A comparison of the ZnS(Ag) scintillation detector to the silicon semiconductor detector for quantification of alpha radioactivity in aqueous solutions

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A COMPARISON OF THE ZnS(Ag) SCINTILLATION DETECTOR TO THE SILICON SEMICONDUCTOR DETECTOR FOR QUANTIFICATION OF ALPHA RADIOACTIVITY IN AQUEOUS SOLUTIONS

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ABSTRACT

A ZnS(Ag) detector was successfully used in the direct detection of alpha particles from aqueous solutions and the results were compared to the passivated ion implanted planar silicon (PIPS) continuous air monitor (CAM) detector. The ZnS(Ag) detector is recommended for on-line detection of gross alpha radioactivity from high-level liquid waste process streams; however, the detector suffers from limitations due to variations in detection efficiency with alpha energies. The beta and gamma interference did not significantly spillover into the alpha region of interest of the ZnS(Ag) detector which provided absolute detection efficiencies of $7.19 \pm 0.13\%$, $5.37 \pm 0.02\%$, and $4.21 \pm 0.03\%$ for $^{244}$Cm, $^{239}$Pu, and $^{234}$U/$^{238}$U solutions, respectively. The detection efficiencies were found with a $3.5 \text{ mg/cm}^2$ ZnS(Ag) scintillation layer, $4.15 \text{ cm}^2$ detection surface, and a $5.2 \text{ mm}$ air gap between the liquid surface and the detector. The absolute detection efficiencies of the PIPS CAM detector for $^{244}$Cm, $^{241}$Am, $^{239}$Pu, and $^{230}$Th solutions in the presence of beta and gamma activity were $4.61 \pm 0.01\%$, $3.87 \pm 0.01\%$, $2.84 \pm 0.01\%$, and $1.32 \pm 0.01\%$, respectively, for a geometrical set-up similar to the ZnS(Ag) experiments. Two ZnS(Ag) scintillator thicknesses were compared. The thicker scintillation layer suffered from light absorption and scattering in the ZnS(Ag) layer resulting in a lower energy signal. Despite the differences, the absolute detection efficiencies of the two thicknesses were equivalent. The large detection area available to the ZnS(Ag) detector proved to be valuable for the detection of radioactivity in liquids as the absolute detection efficiency more than doubled for a surface area of $78.5 \text{ cm}^2$ and resulted in a minimum detection concentration of $0.32 \text{ Bq/mL}$ for $^{238}$U solution for a
3600 second count. The detection capabilities and pulse height spectra of the ZnS(Ag) detector were compared to the PIPS CAM detector as well as computer simulations and theory.
DEDICATION

In dedication to my father, who has always been my role model in the sciences.
ACKNOWLEDGEMENTS

I would like to thank Dr. Timothy DeVol for his guidance throughout my research project and also for the excellent opportunity to work under him. Mr. Matthew O’Hara served as my mentor in many safety and laboratory practices and took time out of his busy day to guide me as I acquired data and conducted experiments. I would like to thank Dr. Oleg Egorov for his direction, ideas, and the opportunity to work at the Pacific Northwest National Laboratory. I am grateful for the electrical engineering assistance which Mr. Joel Simoneau was kind enough to grant. I appreciate both Dr. Robert Fjeld and Dr. John Coates for being willing and helpful participants on my committee.

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**B.18** The pulse height spectra of the 7151.7 Bq $^{241}$Am EPS on top of the Plexiglas shield with and without the $7.4 \times 10^8$ Bq $^{90}$Sr($^{90}$Y) source detected with the ZnS(Ag) detector and charge-sensitive preamplifier.

**B.19** The electronic signal for the ZnS(Ag) detector, amplifier, and (a) signal splitter and (b) the preamplifier. The 7151.7 Bq $^{241}$Am electroplated standard was set on top of the 0.635 cm Plexiglas sheet placed above a $7.4 \times 10^8$ Bq $^{90}$Sr($^{90}$Y) source.

**C.1** The moving spherical coordinate system used in the computer simulations (Relative thicknesses were not drawn to scale).

**C.2** The Microsoft Excel worksheet referred to by the Visual Basic source codes.
LIST OF SYMBOLS AND ABBREVIATIONS

Greek Symbols

Alpha particle: \( \alpha \)
Beta particle: \( \beta \)
Delta (change of): \( \Delta \)
Gamma ray: \( \gamma \)
Micrometer: \( \mu m \)
Pi: \( \pi \)
Counting Sensitivity: \( \varepsilon \)

Abbreviations

SRS: Savannah River Site
ZnS(Ag): Silver doped Zinc Sulfide
PIPS: Passivated Ion implanted Planar Silicon
CAM: Continuous Air Monitor
HLW: High-level Waste
PUREX: Plutonium and Uranium Recovery by Extraction
PAM: Personal Alpha Monitors
PNNL: Pacific Northwest National Laboratory
EPS: Electroplated Standard
WAC: Waste Acceptance Criteria
PVC: Polyvinylchloride
FWHM: Full Width Half Maximum
List of Symbols and Abbreviations (Continued)

SRIM: Stopping and Range of Ions in Matter
TRIM: The Transport of Ions in Matter
GEANT4: Geometry and Tracking: A toolkit for simulation of the passage of particles through matter.
MCNPX: Monte Carlo N-Particle Transport Code
AASI: Advanced Alpha- spectrometric Simulation
LLD: Lower Level Discriminator
FWTM: Full Width Tenth Maximum
ULD: Upper Level Discriminator
CERN: Conseil European pour la Recherche Nucleaire: European Laboratory for Particle Physics
NIST: National Institute of Standards and Technology
ASTAR: A Stopping power And Range table for helium atoms
MDA: Minimum Detectable Activity
MDC: Minimum Detectable Concentration
CHAPTER 1

INTRODUCTION

There is a need for the direct detection of alpha radioactivity in aqueous solutions. There are various detectors which detect alpha particles; however, the short range of alpha particles originating from a solid or liquid matrix makes detection in these media problematic. Actinides are present in high-level waste (HLW) within the Department of Energy complex at concentrations of approximately $8 \times 10^4$ Bq/mL for the HLW supernate and $8 \times 10^8$ Bq/g for HLW sludge. The total alpha activity of the liquid waste is quantified and monitored during the chemical processing of the HLW. At the Savannah River Site (SRS), the concentration of the treated HLW supernate can not exceed 970 Bq/mL in order to be accepted for grouting at the Saltstone Facility [1].

