radiochemical analysis.

$$^{90}_{38}Sr \longrightarrow ^{90}_{39}Y + e^- + \overline{\nu} \tag{1}$$

Beta particles are emitted over a continuous energy spectrum, from zero to their maximum end-point energy, see Figure 2. Depending on their initial energy, beta particles may have a range of a few metres in air, centimetres in water and millimetres in aluminium [9]. Fast moving electrons typically ionise matter as a result of inelastic coulomb collisions. When a fast-moving

electron is decelerated, usually by the electric field of an atomic nucleus, excess energy may be released in the form of electromagnetic radiation, known as Bremsstrahlung radiation [10].

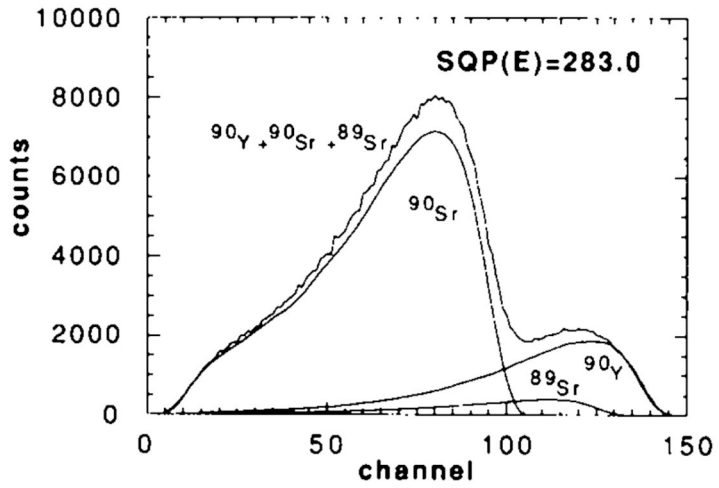

Figure 2: The gross beta spectrum observed, with a liquid scintillation spectrometer, from a sample containing ^{90}Sr , ^{90}Y and ^{89}Sr, where the constituent spectra have been deconvolved. SQP(E) refers to the quenching parameter attributed to the sample [11].

As a beta particle travels through matter, coulombic interactions cause it to lose energy, eventually to the point at which it is no longer detectable. This limited range in matter has presented difficulties when designing beta detectors. In an ideal scenario a direct detector would be small, lightweight and immersible in the detection medium. The detector must operate at very close range in order to capture particles before they have lost a significant fraction of their energy. In addition, an ideal device would be compatible with existing borehole dimensions, be low maintenance and produce no waste. Wireless communications and solar power could remove the need for obstructive headworks, thereby reducing infrastructure requirements. However, in the past these technologies have not been sufficiently developed or readily available. In light of this, many existing techniques have adopted an indirect approach to detection. Indirect radiation detection sees the source stimulate a scintillator, which produces flashes of light which are in turn detected by a photomultiplier tube (PMT) or photodiode. These techniques see water pumped to the surface for sample collection, necessitating surface infrastructure and regular maintenance.

3. Existing Methods for ^{90}Sr monitoring

When a sample of unknown radionuclear composition is collected from the environment, the overlapping of their energy spectra makes it difficult to identify individual beta emitters by spectroscopy. For many beta monitoring techniques, it is essential to isolate the target radionuclide from the sample matrix, thereby removing any other source of radiation entering the detector. There are a number of different methods which can be employed to achieve this, including precipitation [12], liquid-liquid extraction [13], solid-phase extraction [14] and chromatography [15]. When multiple radionuclides are present in the sample, which is inevitable as ^{90}Sr decays to ^{90}Y , it is possible to resolve radionuclides by their spectra provided their beta energies differ significantly [16]. This is achieved by measuring activity over multiple energy windows, and using the resulting information to mathematically resolve the individual energy spectra of the radionuclides present. C.K. Kim et al. demonstrated a variation on this technique designed for rapid response to emergency scenarios. Their two window approach generated results with minimum detectable limits compliant with IAEA safety standards, with a counting time of just 1.5 hours [17].

Once a radionuclide has been isolated, it can be measured using existing beta counting devices, such as gas ionisation chambers and liquid scintillation counters (LSC). Gas ionisation counting is one of the oldest radiation counting techniques, and measures the avalanche of charge induced as ionising radiation traverses a gas. Despite no longer being at the cutting edge of technology, proportional gas counters have remained popular over the years due to their simplicity, cheap construction and low operational costs. They are still used in the standard procedure for ^{90}Sr monitoring in seawater by the Japanese Government [18]. Liquid scintillation counting (LSC) sees a cocktail of organic fluorescence compounds stimulated by ionising radiation into the emission of light which can be detected and used to determine the activity of a radioactive source. Given the low energy of some beta-emitters and the relatively short penetration depth, LSC has become the most widely used technique for measuring pure beta emitters [19]. However, beta counting by Cherenkov radiation and gas proportional counters are the primary groundwater counting procedure used at Sellafield. Provided the composition of the groundwater sample is known, gross beta counting may offer a cheaper and quicker alternative. However, naturally occurring ^{40}K and ^{90}Sr may conceal ^{90}Sr contamination from nuclear waste, meaning results are not

suitable. This section shall review the Cherenkov counting procedure, from sample preparation to activity counting, and highlight the short-comings of this technique for nuclear decommissioning applications.

3.1. Groundwater sample collection and pre-treatment

In a traditional groundwater monitoring programme, samples of groundwater are obtained from installed monitoring boreholes, typically through either a dedicated or portable pump [20]. Samples for gross beta analysis are then filtered, if a dissolved concentration measurement is required, before collection in a pre-acidified plastic container [21], which gives a maximum recommended holding time of 1 month or 2 months for ^{90}Sr .

3.2. Radiochemical separation

Many beta counting procedures cannot resolve the spectra of different beta emitting particles, therefore in order to accurately determine the activity of ^{90}Sr in an environmental groundwater sample, it must be separated from contaminants and other radionuclides which may interfere with the counting process. A number of techniques have been developed over the years, each with its own advantages and disadvantages. This section shall investigate the three most commonly used procedures, precipitation, liquid-liquid extraction and extraction chromatography.

The oldest method for radiostrontium separation is by precipitation. In this procedure, strontium is separated from calcium by exploiting the different solubilities of Ca and Sr nitrates in concentrated fuming nitric acid [22, 12]. Radium, lead and barium are collected with barium chromate and the remaining fission products are eliminated with iron hydroxide. ^{90}Y can be separated with hydroxide precipitation and prepared as an oxalate, ready for counting [12]. This procedure, and ones similar to it, have been developed, popularised and standardised since the 1960s. The precipitation technique was popularised because it is robust, efficient and can be applied to large volumes of samples. However it is also laborious, precipitations must be repeated several times to sufficiently extract strontium from the sample [23, 24]. In addition, the health and safety risks, posed through the use of extremely hazardous chemicals, has motivated the development of more rapid and safe techniques.