Currently, several methods are used at SRS to measure the alpha activity in tank waste supernate and processed waste. Gross alpha counting is conducted by depositing a small aliquot of waste onto a planchet. After evaporation, the alpha activity is quantified with a gas-flow proportional counter. The detection of alpha particles is hindered by the high salt content of the sample which introduces uncertainty into the measurements. SRS also periodically uses radiochemical separation techniques to isolate the alpha activity from the waste matrix prior to quantification by either gas flow proportional counters or silicon detectors. Inductively coupled plasma mass spectroscopy is also used to identify the contents of tank waste; however, the mass spectrometer results are sometimes complicated with isobaric interferences [1].
On-line flow-through alpha radiation detectors are needed at HLW processing facilities along side beta and gamma detectors for continuous monitoring. An on-line alpha detector on a process stream would be able to monitor the solution continuously and nondestructively. Beta and gamma ray radiation are typically easier to detect from waste due to the longer range and mean free path, respectively, in process solutions. The ZnS(Ag) detector would simplify the detection of alpha particles from aqueous solutions if the alpha particles are efficiently detected from the surface of the liquid. An efficient on-line method of total alpha detection would be useful and practical for chemical processing applications and numerous other purposes.
CHAPTER 2
LITERATURE REVIEW

Defense Waste

In the 1940’s, the United States embarked on a series of military and civilian activities which initiated the accumulation of radioactive wastes. The activities began in earnest with the Manhattan Project and continue today through the use of nuclear power in the generation of electricity. Various forms of radioactive wastes are produced through different kinds of activities. Civilian waste consists mainly of spent fuel from reactors and low-level wastes. Wastes stockpiled from the national defense taskforce consist of irradiated target materials, spent fuel, transuranic wastes, low-level wastes, and HLW. In comparison, research laboratories and the nuclear power industry (exclusive of nuclear power reactor stations) generate mostly low-level wastes and only produce high-level waste in special cases where nuclear fuel or fissile material is handled for research [2].

Out of the nation’s radioactive waste, HLW is the top concern as it contains the bulk of the radioactivity found in nuclear waste. HLW is defined as the highly radioactive material which typically contains significant concentrations of fission products and is the consequence of reprocessing spent fuel [2]. Tritium and plutonium, the materials required for nuclear warheads, were extracted by means of reprocessing irradiated targets and nuclear fuel rods from the United States’ original fourteen materials production nuclear reactors. The first materials production reactor went critical in 1944 and the last materials production reactor was shut down in 1988 [3].
High-level waste is a byproduct of plutonium and uranium recovery by extraction (PUREX) process. The PUREX process is the leading method of reprocessing and was widely used by the defense department at both the Hanford Site and SRS. The process requires that the irradiated target be chopped and dissolved in nitric acid. The heavy elements went into solution while the cladding remained whole. The nitric acid solutions were processed through solvent extraction which separated the uranium and plutonium from the fission products and other transuranic elements. The resultant fission products and transuranic elements are discarded as HLW [2]. The fission products are primarily responsible for the high activity and external dose rate of the waste while the high-activity transuranic elements generate considerable thermal energy [3].

After nuclear warfare production declined, the demand for plutonium and highly enriched uranium decreased and the pace for reprocessing was reduced. The Department of Defense phased out reprocessing and the production of nuclear weapons and eventually stopped work in 1992 [3]. The Department of Energy, however, left an accumulation of HLW, typically stored in large tanks, for the nation to manage. The original tanks located on the Hanford Site corroded as the single carbon steel walls were only designed for a useful lifetime of 25 years. Sixty seven Hanford tanks leaked HLW into the environment causing contamination issues. The three largest leaks topped the chart at 115,000, 70,000, and 55,000 gallons of HLW liquid into the ground [3]. Twenty eight new double-walled carbon steel tanks were constructed in 1980 to rectify the problem. The waste was transferred to the safer containers which were designed to hold the waste for fifty more years. The Department of Energy is storing approximately 100 million gallons of HLW in 243 underground tanks [3].
The Hanford Site and SRS are currently assessing the existing method of converting HLW into a more stable form. The sludge, liquid, and solid components of the aged, HLW on these sites are mixed with molten glass and poured into metal cylinders. This process is called vitrification, and with the waste in the vitrified, glass form, it is more stable and less likely to contaminate the environment. The Department of Energy has already begun vitrification at a plant at SRS and by 2005, 1.64 million gallons of high-level sludge were successfully vitrified [4]. After vitrification, the canisters were placed in a specially designed storage facility. Vitrified waste costs less to store and monitor compared to liquid waste and decreases the threat to security. The canisters will be kept in the storage facilities until a geologic repository is open then they will be moved to the repository permanently [3].

**Alpha Particle Theory**

Alpha particles are heavy, charged particles which are typically emitted from disintegrating, heavy nuclei. The alpha particle is comprised of two protons and two neutrons and is equivalent to a doubly ionized helium atom, as it lacks electrons. The alpha particle is the least penetrating form of radiation due to its large size and positive charge [5]. The alpha particle interacts with matter through attraction with the orbital electrons of the absorber atoms via the Coulomb force. The impulse caused by the passing alpha particle is enough to excite the electron or to ionize the absorber atom—the primary means of energy transfer. The interaction between the alpha particle and the nucleus of the absorber atom via Rutherford scattering is infrequent and typically not significant in the detection of alpha particles. The kinetic energy of the alpha particle decreases after each interaction which reduces the particle velocity. The alpha particle
loses approximately one five hundredth of its total energy after every interaction [6]. The alpha particle interacts continuously in the absorber material thus losing energy linearly, consequently slowing the particle until it stops [6].

The alpha particle intensity passing through an absorber does not decrease until near the end of the range as shown in Figure 2.1. The energy of the alpha particles decreases with increasing absorber thickness; however, the number of alpha particles detected does not decrease until the approximate range is reached and the alpha particles begin to terminate [7]. The alpha particle range, the distance which particles penetrate, is much shorter than that of beta particles or the mean free path for gamma rays. The mean range, the most common reference, refers to the absorber thickness which reduces the alpha count to half. The extrapolated range is obtained by extrapolating the alpha absorption curve in Figure 2.1 to zero transmission and represents the largest possible range [7]. The mean range of alpha particles from common alpha emitting radionuclides is only a few centimeters through air. The range of alpha particles through liquids and solids is significantly shorter than the range in air due to the high density of the media [6]. The energy-range relationship of alpha particles through air is estimated as

\[ R(cm) = 0.56E, \quad E < 4\ MeV \]
\[ = 1.24E - 2.62, \quad 4\ MeV < E < 8\ MeV \]

at 1 atm and at 15°C [6]. The alpha particle energy-range relationship through other media, the Bragg-Kleeman Rule, is found through the relationship

\[ \frac{R_1}{R_0} = \frac{\rho_1\sqrt{A_1}}{\rho_0\sqrt{A_0}} \]
where $R$ is the range, $\rho$ is the density of the medium, and $A$ is the atomic mass number of the medium [7]. If the medium is a mixture of elements, the square root of the effective atomic mass number is

$$\sqrt{A} = \frac{n_1A_1 + n_2A_2 + n_3A_3 + \ldots}{n_1\sqrt{A_1} + n_2\sqrt{A_2} + n_3\sqrt{A_3} + \ldots}$$  \hspace{1cm} (3)$$

where $n$ is the atomic fraction of the elements and $A$ is the atomic mass number of each element [5]. The square root of the effective atomic mass number of air is 3.82 and the density of air is $1.226 \times 10^{-3}$ g/cm$^3$ at 1 atm and at 15°C [5].

![Figure 2.1](image)

**Figure 2.1** The alpha particle absorption curve demonstrates the consistent count rate and alpha range [5].

The alpha particle tracks through matter are linear as they are not easily deflected due to their large momentum. The Coulomb force is not significant in changing the path of an alpha particle because the alpha particles undergo interactions from all directions as they travel through matter. Deflections in the alpha particle track do not usually occur until the end of the range when the alpha particles have decreased energy [6]. Alpha particles are mostly affected by straggling, the fluctuation in path length of particles with the same initial velocity, towards the end of the range. The alpha particle energy loss is a stochastic process and an individual alpha particle range varies by a few percentage points of the mean range [7].
The ionization of air requires roughly 35 eV per ion pair produced, where the ion pair is defined as the positive ion and free electron. The exact value for ionization depends on the velocity of the alpha particle though the medium. Approximately half of the alpha particle energy lost in air is due to electron excitation as opposed to ionization [5]. The ionization potentials for O₂ and N₂ are 13.6 eV and 14.5 eV, respectively. The specific ionization along the alpha particle path is defined as the number of ion pairs per unit path length, which equals approximately 2,000 to 6,000 ion pairs per millimeter of air. Alpha particles ionize the air most heavily near the end of their range as shown in the Bragg Ionization curve, Figure 2.2, which establishes the relationship between the average specific ionization and the distance from the source [5].

![Figure 2.2](image)

**Figure 2.2** The Bragg ionization curve for Polonium alpha particles [5].

The average linear rate of the energy loss of a heavy, charged particle is called the stopping power of a medium, S.

\[
S = -\frac{dE}{dx}
\]

Hans Bethe improved upon Bohr’s classical equation for the average linear rate of energy loss by incorporating quantum mechanical theory which allowed the equation to be useful for all heavy, charged particles [8].
\[-\frac{dE}{dx} = 4\pi k_0^2 z^2 e^4 n \frac{mc^2 \beta^2}{I(1 - \beta^2)} \left[ \ln \frac{2mc^2 \beta^2}{I(1 - \beta^2)} - \beta^2 \right] \tag{5}\]

where:

\( k_0 = 8.99 \times 10^9 \text{ N m}^2 \text{ C}^{-2} \)
\( z = \text{atomic number of heavy particle} \)
\( e = \text{magnitude of the electron charge} \)
\( n = \text{number of electrons per unit volume in the medium (e\(^{-}/m^3 \))} \)
\( m = \text{electron rest mass} \)
\( c = \text{speed of light in vacuum} \)
\( \beta = v/c = \text{speed of the particle, v, relative to c} \)
\( I = \text{mean excitation energy of the medium} \)

The number of electrons per unit volume in the medium is

\[ n = \frac{6.022 \times 10^{23}}{\text{molecules/moles}} \rho \left( \frac{\text{g}}{\text{m}^3} \right) Z \left( \frac{\text{e}^-}{\text{molecule}} \right) A \left( \frac{\text{g}}{\text{mole}} \right) \tag{6} \]

where \( \rho \) is the density of the medium and the atomic number, \( Z \), represents the number of electrons in the medium. The mean excitation energies, \( I \), are approximated from previous experiments for elements of atomic number, \( Z \), using empirical formulas in equation 7 [8].

\[ I \begin{cases} 19.0 \text{ eV}, Z = 1 \\ 11.2 + 11.7Z \text{ eV}, 2 \leq Z \leq 13 \\ 52.8 + 8.71Z \text{ eV}, Z > 13 \end{cases} \tag{7} \]

Equation 5 is simplified greatly by the incorporation of constants into the equation. The atomic number of the particle is defined as 2, which specializes the equation for alpha particles in equation 8 [8].

\[-\frac{dE}{dx} = \frac{2.032 \times 10^{-30}}{\beta^2} n \left[ \ln \frac{1.02 \times 10^6 \beta^2}{I(1 - \beta^2)} - \beta^2 \right] \text{MeV cm}^{-1} \tag{8} \]
The stopping power of a medium for an alpha particle depends greatly on the energy or velocity of the alpha particle which dictates the only remaining variable, $\beta$. The stopping power of water in MeV/cm for several particles at various energies is shown in Figure 2.3 [8].

![Figure 2.3](image)

**Figure 2.3** The stopping power of water [8].

**Scintillation**

The energy states are determined by the crystalline lattice of an inorganic material thus influencing the scintillation mechanism. The lowest band of the valence states represents electrons which are bound to the lattice sites. The conduction band contains higher energy electrons which are free to migrate throughout the crystal. The intermediate region between the valence and the conduction bands, where electrons do
not normally reside, is called the forbidden band. When energy is absorbed from an alpha particle, an electron is elevated from the valence band into the conduction band thus leaving a hole in the valence band. If scintillation is favorable then an electron drops from the conduction band into the valence band resulting in the emission of light or scintillation photons. An activator is used to dope the pure crystal and to modify the energy band structures as shown in Figure 2.4. The activator creates energy states in the forbidden band of the crystalline states of the host crystal which allow electrons to de-excite with photons in the visible range of the spectrum [6].

![Energy Band Structure](image)

**Figure 2.4** The energy band structure of an activated crystalline scintillator [6].

**Zinc Sulfide**

Zinc sulfide, ZnS, and silver activated zinc sulfide, ZnS(Ag), are common detectors of alpha particles and are among the oldest inorganic scintillators. The ZnS screen utilized in the famous Rutherford experiment was used to prove the existence of the atomic nucleus. The scintillation screen allowed for the locations of the alpha particles to be recorded after interaction with the gold foil. The majority of Personal Alpha Monitors (PAM) which are used in laboratories for the detection of alpha contamination are constructed of ZnS(Ag) due to its potential large area and low cost. ZnS is also commonly used in its un-doped form in “glow-in-the-dark” paints and products due to its lengthy phosphorescence.
ZnS(Ag) detectors are convenient to use in applications where gross counting is involved due to the portability of the scintillation detector. The ZnS(Ag) scintillation crystals are either sprayed onto the light guide, attached with double-sided tape, or embedded into an epoxy. A photomultiplier tube converts the light from the ZnS(Ag) into an electronic signal which can be counted with appropriate electronic devices. A thin layer of Mylar® covers the scintillation layer to create a light-tight atmosphere; although, the Mylar® layer is delicate and easily punctured or scratched [9].

ZnS(Ag) is only available as a polycrystalline powder and is therefore primarily limited to thin screens. Thicknesses ranging between 3.5 mg/cm² and 25 mg/cm² are recommended by the Ludlum manufacturers. The ZnS(Ag) layer is unusable at thicknesses greater than 25 mg/cm² due to the opacity of the layer to its own luminescence [6]. At greater thicknesses, ZnS(Ag) crystals are opaque due to the absorption of light into the material and from light scattering. The extinction coefficient, a measure of how well a substance absorbs electromagnetic radiation at a given wavelength, is best used to describe the absorption of the scintillated light. The intensity of the electromagnetic radiation is dampened as the light propagates through the material. The light intensity after transition through a material of z thickness is defined as I,

\[ I(z) = I_0 e^{-\frac{2\pi k z}{c}} \]  

(9)

where k is the extinction coefficient, \( \omega \) is the angular frequency of the electromagnetic wave, and c is the speed of light. The extinction coefficient can be determined from Figure 2.5, a graph of ZnS extinction coefficients at various photon energies [10].
Scintillation decay times for ZnS(Ag) are reported between several hundred nanoseconds and 10 µs with an accepted decay constant of 110 ns [10]. The decay time recorded in Knoll is 0.2 µs or 200 ns; although phosphorescence of a longer duration was noted in ZnS(Ag) scintillation [6]. ZnS(Ag) possesses several other physical properties listed in Table 2.1: the specific gravity of the scintillator, the wavelength of the emission [10], and the refractive index [6].
Table 2.1 Common properties of ZnS(Ag) scintillator [6].