The liquid-liquid extraction technique selectively isolates radionuclides with the use of two immiscible chemical solvents, typically water and an organic solvent. When the analyte is favourably soluble in the solvent, it

will distribute itself from one phase to another, almost completely [13]. This concept can be used to either separate ^{90}Y from the sample, for indirect measurement of ^{90}Sr activity, or for selective extraction of ^{90}Sr using crown ethers. ^{90}Y stripping from the sample is achieved with the use of tri-n-butyl phosphate (TBP), an organic extractant compound [25]. The organic solvent must then be discarded by washing the sample with water, leaving the remaining ^{90}Y to be precipitated to oxalate form and counted by Cherenkov methods.

Extraction chromatography with crown ethers was investigated by Horowitz et. al. in 1990. A crown ether, 4,4,(5')-bis(tert-butyl cyclohexano)-18-crown-6 in 1-octanol, was sorbed onto an inert substrate and used to selectively capture the strontium ions of interest [15]. Given the simplicity of preparation of the ether, and its strong performance in removing strontium from a nitric acid sample, the ether was commercialised and is now sold as Sr Resin, produced by EiChrom Industries, Inc [24]. Sr Resin has been widely adopted in the nuclear industry because it is simple, can be completed in a few hours and is attractive economically since the resin can be reused. However, as the properties of Sr Resin were further investigated, some downsides were revealed. The process of acidifying large volumes of water samples requires precipitation which is time-consuming and may be completed at the expense of some strontium [26], particularly relevant when considering low activity samples. Other extraction chromatography products have been manufactured, including 3M Empore^{TM} Strontium Rad Disks and AnaLig®(R) gels. Empore^{TM} Rad Disks consist of a mesh of PTFE (teflon) fibres hosting AnaLig Sr-01^{TM} selective adsorption chromatographic ligands [14, 27]. Essentially these filters consist of specifically crafted polymers, templated on the desired molecule for extraction. The result is a very selective procedure which is capable of separation ^{90}Sr from even its daughter nuclei, ^{90}Y [28].

Table 1: A comparison of radiochemical separation procedures for ^{90}Sr in groundwater.

Method	Avg. radiochemical yield ^{85}Sr %	Avg. Activity ± 2U (Bq dm^{-3})
3M Empore^{TM} Sr Rad Disk	96	377.6 ± 43.2
AnaLig® Sr-01(60–100 mesh)	99	328.8 ± 36.9
AnaLig® Sr-01(230–425 mesh)	97	383.9 ± 43.3
Sr®(R) Resin	89	319.8 ± 34.4
Liquid extraction —TBP	86	377.9 ± 25.0
Carbonate precipitation	54	375.5 ± 45.5

A comparative investigation of five different radiochemical separation

techniques for ^{90}Sr in water was undertaken by J. Ometakova et al. in a 2011 study [29]. The traditional techniques of strontium separation, carbonate co-precipitation and TBP liquid-liquid extraction, were compared along side commercial Solid Phase Extraction (SPE) techniques using 3M Empore^{TM} Strontium Rad Disks and AnaLig(R) Sr-01 resin at two different mesh levels. The results, summarised in table 1, compare modern SPE extraction with older techniques. SPE achieved higher radiochemical yields while also being substantially quicker and easier to complete compared to liquid-liquid extraction and precipitation. Separation using 3M Empore^{TM} Strontium Rad Disks was possible in 20 minutes. The authors also found that the traditional methods incurred large volumes of liquid waste as well as the use of hazardous concentrated acids. This is of significance to the nuclear decommissioning industry where thousands of samples must be prepared each year, it is highly desirable to reduce the production of secondary waste as much as possible.

3.3. Cherenkov radiation counting

When a charged particle moves through a medium with a velocity greater than the phase velocity of light in that medium, energy is released in the form of light known as Cherenkov radiation. Typically the photons released as Cherenkov radiation are from the UV and visible portion of the electromagnetic spectrum, hence the characteristic blue glow which can be observed in images from the interior of nuclear reactors. This phenomenon has been utilised in the detection of beta particles released from ^{90}Sr and its daughter nuclei. To produce Cherenkov radiation in a medium, such as water, beta-particles must exceed a threshold energy which is dependent on the refractive index of the medium [30, 31]. As such, the refractive index of the selected medium can be used to discriminate between different sources of radiation as the maximum energy of emitted energy from the radioisotope must greatly surpass the threshold energy, given the spectrum of energised beta-particles released [32]. The light produced by Cherenkov radiation can be measured with existing commercial liquid scintillators [33, 34, 35].

Cherenkov radiation counting has a few advantages over similar liquid scintillation techniques. The sample used in Cherenkov counting does not need to be incorporated into a scintillation cocktail, resulting in more efficient sample preparation, disposal and the ability to reuse samples [34]. One of the primary performance limiting factors in Cherenkov counting is known as quenching. This is any process which reduces the intensity of

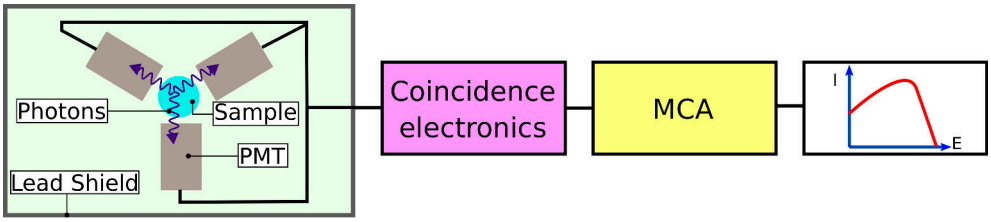

Figure 3: A schematic view of the primary components in a typical TDCR LSC device.

light available for detection by the PMTs. The primary quenching effect in Chernekov counting is colour quenching, simply a consequence of discolouration of the sample, which contributes to the absorption of light emitted during the Cherenkov process. However, this can be compensated by calibration with a colour quench correction curve [35, 36].

An alternative is to use the Triple to Double Coincidence Ratio (TDCR) technique. TDCR has recently become popular with many national metrology institutes as a method of determining primary activity standards. It is an absolute method of determining radioactivity in a source and requires no reference to internal or external sources. TDCR requires a liquid scintillation detector with three photomultiplier tubes (PMT) uniformly arranged around a sample, Figure 3, with an electronics package capable of recording triple and double coincidence events [37]. The activity of the source is calculated with a free-parameter statistical model, which considers assumptions about the number of electrons generated during a decay event in the detector [38, 39].

J. M. Olfert et. al. investigated a method for the rapid determination of ^{90}Sr and ^{90}Y in water samples by liquid scintillation and Cherenkov counting [40]. Groundwater samples were collected from the discharge of a groundwater plume, filtered and acidified in preparation for counting. This study compared five different techniques for ^{90}Sr analysis: direct TDCR counting of ^{90}Y , LS counting for ^{90}Sr and ^{90}Y after radiochemical separation, Cherenkov counting for ^{90}Y after radiochemical separation and LS counting of the ^{90}Sr sample for ^{90}Y in growth. After direct Cherenkov counting of ^{90}Y , the samples were radiochemically separated, using Sr and DGA-N resins, into ^{90}Sr and ^{90}Y . The ^{90}Sr sample was counted via LSC, and recounted after 8 days to allow for ^{90}Y in growth. Meanwhile, the ^{90}Y sample was measured by Chernkov and LSC. Each sample was counted with a Hidex 300SL TDCR Liquid Scintillation counter [41]. The results produced by each technique

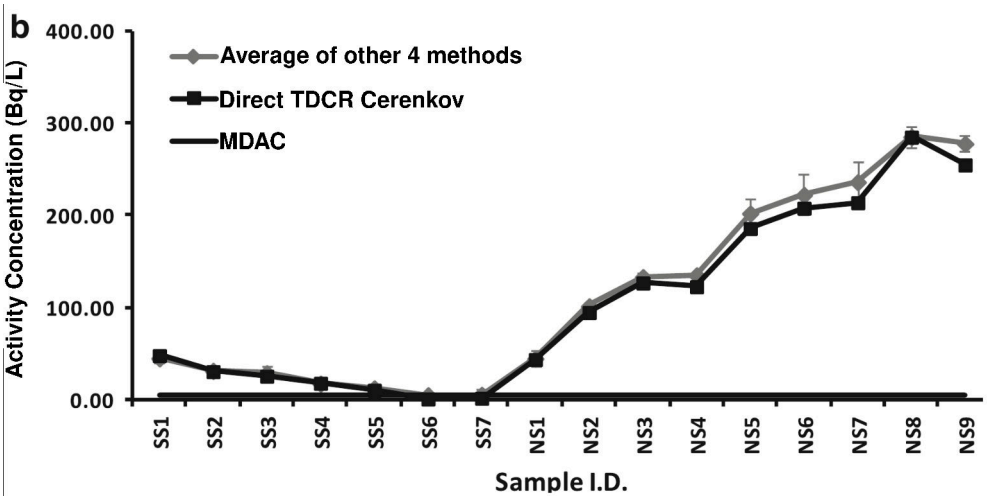

Figure 4: A comparison of radiochemical separation procedures for ^{90}Sr in groundwater [40]. MDAC refers to the minimum detectable activity concentration.

were consistent with each other, validating the TDCR technique against the standard radiochemical procedure as a method for detecting beta-emitting radionuclides (see Fig. 3). This affirmation highlights TDCR as a technique with a number of advantages over radiochemical separation. The procedure is fast, requires no sample preparation and does not suffer from chemical quenching. However, radiochemical separation assures that no interfering radionuclides are present in the sample and results were produced within 1 day.

Initially, with a lack of suitable detectors commercially available, many TDCR systems were purpose-built in laboratories. In the last few years, commercially produced detectors have become available such as the Hidex 300SL. However, these detectors are large and immobile, rendering them ill-suited for in situ detection purposes. The European Metrology Research Programme (EMRP) and the Joint Research Project MetroFission launched an initiative to design and develop a portable TDCR device for use in situ at next generation power plants. Four different national metrology institutes, NPL (UK), ENEA (Italy), LNHB (France), and PTB (Germany) were tasked with producing a device. Each design had to be distinct yet conform with a number of fundamental design principles. Chiefly that the device must fit into a standard car and be light enough to be comfortably handled by one person [42].

The PTB design featured three channel photomultipliers packed into a compact optical chamber itself ensconced in a foam carrying case, a portable mini NIM bin to house the electronics and a portable PC for data acquisition and processing [37, 43]. Initial validations of the device found that it could measure the activity of some high energy nuclides, such as ^{90}Sr , with uncertainties under 1% and a similar percentage deviation from reference TDCR measurements. However, the performance of the device suffered when measuring lower energy emitters as uncertainty contributions from low count statistics and background radiation took on increased significance in the model. It was concluded that while the device was not of the standard required for metrology applications, the device could be sufficient for other field-based research. Indeed each of the devices produced in the design initiative showed promising results in their initial validation measurements and offer promising potential for further development.

Many procedures for Cherenkov counting of ^{90}Sr have been developed over the years [44, 45] and the technique has become the primary method for ^{90}Sr analysis at the Sellafield decommissioning site. Currently, groundwater samples are pumped from boreholes and transported to laboratories for analysis. Strontium is separated using ion exchange resins, and counted in a two window approach[46]. The activity of the ^{90}Sr source is counted immediately after separation and again, after 20 to 30 days, once it has achieved secular equilibrium with its daughter nuclei ^{90}Y . The activity of the original ^{90}Sr source is determined by the ingrowth of ^{90}Y . Across the Nuclear Decommissioning Authority, thousands of groundwater and solid samples require analysis each year, and there is demand for ever more data to provide a greater understanding of groundwater systems [46]. As this demand increases, the financial and temporal costs associated with Cherenkov counting will mount. This puts financial and organisational strain on decommissioning sites and is the motivation for an alternative approach.

3.4. Demand for next generation beta detectors

Sellafield, and other nuclear decommissioning sites, must plan their operation for the next 100+ years and there is an increasing demand for data to be collected more frequently and in real-time while decreasing lifetime monitoring costs. This functionality will allow sites to immediately respond to unexpected spikes in groundwater mobility, which may have gone undetected with existing monitoring routines. In addition, more frequent data acquisition would allow decommissioning sites to enhance their understanding of

groundwater systems and the daily factors which influence contaminant mobility. This would provide evidence in the development of conceptual models for radionuclide transport in groundwater, and enhance safety assessments which indicate whether groundwater strontium is being managed correctly. Although more rapid and streamlined versions of existing techniques have been developed [18, 28] , these are reserved for use in emergency scenarios and still suffer from the same drawbacks in terms of chemical waste, sample collection and expenses.

Beta detection by gas ionisation chambers, liquid scintillation counting and Cherenkov counting is very sensitive and precise, with minimum detectable limits of activity well below the standards required to meet World Health Organisation (WHO) guidelines, 10 BqL^{-1} [47]. Indeed, LSC has been adopted by many metrology institutions across the world, such is the calibre of results it can provide. However, the practicalities of these procedures present hazards for workers through manual handling of samples and the risk of exposure to radiation. Samples must be collected from remote locations, delivered to laboratories, treated with chemicals, counted and finally disposed of. As these procedures must be completed hundreds of times per year for the duration of the facility's lifetime, there will be significant operating costs and production of secondary wastes.

This paper seeks to propose an alternative approach to beta spectroscopy, through direct in situ detection of beta radiation. Attempts have been made to produce in situ versions of the weighty lab-based detectors required for existing techniques, but this does little to satisfactorily address the demands for real-time radiostrontium monitoring in groundwater. This paper considers a novel approach where the detector is deployed within groundwater boreholes, directly at the source of radiation. This would require a radiation detector that is sensitive to beta radiation, offers real-time detection, while also being highly portable. Such a device would be unburdened by time-consuming sample collection and chemical treatment procedures. One potential solution comes in the form of photodiodes, adapted for use as direct radiation detectors. Photodiodes are devices designed to convert light into electrical current, recognisably used in solar panels. The same mechanisms that allow for the conversion of visible light to current also apply to ionising radiation.