<table>
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<th>Properties of ZnS(Ag) Scintillator</th>
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<tr>
<td>Specific Gravity</td>
<td>4.09</td>
</tr>
<tr>
<td>Emission Wavelength (nm)</td>
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<tr>
<td>Peak</td>
<td>450</td>
</tr>
<tr>
<td>Minimum</td>
<td>330</td>
</tr>
<tr>
<td>Refractive Index</td>
<td>2.36</td>
</tr>
</tbody>
</table>

A pulse height spectrum for the detection of 5.5 MeV alpha particles is shown in Figure 2.6. ZnS(Ag) is primarily used for the detection of alpha particles; however, thermal neutrons are detected by incorporating $^6$Li into the scintillation material, also shown in Figure 2.6. Fast neutrons are detected with ZnS(Ag) when the scintillation powder is embedded into a hydrogenous compound. The ZnS(Ag) detects the recoiling proton from a neutron-proton scattering interaction [10].

![Graph](image_url)

Figure 2.6 The ZnS(Ag) pulse height spectra for 5.5 MeV alpha particles and thermal neutrons [10].
**Alpha Radiation Monitoring Detector**

A flow-through zinc sulfide phosphor scintillation detector which quantified alpha radioactivity in low-level radioactive liquids was utilized in 1973 by M.M. Chiles at Oak Ridge National Laboratory. The scintillation detector was coupled to an alarm, and when the alpha radiation count rate was above a set concentration discriminator, the alarm sounded. The system was used to find contamination in the laboratory’s process water. The detector did not determine the alpha particle energy and therefore was not useful in the determination of the present radioisotopes. The system was used to distinguish between those liquids which had alpha emitting radionuclides present and those which did not. The system, however, had a printed record of the count rate and the time at which the liquids flowed past the detector [12]. The zinc sulfide phosphor scintillation layer, 4 or 5 mg/cm², consisted of a powder which was applied with adhesive to the light pipe in front of a two inch diameter photomultiplier tube. A thin layer of Mylar® was placed between the scintillation layer and the low-level liquids in order to not disrupt or dissolve the scintillation powder. The zinc sulfide phosphor detector was encased in stainless steel housing and the low-level process waste water was injected into the system. The injected liquid spread out radially over the Mylar® window as depicted in Figure 2.7. The contamination of Mylar® was not a concern as the continuous flow of water had the effect of flushing away any contamination. The alpha background of the detector quickly returned to normal after the contaminated water had passed through the detection system. In rare circumstances where the background was elevated because of cross-contamination, the detector was cleaned with a detergent or the Mylar® was replaced [12]. The zinc sulfide phosphor detector also boasted a large six-inch diameter
scintillation area, a considerable benefit for the detection of alpha particles from a low-level liquid source [12].

Figure 2.7 The flow through ZnS phosphor detector used to detect contamination in low-level aqueous solutions [11].

The relationship between the count rate and solution concentration was found for the ZnS phosphor detector along with the intrinsic detection efficiency and minimum detectable concentration. The count rate and concentrations of $^{238}\text{U}$ solution demonstrated a linear relationship. The intrinsic detection efficiency of the ZnS phosphor detector for $^{233}\text{U}$ was calculated as 87.7% and the minimum detectable concentration for the detector was found to be 0.48 Bq/mL for the $^{238}\text{U}$ solution [12].

A mathematical model was created to quantify the number of alpha particles which reached the detector. The model investigated the range of $^{238}\text{U}$ alpha particles through water and Mylar® and was used to determine the effective volume of liquid that an alpha particle could be detected in [12]. The location of the alpha particle when it was released from the nucleus, solid angle subtended by the detector, and the Mylar®
absorption layer between the solution and detector were accounted for in the model. The Mylar® window thickness, the only layer between the ZnS phosphor and water, was converted into water equivalent absorption thickness using absorption data in order to treat the materials as one. The number of alpha particles, \(N\), per unit time within an element of thickness which interacted in the detector was calculated as

\[
N = \int_{\theta_{\text{min}}}^{\theta_{\text{max}}} \frac{CAR_m}{2}(1 - \cos \theta) \sin \theta d\theta
\]  

(10)

where \(C\) was the specific activity, \(A\) was the surface area, \(R_m\) was the maximum range of the alpha particle through solution and Mylar®, and \(\theta\) was the angle between the directional vector of the alpha particle and the normal of the solution, Figure 2.8 [12]. The maximum angle of the alpha particle, \(\theta_{\text{max}}\), occurred when no alpha energy was deposited in the solution or ZnS phosphor layer. The minimum angle, \(\theta_{\text{min}}\), represented a scenario where the alpha particle was emitted perpendicular to the solution and was zero. The distance the alpha particles traveled through the water was determined by taking the integration of the thickness of water and the geometry fraction. The geometry fraction in equation 10, defined as \(\frac{1}{2}(1-\cos \theta)\), was simplified for a point source located in the middle of the surface area. In the experimental homogeneous solution, the alpha particles were originating throughout the solution which resulted in variations in the x, y, and z coordinates. Despite the variations in the experimental set up, the mathematical model only accounted for variations in the x direction. Although the mathematical model did not precisely portray the physical set up, the calculated activity of 0.83 Bq/mL was only slightly less than the experimental activity, 0.95 Bq/mL.
In 1984, Robert et al., reported using a flow-through alpha detection system which utilized a cerium-doped SiO$_2$ scintillator sandwiched between the circulation cell of the liquid and photomultiplier tube, Figure 2.9 [13]. This flow-through alpha detector was designed for the spent fuel reprocessing industry and for the general use in actinide laboratories for the detection of alpha particles from aqueous solutions. The liquid solution came in direct contact with the cerium-doped SiO$_2$ scintillation detector. The absence of the air gap was an advantage since the alpha particles did not lose energy in the absorption thickness. The scintillation glass was placed in contact with 5 N nitric acid for four months without showing signs of deterioration [13]. However, spent fuel associated with reprocessing is typically 14 M NaOH, a very corrosive liquid which is able to etch glass. The detector was coupled to a multi-channel analyzer which provided energy spectra of the alpha emitting samples. A spectrum of a $^{244}$Cm solution was detected using two different scintillation thicknesses: 35 µm and 2 mm. The thinner scintillator had a greater peak resolution [13]. The cerium-doped SiO$_2$ detector was tested with three different radionuclide solutions: $^{233}$U, $^{239}$Pu, and $^{241}$Am. For each radionuclide test solution, the detector showed a linear increase in count rate with the radionuclide concentration. Beta and gamma emissions, present from fission products in
the waste, were differentiated from alpha isotopes by using the spectrometric properties of the detector. The spectra of alpha and beta sources, Figure 2.10, showed the alpha peaks at higher energies than the beta peaks. A lower-level discriminator (LLD) was set as a means of separating the two types of radiation. The cerium-doped SiO$_2$ scintillator also possessed a quick decay time of 78 ns, the shortest time for scintillating glass [13]. The fast decay time was an advantage as pulse pile up did not occur in the detector. The study determined that this detection system was appropriate for use in spent nuclear fuel reprocessing plants [13].

![Figure 2.9](image.png)  
**Figure 2.9** The flow through detection system set up with scintillation glass detector and radioactive liquid [13].
A ZnS(Ag) detector was developed by McElhaney et al., which possessed a more rugged surface and better detection efficiency than the conventional ZnS(Ag) detector [9]. A Mylar® layer of 0.8 mg/cm² is typically used to achieve light tight properties for conventional ZnS(Ag) detectors; however, it is easily scratched or punctured as it is not a robust material. The ZnS(Ag) detector was developed to have a sturdier surface which consisted of a scintillation layer of ZnS(Ag) crystals and epoxy. The ZnS(Ag) powder was mixed with an optically transparent, low viscosity epoxy and poured into a mold. The ZnS(Ag) settled to the bottom of the strong, durable epoxy layer which allowed for a minimum layer of epoxy in front of the ZnS(Ag) crystals. The viscosity of the epoxy was lowered further by placing the layer in the oven. The scintillation layer consisted of an 8.0 mg/cm² ZnS(Ag) layer with an epoxy backing. A 0.087 mg/cm² aluminum layer was then spin-coated onto the scintillation layer to ensure an opaque detection window. A 0.02 mg/cm² cyanoacrylate hardcoat was also applied to the scintillation layer through spin-coating to increase the ruggedness of the detector. The detection layer was sturdier.
than the traditional ZnS(Ag) detector, more resistant to scratches and punctures, and waterproof. The detection of alpha radioactivity increased with the more rugged ZnS(Ag) detector due to the thin nature of the protective layers. The count rate of a $^{239}$Pu alpha source with unknown activity at a high voltage of 800 V was 73.4 Bq for the more rugged detector and 60.6 Bq for the conventional detector [9]. Resistance towards highly corrosive liquids remains to be determined.

A study of the theoretical response of a ZnS(Ag) scintillation detector, conducted by Skrable et al., investigated the self absorption of alpha radiation within a sample [14]. A thick solid and a source with minimal weight were studied as dense and light samples of infinite thickness. The detection of alpha particles from a thick dust deposit on an air filter proved to be difficult as the thickness resulted in the deposition of the majority of the alpha energy in the dust particles. The dust deposit resembled an infinitely thick alpha source as the deposit contained a thickness greater than the range of the alpha particles in the medium. An 11.4 cm diameter ZnS(Ag) scintillation layer was used to detect the count rate of the air filter and was then compared to theoretical calculations. The Mylar® layer, used to keep out light contamination and to keep the scintillation layer from being contaminated, was included in the calculations, and an air gap was not present [14]. The weightless sample model consisted of air that contained radon. Alpha particle ranges for various mixtures and compounds were investigated while $^{222}$Rn was used to model radionuclides in an infinite homogeneous gaseous medium [14].

The total specific alpha activity from soil was directly detected by Phoenix et al., using a ZnS(Ag) detector instead of eliminating the sample matrix through radiochemical procedures [15]. The experimental set up consisted of a Ludlum Model 43-1 ZnS(Ag)
detector coupled to a Ludlum Model 2200 scalar and a printer. The theoretical detector response count rate equation (cps), $y$, was

$$y = \frac{C A R_s}{4} - \frac{C A R_s}{2 R_w} x + \frac{C A R_s}{4 R_w^2} x^2$$

(11)

where $C$ was defined as the specific alpha activity, $A$ was the area of the source in contact with the detector, $R_s$ and $R_w$ were the mean alpha particle ranges in source and window material, respectively, and $x$ was the window mass density thickness [15]. The three soil samples investigated included natural uranium, natural thorium (both in secular equilibrium with their prodigies), and a sample of uranium oxide ($99\%$ U$_3$O$_8$). A two-inch diameter steel planchet was filled with soil and covered with a single piece of $0.297$ mg/cm$^2$ Mylar®. The covered planchet was turned on top of a large ZnS(Ag) detector. The Mylar® layer on top of the soil and the $0.297$ mg/cm$^2$ Mylar® layer included with the detector were between the sample and detector; however, additional Mylar® layers were added. The count rate for each respective Mylar® thickness was recorded from the two original layers ($0.594$ mg/cm$^2$) to a total of ten layers ($2.673$ mg/cm$^2$). The plot of count rate versus absorption thickness showed a quadratic curve, Figure 2.11. The curve was fit and by using the coefficient on the linear term, the specific activity was calculated. To calculate the specific activity of the sample, the surface area, mean alpha ranges in the source and window material, and the window mass density thickness were known. The total specific alpha activities from the soil samples were determined with a mean percent deviation of $17\%$. The alpha activities obtained from the soil samples confirmed the detection of alpha particles from thick sources using the detector response equation, however, with limits imposed by stochastic uncertainty and uncertainty from various parameters. The direct measurement technique was used as a cheap and easy way to
screen soils, ores, and industrial process materials. If the soil was not homogeneous, however, then the material detected did not represent the entire sample [15].

![Graph showing the count rate for a 232Th soil sample versus Mylar® absorber thickness](image)

**Figure 2.11** The count rate for a 232Th soil sample versus Mylar® absorber thickness [15].

Ayaz et al., compared the theoretical and experimental detection efficiencies of the ZnS(Ag) detector in the detection of liquid samples using equation 11 [16]. The gross alpha activity was measured from an infinitely thick alpha emitting source; however, instead of using soil to test the theory, manganese dioxide coated glass fiber filters and radioactive solutions were used as thick alpha emitting sources. MnO₂ coated glass was used to concentrate radium and uranium from neutral pH water and was then detected with the ZnS(Ag) detector [16]. In another experiment, aqueous alpha radioactivity was pumped into a light-tight black box where the radionuclide concentration was quantified with the aid of a ZnS(Ag) scintillation disc on a photomultiplier tube. The detection system was designed to be a flow-through alpha particle detector. The only barrier
between the sample and the scintillation layer was air as the Mylar® layer was left off the
detector face for better detection. The experimental and theoretical efficiencies were
within 7% of one another [16].

**ZnS(Ag) Spectroscopy**

The ZnS(Ag) alpha detector and a plastic veto detector were proposed for the
detection of low activity environmental samples by eliminating background counts in the
ZnS(Ag) detector. Ardid et. al, focused on determining the various sources of ZnS(Ag)
detector background in an attempt to improve the detection of low activity samples [17].
A 3.25 mg/cm³ ZnS(Ag) detector was connected to a plastic veto detector, a detector
typically used in the detection of beta and gamma radioactivity. A multi-channel
analyzer was used for monitoring data collection and the optimization of the operation
conditions [17].

The plastic veto detector and ZnS(Ag) detector, placed face-to-face, were
connected in a coincidence set-up to determine the origin of the ZnS(Ag) detector
background. The plastic veto detector had a higher detection efficiency of beta particles,
gamma rays, and cosmic rays. Although the ZnS(Ag) detection efficiencies for cosmic
rays were low, the parameter was investigated due to increased chance of interaction
from long counting times and large cosmic radiation flux. Coincident events were
detected by the plastic veto detector and ZnS(Ag) detector during a background count. A
coincident count rate of only $(8.4 \pm 1.8) \times 10^{-4}$ cps was detected while the total alpha
background count rate from the ZnS(Ag) detector without the veto detector was $(2.5 \pm
0.3) \times 10^{-3}$ cps, a significant fraction. The coincident events were tracked as they
represent potential rejects to be subtracted from the alpha background of the ZnS(Ag)
detector. The coincident events were caused by entities such as cosmic rays, gamma, or beta rays from the sample, or a gamma ray from the environment. A gamma ray interaction involving Compton scattering was another coincident event possibility as a secondary gamma ray could be detected in the plastic veto detector. The subtraction of the background coincident events from the total alpha background count rate from the ZnS(Ag) detector demonstrated the decrease in the background of the ZnS(Ag) detector. The pulse height spectra of the coincident events of the ZnS(Ag) detector and plastic veto detector were plotted on top of the total ZnS(Ag) background counts in Figure 2.12. The pulse height spectrum of the $^{241}$Am source, detected with only the ZnS(Ag) detector, was displayed to demonstrate the shape and location of an alpha peak [17].