4. PIN photodiodes

This research aims to develop a highly mobile, fast and efficient beta-radiation detector, free from the lengthy chemical separation and counting procedures outlined in the previous section. To this end, PIN photodiodes are being investigated as candidate for a light weight radiation detector. Initially developed to detect photons and used as an alternative to PMTs, photodiodes have been increasingly investigated in recent years as a tool for direct radiation detection. In comparison with gas-filled or scintillation detectors, semiconductors have a lower energy requirement for charged particle detection resulting in superior energy resolution [48]. They now have a number of applications in medical imaging, dosimetry, power generation and high-energy radiation experiments[49, 50, 51, 52].

In contrast to PN-junction semiconductors, PIN photodiodes have a large intrinsic layer separating the p and n-type layers. Figure 5 illustrates a simplified configuration of a PIN photodiode and its interaction with ionising radiation. As radiation enters the intrinsic layer, it disrupts electron-hole pairs which are swept up by a reverse-biased voltage and the resulting current is measured. The more energy is deposited, the greater the current pulse produced. By extending the size of the depleted region, a larger volume is presented for ionising radiation to fully deposit its energy within the sensitive region of the detector.

A number of materials are used to construct semiconductor photodiodes, each with their own strengths and weaknesses. Silicon and germanium rapidly emerged as widely used semiconductors, largely due to early advances in manufacturing allowing for high quality devices to be made cheaply and quickly. However, these materials have properties which make them less than ideal candidates for in situ beta detection. Germanium detectors offer excellent energy resolution but, due to a very narrow bandgap of 0.66 eV, require cooling to liquid nitrogen temperatures to reduce thermally induced noise [10]. This clearly makes germanium ill-suited for mobile applications. While silicon has a wider bandgap, 1.1 eV, it still requires cooling and its low atomic number, 14, means it has relatively poor stopping power for ionising radiation.

4.1. Gallium arsenide

Gallium arsenide has a number of properties which make it an attractive alternative to silicon and other semiconductor materials. Gallium and

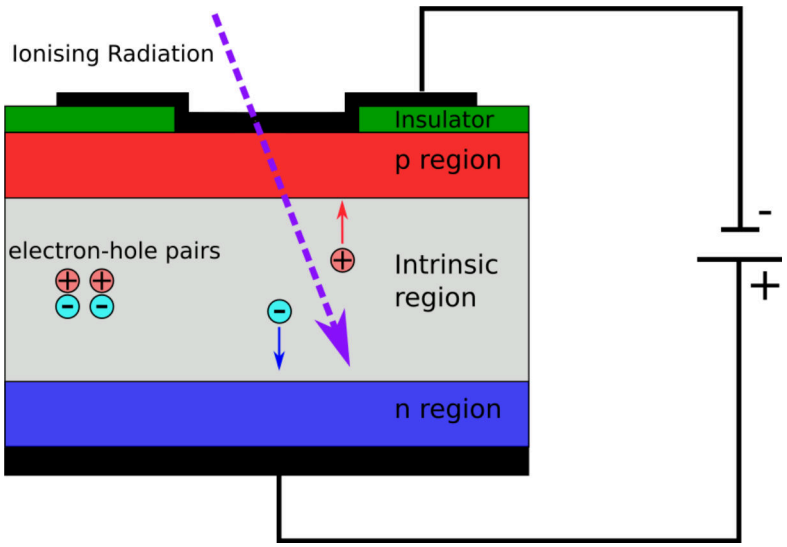

Figure 5: A simplified view of ionising radiation generating a current pulse in a PIN photodiode operating under reverse biased voltage.

arsenic have atomic numbers of 31 and 33, respectively, resulting in the material having greater stopping power for ionising radiation, like X-rays, in comparison with silicon devices [53, 54]. The bandgap of the compound material, 1.42 eV, is wide enough that devices can be operated at room temperature [55, 53, 54] without overwhelming thermal noise. Table 2 compares some of the fundamental properties of GaAs and Si. As the electron mobility of the GaAs is approximately 6 times greater than Si, this should allow for a device which functions over a shorter time scale.

Table 2: A comparison between the semiconductor properties of Si and GaAs [56].

Property	Silicon	Gallium-arsenide
z	14	32
ρ (gcm^{-3})	2.33	5.31
Radiation Length (cm)	9.36	2.3
Pair Production Energy (eV)	3.55	4.27
Electron Mobility (cm^2/Vs)	1500	7000-8500

Radiation detectors, unsurprisingly, are bombarded with ionising radiation and it is important that the device is not degraded or significantly damaged over time. The ability of a material to withstand the damage is known

as its radiation hardness. There are two principle ways in which radiation can negatively affect a semiconductor, displacement damage and ionisation damage. Displacement damage refers to the permanent physical dislocation of atoms from their lattice positions by incoming radiation. This produces defects in the material resulting in intermediate energy states, facilitating easier separation of electron-hole pairs, thus generating current and contributing to noise in the detector. Additionally, charges can become trapped on intermediate levels, which will negatively affect counting statistics. Ionisation damage occurs after energy deposition in the detector frees electron-hole pairs which drift to other locations and become trapped. When sufficient concentrations of charge are trapped, localised parasitic electric fields can develop [57].

A. Sagatova et al investigated the radiation hardness of GaAs devices against gamma radiation, high energy electrons and fast neutrons [58]. The detectors, beams and doses used in their experimentation is summarised in Table 3. An Am^{241} gamma spectrum was captured at each dosage and the results of electron damage on the spectra acquired, photopeak area, charge collection efficiency (CCE) and full width half maximum (FWHM), indicative of the energy resolution, were documented. Curiously, the results reported indicated that low doses, 1 kGy, of electron damage may even improve the performance of the detector, and this was attributed to the radiation damage compensating for pre-existing defects in the device. The study found that the damaging induced by electrons was up to 10 times worse than that of gamma photons, and up to 1000 time worse for neutrons. Indeed, the device was no longer functional after a dose of 0.576 kGy of fast neutrons, and was still functional, albeit in a limited capacity, after the full course of electrons and gamma.

Table 3: Experiments carried out by A. Sagatova et al [58] determining the radiation hardness of GaAs.

Radiation	Detector Thickness (μm)	Energy (MeV)	Max Dose (kGy)
Gamma	250	1.33	1140
Electrons	230	5	104
Neutrons	300	2-30	3.215

The radiation damage in GaAs sensors has been investigated by numerous studies [56, 59, 60] and has been compared favourably with silicon [60]. The strong radiation hardness of GaAs, has seen the material used radiation harsh environments like high energy particle accelerators and satellites, and

suggests that GaAs is a viable candidate for use at nuclear waste disposal facilities.