A lead shield was constructed around the instrumental apparatus in an attempt to decrease the beta and gamma radiation from the environment. The lead shield decreased the total counts in the plastic veto detector by a factor of 20; however, it did not reduce the coincident events in the ZnS(Ag) and plastic veto detectors. Due to the consistency of the coincident events in the presence of shielding, it was determined that cosmic rays constituted the main contribution to the background [17].

A $^{137}$Cs gamma ray source was detected by the ZnS(Ag) and plastic veto detectors in order to determine the detection of coincident events due to Compton scattering. The gamma component of the ZnS(Ag) background was determined to be very small with less than 10% of coincident events from gamma rays [17].

The effect of radon on the detector background was studied by varying the air gap distance between the detectors. The total alpha rate increased with increasing air gap distance; however, the coincident events remained the same. Radon in the environment
was determined to be a significant factor in the detector background, but did not contribute to the detected coincident events [17].

![Figure 2.12](image)

**Figure 2.12** The pulse height spectra for the total events of the ZnS(Ag) detector background and the coincident events of the ZnS(Ag) detector and plastic veto detector are shown. The pulse height spectrum for a $^{241}$Am source was detected with a ZnS(Ag) detector [17].

**PIPS CAM Research**

The usefulness of the passivated ion implanted planar silicon (PIPS) continuous air monitoring (CAM) detector in the direct measurement of actinides from a liquid was investigated by Egorov et al., for the application of process waste streams. The study was aimed at producing an on-line detector for a process waste stream in the chemical processing of high-level nuclear waste. Although the study was intended for a flow-through waste stream, the experiments were conducted with 2 mL of solution in a sample holding cup constructed of polyvinylchloride (PVC), Figure 2.13, with an air gap of 0.5 cm between the detector and solution surface. The detector was connected to a Canberra
alpha spectrometer and the data was recorded with an Aptec analog to digital converter and multi-channel analyzer [18].

Figure 2.13 The PIPS CAM detector was placed over 2 mL of radioactive solution in a sample holding cup [18].

Five alpha solutions were used in order to obtain the pulse height spectra in Figure 2.14(a) using the PIPS CAM detector. The pulse height spectra, an alpha energy histogram with 1,022 channels or bins, had well-defined leading edges corresponding to the maximum energy of the alpha particles. The spectral shapes were consistent with the theoretical process of alpha particles traveling through an infinitely thick source. The continuous energy distribution was attributed to the decreasing alpha particle energy as the location of the nucleus which was emitting the alpha particle was at a greater depth. The leading edge channel numbers, Table 2.2, scaled linearly with the maximum alpha energy of the radionuclide solution as channel and energy were related. The counting sensitivities, the net count rate (cps) over activity concentration (Bq/mL), of the PIPS CAM detector were $0.221 \pm 0.005$, $0.180 \pm 0.004$, $0.160 \pm 0.005$, $0.139 \pm 0.005$, and $0.121 \pm 0.003\%$ for $^{244}$Cm, $^{241}$Am, $^{239}$Pu, $^{233}$U, and $^{230}$Th, respectively [18]. The counting sensitivity of the detector increased with alpha energy, Table 2.2. Despite the changing detection efficiency for the alpha particle energy, a disadvantage in gross alpha counting, the solution could be calibrated or the spectra de-convolved.
Beta and gamma interferences are problematic for the detection of alpha radiation. The radionuclides chosen for the study, $^{99}$Tc, $^{60}$Co, $^{137}$Cs, and $^{90}$Sr($^{90}$Y) are common fission and activation products in aged nuclear waste and would interfere in an on-line detector. The counting sensitivities (cps/Bq/mL) of the beta and gamma solutions were $4.06 \times 10^{-4}$, $4.69 \times 10^{-6}$, $1.89 \times 10^{-7}$, and $8.53 \times 10^{-10}$% for $^{90}$Sr($^{90}$Y), $^{137}$Cs, $^{60}$Co, and $^{99}$Tc as shown in Table 2.3 [18]. Although the beta and gamma interferences were located in the lower energy region in Figure 2.14(b), when a lower level discriminator was set, the alpha region of interest was reduced. The beta inference from the $^{90}$Sr($^{90}$Y) solution was located at channels as high as 550 while the leading edge of the $^{230}$Th pulse height spectra was at channel 643 in Figure 2.14. The counting sensitivities (cps/Bq/mL) for the reduced region of interest, 550-900 channels, were $0.113 \pm 0.004$, $0.072 \pm 0.003$, $0.057 \pm 0.003$, $0.033 \pm 0.002$, and $0.023 \pm 0.001$% for $^{244}$Cm, $^{241}$Am, $^{239}$Pu, $^{233}$U, and $^{230}$Th, respectively as shown in Table 2.2 [18]. The counting sensitivity of the detector decreased significantly from that of the larger alpha region of interest, channels 200-900.
Figure 2.14 The pulse height spectra from a) alpha emitting solutions and b) beta and gamma emitting solutions detected with a PIPS CAM detector [18].
Table 2.2 The counting sensitivities of $^{230}$Th, $^{235}$U, $^{239}$Pu, $^{241}$Am, and $^{244}$Cm solutions using the PIPS CAM detector in direct alpha assay using two different channel ranges: 200-900 and 550-900 channels [18].

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Alpha energy (MeV)</th>
<th>High energy spectral edge (channel)</th>
<th>Counting sensitivity (200-900 channels) (%)</th>
<th>Counting sensitivity (550-900 channels) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{230}$Th</td>
<td>4.688</td>
<td>643</td>
<td>$0.121 \pm 0.003$</td>
<td>$0.023 \pm 0.001$</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>4.821</td>
<td>672</td>
<td>$0.139 \pm 0.005$</td>
<td>$0.033 \pm 0.002$</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>5.157</td>
<td>735</td>
<td>$0.160 \pm 0.005$</td>
<td>$0.057 \pm 0.003$</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>5.486</td>
<td>785</td>
<td>$0.180 \pm 0.004$</td>
<td>$0.072 \pm 0.003$</td>
</tr>
<tr>
<td>$^{244}$Cm</td>
<td>5.806</td>
<td>838</td>
<td>$0.221 \pm 0.005$</td>
<td>$0.113 \pm 0.004$</td>
</tr>
</tbody>
</table>

*Principal peak of the highest energy.
*Point at which high energy edge intercepts zero.
*Counting sensitivity = count rate (c/s)/solution activity (Bq/mL); sample-detector stand-off distance 0.5 cm.

Table 2.3 The counting sensitivities for beta and gamma solutions, $^{99}$Tc, $^{60}$Co, $^{137}$Cs, and $^{90}$Sr($^{90}$Y) for the same channel ranges as Table 2.2 [18].