In recent years, GaAs photodiodes garnered increasing interest for their potential as X-ray imaging devices. One of the hurdles in this has developing detectors with intrinsic layers thick enough to stop energetic radiation such as X-rays, with as few defects as possible to maintain strong energy resolution. Detectors designed for beta detection will likely require similarly thick intrinsic layers.

The major imperfection associated with GaAs crystals is known as the EL2 defect. This defect is present in many fabrications techniques, including Metal Organic Chemical Vapour Deposition (MOCVD) and Liquid Encapsulated Czochralski (LEC), however is notably absent in Molecular Beam Epitaxy (MBE) [61]. The exact nature of this defect is the subject of much debate, but it is known to produce a midgap deep level, essentially a trap for electrons. The presence of the EL2 defect restricts the sensitive volume of the device, which is crucial for radiation detection. The ionised form of the defect meanwhile reduces electron lifetime, dampening charge collection efficiency and therefore energy resolution [51]. This defect has presented a stumbling block in the use of GaAs photodiodes in sensitive applications, such as medical imaging and radiation detection.

4.2. Applications in radiation detection

One of the limiting factors in the adoption of GaAs devices as radiation detectors has been the presence of defects, such as EL2, in the bulk material. This induces a small sensitive layer thickness, low values of electron charge collection efficiency, current oscillations in the detector and non-uniform field distribution. A. V. Tyazhev et al [62] noted these flaws in the LEC process. As an alternative, they proposed using chromium compensated GaAs layers. Their study validated the composition through I-V characterisation, electric field distribution tests and CCE assessment. It was found that chromium doped GaAs offers high resistivity, thickness approaching 1 mm, more uniform electric field distribution and functional levels of CCE for use in X-ray pixel detectors. Figure 6 offers a point of comparison between a LEC GaAs device, operating at 250 V, and a chromium compensated device, on the right. The function plotted is $F = 1 - T$, where T is the spatial distribution of light transmission through the detector thickness. This demonstrates that chromium compensated GaAs structures offer a more uniform electric field

distribution, which is stable over a range of operating voltages. This uniformity of electric field is significant, because it allows for detectors with very thick intrinsic layers to be operational.

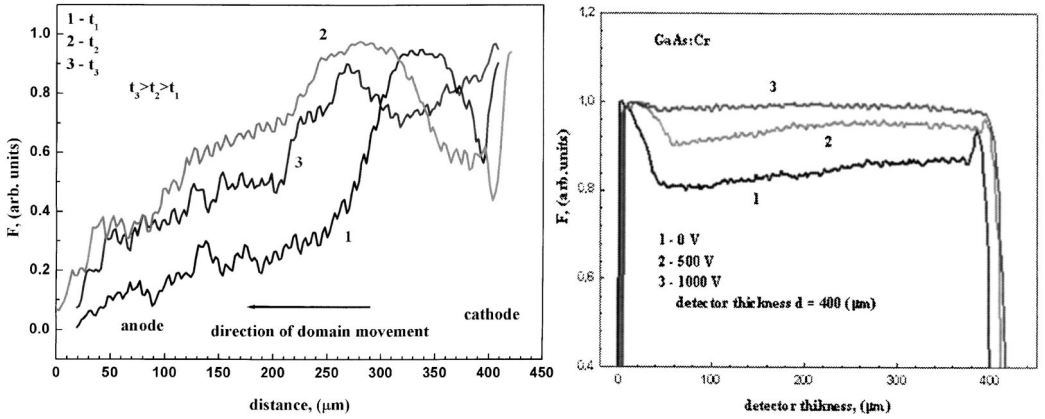

Figure 6: These graphs plot the function F, a cypher for electric field distribution, through a LEC frabricated detector, left, and a chromium compensated detector, right [62].

Building on A. V. Tyazhev's research, M.C. Veale et al [51] produced 500 μm thick chromium compensated GaAs devices and tested their potential for X-ray and γ-ray spectroscopy and imaging. The GaAs wafer was affixed to Schottky electrodes, etched with an 80x80 array of 200 μm anode pixels and bonded to a HEXITEC ASIC readout chip. I-V characteristics for the device were measured at 280 K and 298 K, with the room temperature resistivity measured as 2.5×10^9 Ωcm. To investigate the spectroscopic ability of the detector, an ^{241}Am γ spectrum was collected at 280 K. The FWHM of the 60 keV photopeak in this figure is 2.9 keV. As a proof of concept for X-ray imaging a test object, fitted with different materials, and imaged by 5-80 keV X-rays. This is illustrated in Figure 7. The materials, from clockwise, are adhesive putty, indium, lead, tin, and indium. The image produced, right, indicates the ability of the detector to distinguish between different materials thereby functioning as an imaging device.

Elsewhere, C. Erd et al [63] developed a spectroscopic X-ray imaging device based on epitaxially grown GaAs. The prototype array was fabricated by growing a 325 μm epitaxial intrinsic GaAs layer onto a 200 μm n+ substrate, topped with a 6 μm p+ layer, completing the PIN structure. The 1.1 cm^2 surface area was etched into a grid of 32x32 pixels. Optimal operating conditions for the reverse bias, 60 V, were established and the energy resolu-

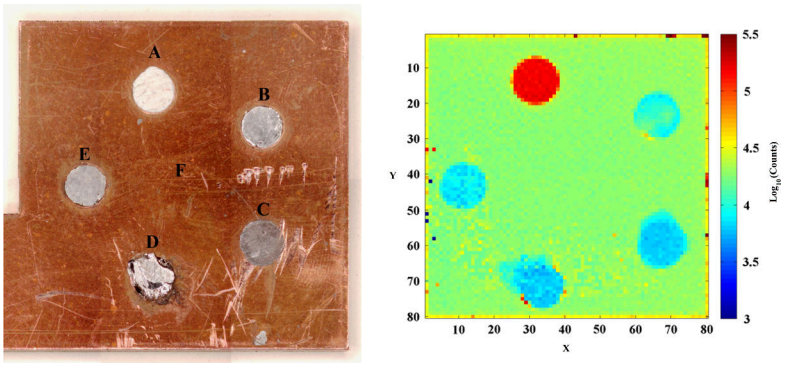

Figure 7: A proof of concept demonstration of X-ray imaging with a GaAs sensor [51].

tion of individual pixels were found to range from approximately 290 eV for a 5.9 keV beam and 780 eV for a 100 keV beam. These results were obtained at room temperature, and the investigations into variation of resolution with temperature found a 10% gain in resolution as the device was cooled to 5 °C with negligible improvements at cooler temperatures.

The results found here can be contrasted with the previously cited study into chromium compensated GaAs. C. Erd et al's device was anticipated to achieve a resolution of 0.5 keV for the 60 keV photopeak on an ^{241}Am source at room temperature. The same photopeak has a resolution of 2.9 keV for the GaAs:Cr detector. The GaAs:Cr detector has a thicker intrinsic layer, which should contribute to better energy resolution in principle, however this device was not based on epitaxially grown GaAs, as was the case with C. Erd's device. Veale et al selected the LEC growth technique in order to grow a thick material bulk whereas the MBE technique allows for the precise and orderly growth of crystal layers, at lower temperatures than LEC, reducing the risk of defect inducing effects like interdiffusion [64]. Deficiencies in material quality can give rise to charge collection inefficiencies which introduce noise to the detriment of energy resolution. Furthermore, C. Erd et al developed and tested low noise pre-amplifier designs which may have played an additional role in the superior energy resolution of their device. C. Erd, et al also found the FWHM of a 5.9 keV photopeak, 0.26 keV, which can be contrasted with a more recent study by G. Lioliou et al [54]. They conducted a comprehensive characterisation of MOVCD GaAs photodiodes, fabricating 200 μm and 400 μm diameter device with 10 μm thick intrinsic layers. The detectors have energy resolutions of 0.69 keV and 0.73 keV respectively. The

authors suggest their relatively thin device suffers primarily from the effects dielectric noise in addition to white series noise and Fano noise. These factors are all correlated with the capacitance of the device, which is reduced in thicker devices. This can give rise to a number of benefits including increased quantum efficiency and lower pulse shaping times [10]. Future work will see the authors reconsider the design of the pre-amplifier by combining the photodiode and the junction gate field-effect transistor (JFET) into the same substrate with the aim of reducing dielectric noise. Additionally, further refinements in device passivation may reduce surface leakage current and bring the energy resolution performance closer to the previously discussed devices.

There are many other studies investigating the burgeoning field of X-ray spectroscopy by GaAs photodioes and it can be concluded that these devices have potential in this field [65, 66]. These studies have validated some of the advantages of using GaAs including radiation hardness, energy resolution and room temperature operation. In addition to giving insight into suitable fabrication techniques, I-V characteristics and potential readout electronics. However, there have been few documentations of their application to beta radiation.

Barnett, Lees, and Bassford attempted direct detection of ^{3}H and ^{14}C beta particles with GaAs photodiodes [67]. Their detectors were grown by MBE and photolithographically etched into 200 μm diameter diodes with 2 μm thick intrinsic layers. Beta propagation through the device was simulated with the Monte Carlo software, CASINO [68]. Particles ranging in energy from 1 keV to 156.48 keV were simulated, investigating their penetration depth and deposition of energy within the detector. Figure 8 shows the results of the simulation of 156.48 keV electrons. It can be observed that the thin intrinsic layer of this detector is not sufficient to stop incoming radiation of this energy, and the electrons penetrate into the substrate layer of the diode. This indicates that a much thicker intrinsic layer would be required for beta particles approaching the energy of those released by ^{90}Sr and ^{90}Y . Other results from this study indicate that the limiting factor on the detection of low energy beta particles, less than 5 keV, is the p-type layer on the surface of the detector. This region attenuates the particles sufficiently such that the maximum proportion of their energy deposited in the intrinsic layer is only 50%. The detector was used to capture ^{3}H and ^{14}C spectra which, after calibration, showed accordance with accepted spectra for these nuclides. The results presented here are promising for the potential of GaAs detectors, although evidently the intrinsic layer is likely much too thin for

efficient ^{90}Sr detection.

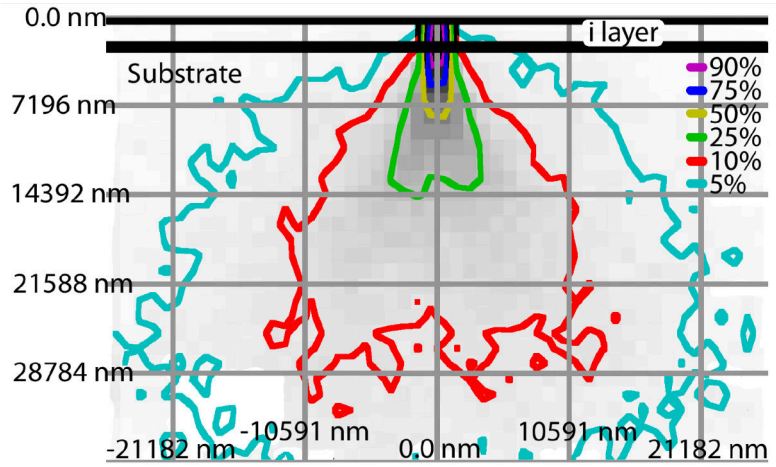

Figure 8: The percentage of initial energy, 156.48 keV, of beta particles deposited in a GaAs detector [67]

Lioliou and Barnett [69] characterised GaAs p^+-i-n^+ mesa photodiodes to assess their potential as low energy beta spectrometers with a view to using them in applications for space plasma physics. GaAs photodiodes were fabricated for this study at the EPSRC National Centre for III-V Technologies, Sheffield, with a 10 μm undoped GaAs intrinsic layer sandwiched between a 0.50 μm thick GaAs p^+ layer and a 1 μm n^+ layer. Reportedly the thickest X-ray mesa photodiodes produced to date, the wafers used for beta spectroscopy had a 200 μm diameter. Initially, the detectors were simulated with the Monte Carlo simulation software, CASINO. A point source of 4,000 electrons, varying from 1 keV to 66 keV, were fired at the detector surface and the depth of their penetration is summarised in Figure 9. Simulations were run with and without the presence of the Ohmic contact required on the detector, which covered 45% of the detector's surface. The simulation predicted a maximum external quantum efficiency of 49% from a 66 keV source, with the major limiting factors being the absorption of electrons in the top layers of the diode and the Ohmic contacts.

Following on from the simulation, a real-world validation was carried out. A ^{63}Ni source was placed 5 mm above the surface of the GaAs photodiode, which was operating under a 10 V reverse bias. After a counting time of 400 s the collected beta spectrum was compared with the accepted spectrum

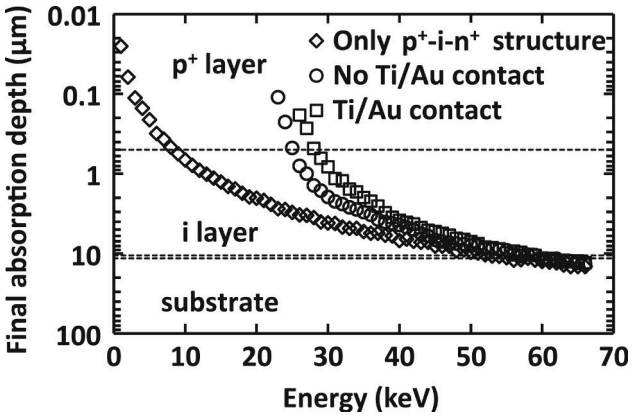

Figure 9: The simulated absorption depth of electrons in a GaAs p^+-i-n^+ detector as a function of their energy [55]

of a ^{63}Ni beta source, normalised to reflect experimental conditions. The maximum energy observed in the intrinsic region was approximately 50 keV, suggesting that maximum energy particles were losing 16 keV. This was a 9 keV difference from the maximum energy predicted in the simulation. This discrepancy was attributed to the absorption of energy in insensitive sections of the detector, including the p^+ layer in the device, the nickel protective layer around the source and the air gap between source and detector. While these results were promising for the potential of GaAs photodiodes as electron spectrometers, they were conducted with a device designed for X-ray rather than beta-spectrometry in mind. The response of the device to much higher energy electrons remains to be seen, whether they are stopped by the intrinsic layer in sufficient numbers to produce a clear signal. It is likely that detection of ^{90}Sr with GaAs photodiodes will require devices thicker than the 10 μm devices which have been tested in these studies. The photodiodes fabricated for X-ray detection have demonstrated that it is possible to produce thicker detectors and by producing a chromium doped or epitaxially grown device can address the defects which have previously hindered the development of GaAs devices.