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Counting sensitivity (200-900 channels) (%)</th>
<th>Counting sensitivity (550-900 channels) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{99}$Tc</td>
<td>$8.53 \times 10^{-10}$</td>
<td>$4.52 \times 10^{-9}$</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>$1.89 \times 10^{-7}$</td>
<td>$2.09 \times 10^{-6}$</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>$4.69 \times 10^{-6}$</td>
<td>$8.87 \times 10^{-11}$</td>
</tr>
<tr>
<td>$^{90}$Sr($^{90}$Y)</td>
<td>$4.06 \times 10^{-4}$</td>
<td>$1.31 \times 10^{-9}$</td>
</tr>
</tbody>
</table>

*Counting sensitivity = count rate (c/s)/solution activity (Bq/mL); sample-detector distance 0.5 cm; sample volume 2mL.

The effects of the solution concentration on the detector count rate of the PIPS CAM detector were shown in Figure 2.15. The relationship demonstrated linearity between the count rate and concentration, which showed the absence of pulse pile-up and dead time at these concentrations. The relationship also signified the capability of PIPS CAM detection via direct alpha assay [18].
The effects of solution density on alpha detection were investigated with the PIPS CAM detector. The solution density was graphed against the relative efficiency in Figure 2.16. The relative efficiency assumed unity for the detection efficiency of the 1.0 g/mL density solution. The higher density solutions were associated with lower efficiencies. The data points followed a general decreasing trend between efficiency and density; however, the scattered points showed uncertainty in the data. Aged nuclear waste has a density range between 1 and 1.4 g/mL which resulted in approximately a 20% loss in the relative counting sensitivity for the PIPS CAM detector [18]. The solution density would have to be independently quantified in the process waste stream in order for a correction to be made.
The distance between the PIPS CAM detector and the solution surface was varied in order to observe the spectral response and the effect on count rate. Figure 2.17 showed the detector spectral response to a $^{241}$Am solution with increasing distance. The leading edges of the pulse spectra moved to the lower energy channels as the distance increased. The alpha particles which reached the detector had a decreased energy due to energy absorption in the air gap. Fewer alpha particles reached the detector as the air gap increased due to solid angle effects. Figure 2.18 demonstrated the decrease in count rate as the stand-off distance increased [18].

Figure 2.16  The relative efficiency of the PIPS CAM detector in regards to the effect from the solution density [18].
The minimal detectable concentration (MDC) for the PIPS CAM detector was found using equation 12:

\[
MDC = \frac{4.653 \sqrt{b} + 2.706}{f \varepsilon T}
\]  

(12)
where \( b \) is the background counts, \( T \) is the counting time, \( \varepsilon \) is counting sensitivity, and \( f \) is the yield or branching ratio of alpha particles for a particular energy. The branching ratio was assumed to be one for each radionuclide, the counting sensitivities were taken from Table 2.2 and a background count rate of \( 7 \times 10^{-3} \) cps was used to calculate the MDC for a 10 minute counting time. The minimum detectable concentrations of \( ^{244}\text{Cm} \), \( ^{241}\text{Am} \), and \( ^{239}\text{Pu} \) were found to be 17.7, 23.0, and 35.0 Bq/mL for the PIPS CAM detector [18].

Several sample nuclear waste matrices were analyzed using the PIPS CAM detector in order to evaluate the effectiveness of direct detection of alpha particles from a liquid surface. The count time was kept short—10 minutes—in order to maintain a correlation with an on-line waste stream. The 550-900 channel range was used in order to minimize beta and gamma spillover into the alpha ROI. The detector was calibrated for the detection efficiency of \( ^{241}\text{Am} \) and the linear relationship for density was used. The alpha activities of several solutions were compared in Table 2.4 using direct measurements from a PIPS CAM detector and a conventional radio-chemical method. The activities measured by the two measurement methods are in good agreement [18].

Table 2.4 A comparison of alpha activity measurement between the conventional radio-chemical method and direct alpha measurement using the PIPS CAM detector [18].

<table>
<thead>
<tr>
<th>Sample matrix</th>
<th>Density (g/mL)</th>
<th>Measured alpha activity (Bq/mL)</th>
<th>Laboratory analysis</th>
<th>Direct diode measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dilute HNO(_3)(^b)</td>
<td>1.00</td>
<td>403(\pm)40</td>
<td>375(\pm)38</td>
<td></td>
</tr>
<tr>
<td>Hanford AZ-101(^c)</td>
<td>1.22</td>
<td>187(\pm)19</td>
<td>209(\pm)30</td>
<td></td>
</tr>
<tr>
<td>Hanford Tank AZ-102</td>
<td>1.26</td>
<td>147(\pm)15</td>
<td>166(\pm)47</td>
<td></td>
</tr>
<tr>
<td>Hanford Tank AP-104</td>
<td>1.29</td>
<td>83.2(\pm)8</td>
<td>139(\pm)44</td>
<td></td>
</tr>
<tr>
<td>Hanford Tank AN-107</td>
<td>1.25</td>
<td>86.1(\pm)9</td>
<td>77.1(\pm)32</td>
<td></td>
</tr>
<tr>
<td>Savannah River Tank 43</td>
<td>1.20</td>
<td>279(\pm)28</td>
<td>318(\pm)61</td>
<td></td>
</tr>
</tbody>
</table>

\(^a\)Radiochemical measurement, reported error is \( \pm 10\% \).
\(^b\)Calibration performed using \( ^{241}\text{Am} \) solution, error is propagated counting error (2s); stand-off distance 0.5 cm.
\(^c\)Spiked with \( ^{244}\text{Am} \).
Computer Modeling

The scientific model is a simplification of a real system which is used for prediction and system control. Modeling determines how parameter changes affect the outcome and the system as a whole. There are many types of models including iconic models which pictorially or visually represent a system, analog models that employ a set of properties to represent another set in the desired system, and symbolic models which require logical or mathematical operations to formulate a solution. Symbolic models are classified as either dynamic or static. Dynamic models possess a time variable interaction while static models lack a time variation. Symbolic models are also defined as either deterministic or stochastic [19]. A deterministic population model makes a single prediction about the future state of a population while a stochastic model associates each possible future state with a probability and incorporates at least one random variable. The models could be applied to the same population or example; however, they will not necessarily have a matching mean or outcome [20].

Monte Carlo simulation is a popular form of stochastic modeling which is applied in all areas of science and mathematics [20]. The Monte Carlo method was developed during the Manhattan Project as a way to integrate otherwise impossible mathematical functions. One of the first examples of the method was Buffon’s estimation of $\pi$ as needles were thrown onto a grid. The Monte Carlo method was used in the examination of the Boltzmann equation and also in the estimation of the correlation coefficient in Gosset’s t-distribution along with numerous other applications. The Monte Carlo method is useful in the modeling of a random sampling from a population or for doing statistical experiments [21].
Monte Carlo modeling is the simulation of physical and mathematical problems using random variables and known probability distribution functions. A random variable is one in which the user does not know the value; although, the estimated values and their probabilities are known. The simulations are made possible by modern computers and random number generators. Although a computer generated random number sequence is impossible, pseudo-random numbers have proved to be acceptable for Monte Carlo purposes and are assumed to be independent [22]. A pseudo-random number generator repeatedly simulates variables in a scenario resulting in many possible outcomes with a mean, variance, and the probability density functions of the output variables [20].