5. Monte-Carlo simulations

Reviewing the literature has highlighted some tentative studies applying GaAs photodiodes as beta radiation detectors, however, these have only been applied to low energy beta emitters. The devices used have relatively thin

intrinsic layers, particularly in comparison with some of the detectors being developed for X-ray detection. A proof of concept simulation was developed to investigate the potential for GaAs as a beta detector in a groundwater borehole scenario, for energies on the scale of ^{90}Sr and ^{90}Y decay. The physics simulation package, Geant4, was used to construct a basic model of a GaAs detector and simulate its interaction with beta-particles. Geant4 is a Monte Carlo simulation based software and is written in the object-oriented programming language C++ [70]. Step by step the software tracks the path of radiation as it travels through matter. At each step the probability of interaction and random number generation predict the next step along the particle's track. The exact nature of the physics processes invoked in the simulation and their cross-sections are defined by the "Physics List" selected for the simulation. Geant4 includes a number of reference Physics Lists and this simulation used the FTFP_BERT list, the Geant4 default which is valid for electrons up to 100 TeV.

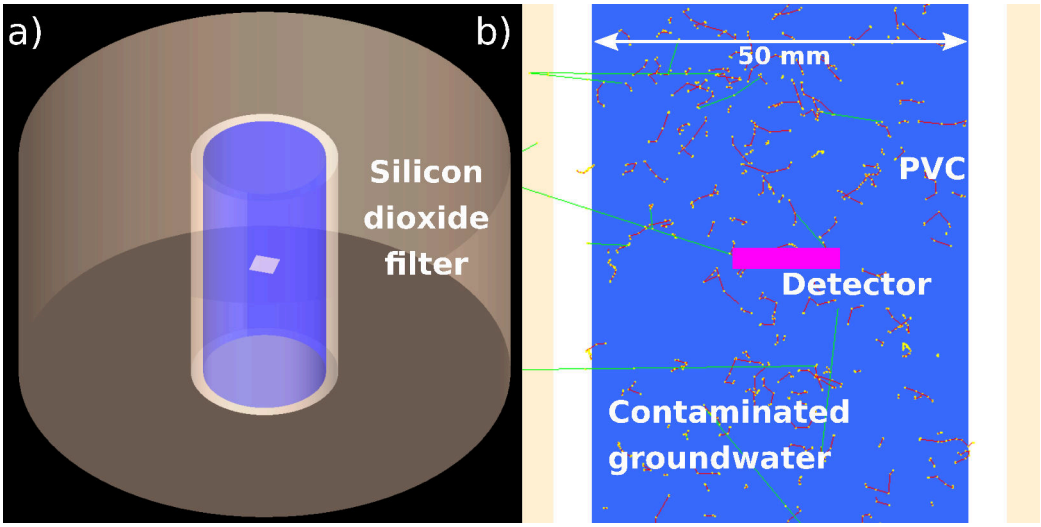

Figure 10: An overall visualisation of the Geant4 simulation can be seen in image a). Image b) is a cross section of the well, filled with ^{90}Sr contaminated groundwater. As nuclei decay beta particles are released and traverse the water in erratic paths. They are either fully absorbed, deflected, or release bremsstrahlung radiation, the long straight lines. Bright dots mark steps in the particle's path.

The proof of concept detector was based on a device created by C. Erd et al [63]. This design was selected due to the relatively thick intrinsic layer, especially in comparison with previous detectors applied to beta radiation.

Matching the thickness of the intrinsic layer, 0.325 mm, and surface area, 1.1 cm^2, a GaAs detector was created in Geant4 code and placed in a modelled groundwater borehole. Figure 10a visualises the borehole with the detector submerged in groundwater. A cross section of the scenario is seen in Figure 10b. Here, decaying ^{90}Sr particles are randomly dispersed throughout the groundwater. As ^{90}Sr decays, electrons, the short erratic trajectories, are released and tracked as they travel through space. Bright dots mark steps along the particle's trajectory before it is fully absorbed by the environment or the detector. As the particles interact the detector they are either backscattered, pass through the detector while only depositing a fraction of their energy, or fully absorbed within the intrinsic layer of the detector. Their energy and path are recorded, along with the number of counts in the detector for the entire run. The long and straight particle lines shown in this image represent photons, likely the result of the Bremsstrahlung effect. The anti-neutrinos released during beta decay are hidden for visual clarity.

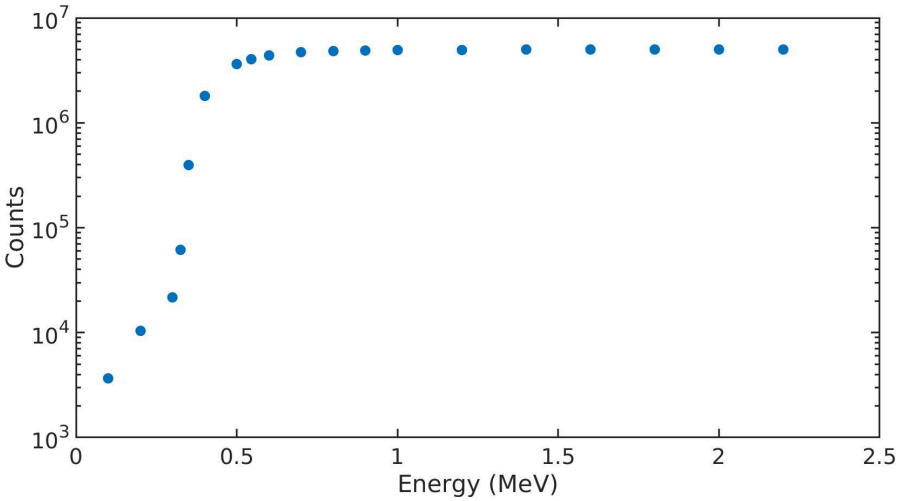

Figure 11: The number of counts recorded in the detector for increasing beams of electron energy. Errors range from ± 35 to $\pm 1.290 \times 10^3$ and as such are not clearly visible on the graph.

The initial simulation examined whether the detector had sufficient stopping power to detect electrons emitted during ^{90}Sr and ^{90}Y decay. A particle gun was positioned 0.1 mm from the surface of the detector and the energy of the beam was increased from 0.1 MeV to 2.2 MeV. Each run consisted of

5×10^6 electrons and the results can be seen in Figure 11. Fewer counts are observed at lower energies, attributed to the increased likelihood of low energy electrons be absorbed before detection and to backscatter on the surface of the detector. As the energy increases to 1.2 MeV, nearly the entire run of electrons, 99% on average, deposit energy in the detector and are counted. However, this does not account for how much energy is being deposited by each particle. Some electrons are backscattered, leaving only a fraction of their energy, or simply pass through the detector. When designing the photodiode, it will be of utmost importance to ensure the intrinsic layer is thick enough to fully capture the energy of particles released by the radionuclides of interest, thereby being capable of fully recording their beta spectra.

The second simulation investigated how the intensity of the radiation detected varied with increasing distance from the source. A 0.546 MeV beam of electrons was fired at the detector from 0.1 cm away increasing to 8.5 cm. Figure 12 displays the results. The number of counts observed by the detector decays exponentially with increasing distance. Particles released within few millimetres from the detector's surface, dropping to tens of counts at a range of 8 cm. This detector aims to be used in situ, and will have to operate at some distance from the source, so the detectable range of the detector is a key characterisation.

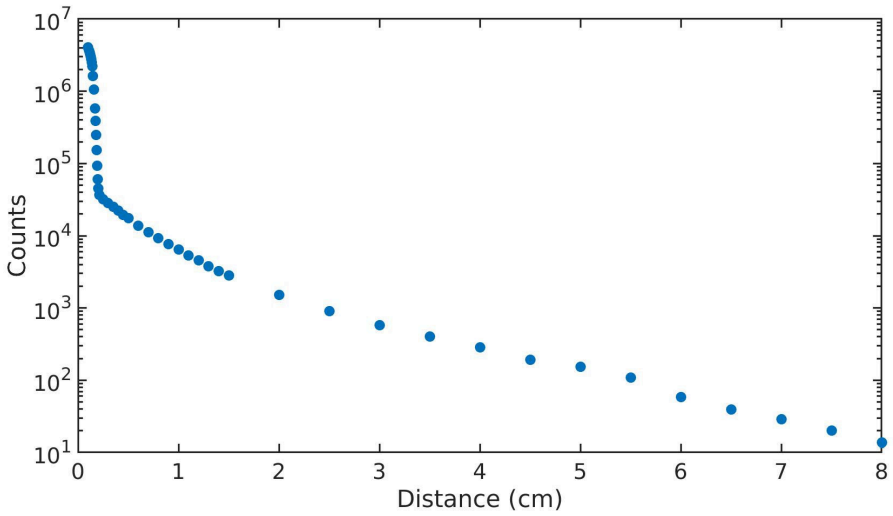

Figure 12: The number of counts detected as the detector is moved further away from a 0.546 MeV electron beam source.

As an exhibition of the potential application of the detector, the device was used to collect a spectrum of radiation from a contaminated groundwater source. A 5 cm deep cylinder of water was randomly filled with decaying ^{90}Sr particles. A full decay chain was realised for each ^{90}Sr particle, resulting in ^{90}Y production and decay to stable Zr. The simulation ran for 1×10^7 decay events. The spectrum of beta radiation captured in the detector is seen in Figure 13. The first peak is largely comprised of beta particles released during ^{90}Sr decay, and the lower second peak is indicative of ^{90}Y decay, which tails off at a much higher energy, approximately 1.8 MeV. It should be noted that the particles generated during this simulation must travel some distance before reaching the detector, and as such will have already lost some of their kinetic energy to the surrounding environment.

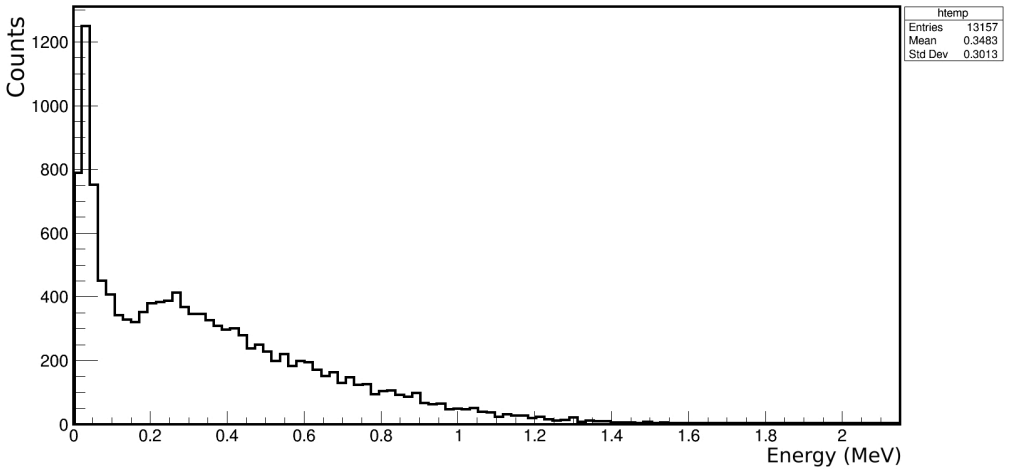

Figure 13: The spectra of beta particle energy recorded in the detector after 1×10^7 ^{90}Sr decay events were simulated.

The simulation results presented here are encouraging for the potential of GaAs photodiodes as in situ detectors for the radiometric assay of ^{90}Sr . It has been demonstrated that a detector of real-world proportions can be successfully detect beta particles of the energy scale required in a situation modelled on a real world scenario. This suggests there is potential to use GaAs photodiodes in the development of an in situ beta detector.

6. Conclusions

This paper has presented a review of existing methods for the radiometric analysis of ^{90}Sr in the environment and their suitability, or lack thereof, for in situ detection has been examined. Nuclear decommissioning sites have a demand for real-time, in situ, monitoring of radionuclides in groundwater to improve their response to fluctuations in groundwater activity and to further evaluate waste management. Current techniques are lab-based, time consuming and expensive. While there have been attempts to reduce the time-scales involved in these procedures, and create more mobile detectors, these only go part way to addressing the practical difficulties associated with these techniques. A novel approach based on the direct detection of beta radiation has been proposed. GaAs photodiodes were examined for their ability to directly detect ionising radiation, and their suitability for beta radiation was validated through simulation.

The research presented here has suggested that GaAs is indeed a strong candidate for an in situ beta detector. The wide bandgap of the material means it can eschew the cooling requirements of other semiconductors, reducing the size and weight of the detector. Meanwhile its radiation hardness suggests GaAs devices are well suited for operation at sites of nuclear waste and spillage. Development of such a device will enable real-time counting of beta radiation in difficult to reach areas, such as groundwater boreholes, reducing risk of exposure to workers.

Initial Geant4 simulations have demonstrated that GaAs has right physical properties to detect beta radiation. The fabrication technique selected can have an influence on the defects present in the device and its operating characteristics, energy resolution and efficiency. Photodiode junction layers and electronics readout systems will also influence the energy deposition of radiation in the detector.

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