Various Monte Carlo modeling software, such as the SRIM/TRIM package, GEANT4, and the MCNP codes, are used for simulating alpha particle behavior in a medium, however, these models were not recommended for spectrometry simulations [23]. The SRIM package, of which TRIM is a subset, was designed for the calculation of the range and stopping power of alpha particles or ions through various materials. The program uses statistical algorithms which allow an averaging of the collision results between the calculated collisions and gives a full quantum mechanical treatment to the ion-atom interactions. The TRIM package is capable of calculating damage cascades, ion distribution, and surface sputtering along with neutron, electron, proton cascades and the ion energy, angle, and position [24]. GEANT4 is also a simulation toolkit for the passage of charged and uncharged particles through matter. The GEANT4 package was created at CERN and is based in the C++ computer language. GEANT4 allows complex geometries, various materials and fundamental particles, and the responses of sensitive detector components. The program tracks particles through materials and external
electric fields, explores particle interactions, and generates event data. The program is particularly useful for nuclear physics, medical physics, accelerator design, and space engineering [25]. The MCNPX, Monte Carlo N-Particle Transport Code Extended, was created at Los Alamos National Laboratory for the transport simulation of nearly all nuclear particles [26]. The Fortran 90 based program allows for three dimensional geometries and time dependence. The program simulates the transport of 34 various particle types at different energies, light ion recoil, and fission multiplicity in addition to many other applications [26].

A Fortran 95 Monte Carlo code, advanced alpha-spectrometric simulation (AASI), was created for the simulation of the energy spectra of alpha particles. The modeling program takes into account the decreasing energy of the charged particles and the Rutherford scattering of the alpha-nucleus interaction. The program also includes the accessibility of various sample types such as thick samples, aerosol particles, and non-uniform samples [23]. The AASI Monte Carlo modeling program was developed for the simulation of energy spectra of alpha radioactivity from an aerosol sample. Previously published data for aerosol experimentation was used to validate the simulation program as the experimental and simulated results agree within one standard deviation. The most significant section of the simulation code, alpha energy loss, was constructed by putting a thin Mylar® sheet between the source and the detector. The simulation proved to be a success, as the simulated spectra followed the experimental data almost exactly [27].

The propagation of particles through matter was dictated by two main processes in the AASI program: scattering and absorption. The emission of a particle from a randomly selected point in the matrix, the distance it traveled to a scattering point, the
determination of the next direction vector or scattering angle, and the continuous loss of energy during each step for a charged particle was incorporated into the alpha spectroscopy simulation code. If the particle energy was below a set cut-off value then the particle tracking stopped, however, if the energy was above the value, the program continued to calculate the distances traveled and the energy loss. The AASI program also took into account the average solid angle which corresponded with the set up geometry in the calculation of the geometrical detection efficiency. The energy loss of the alpha particle was calculated using stopping power parameterizations and the energy loss straggling of alpha particles was approximated by a Gaussian energy distribution [23]. This approximation was not necessarily correct for very thin absorption layers; however, it gave a reasonable estimate for most cases. The alpha particles were assumed to travel in a straight line with the exception of scattering on the planchet. The program also took into account coincidences between particles emitted from the same source. The pulse summation between the alpha particle and daughter emissions occurred as the excited state lifetimes were usually shorter than the integration time of the data acquisition electronics [23].

The AASI computer model contained two limitations: complete energy absorption in the theoretical detector and a dependency on the response of the detector. The remaining energy of the alpha particle after passing through the media between the source and the detector was deposited in the detector. The alpha particles were not tracked inside of the detector and the energy loss was not calculated as it was assumed that all of the remaining energy was deposited in the detector. Additionally, several parameters of the detector were added to the AASI code which eliminated the
independence of the program from the detector such as the detector full width half
maximum (FWHM) and the parameters of the exponential tailing function. The detector
response of a silicon detector to monoenergetic alpha particles was not observed as a
Gaussian fit, but as a double exponential tailing function. The function was entered into
the program with a Gaussian convolution which was then applied to the simulated spectra
[23].

A Monte Carlo simulation was developed by Ferrero et al., to model the scattering
and slowing down of alpha particles in $2\pi$ geometry. The alpha particles traveled in a
straight line with the exception of Rutherford scattering with the nucleus and alpha
backscattering as the particles were absorbed into the media. The scattering of the alpha
particle with the nucleus of the absorbing material was considered to be the most
important consideration in both the backscattering and self-absorption correction factors.
The elastic scattering relationship, the scattering of the alpha particle off the nucleus of
the absorbing atom, was derived from classical mechanical theory using center of mass.
Inelastic scattering, the scattering of the alpha particle off the electrons of absorber
atoms, resulted in a loss of energy without significant angular deviations. The energy
loss was calculated from the electronic stopping power data of Ziegler [28]. The
constructed Monte Carlo model was tested against experimental data from a $2\pi$ ionization
chamber coupled to a multi-channel analyzer. Various thicknesses of UF$_4$ corresponding
to 4 to $18.8$ $\mu$g/cm$^3$ were electroplated onto stainless steel planchets with diameters of $1.0$
and $2.5$ cm, each. The Monte Carlo simulation agreed with equivalent data from
experimental measurements and theoretical calculations within the estimated
uncertainties, in regard to the ratio of counts and activity. The simulated energy spectrum also had good agreement with the experimental energy spectrum [28].

The simulation by Ferrero et al., successfully calculated the backscattering coefficients for different material planchets and various alpha emitters in $2\pi$ geometry. The simulation was applied to a semiconductor detector in the investigation of alpha particles leaving the planchet at extreme angles. Energy resolution depended on the characteristics of the detector, geometry of the setup, composition and thickness of deposited source, and the planchet material. The simulation program was used to model contributing parameters of the peak shape such as the standard deviation, the distance between the source and detector, the window thickness, and the thickness of the source itself. The simulated spectra were distributed using a Gaussian function with various standard deviations and were shown to be symmetric. A standard deviation of 10 keV was used in the rest of the simulations. The alpha pulse height spectrum broadened as the distance between the alpha source and detector decreased due to the increase in extreme alpha angles. The particles which hit the detector at extreme angles lost more energy in the window and caused an asymmetry in the low-energy peak region. An increase in the window thickness also caused asymmetry and the FWHM to increase along with the shift toward the lower energies. As the thickness of the source was increased, the peaks widened and shifted to the lower energies. Experimental data was taken with a semiconductor to correspond with the simulated pulse height spectra. The spectra were in good agreement with the simulation. The backscattering of the alpha particles in the source and support were tracked and determined to be insignificant to the peak shape [29].
The determination of particle size, shape, and composition of radioactive aerosols were attempted by Pickering using alpha energy spectrometry simulations [30]. Monte Carlo calculations of the energy spectra were conducted using spherical and elliptical particles as a function of particle size and orientation inside of a volume. Past methods of particle size determination using radioactivity included autoradiography of filter deposits and also scintigraphy, when alpha particles interacted with ZnS screens. Both methods were slow and inconveniently relied on manual track counting. An attempt to automate the alpha autoradiography by substituting position sensitive semiconductors for the photographic or polymer sheet detectors proved useful only at low concentrations. The procedure failed if more than one particle was present on a detector and also if the particles fell into the space between the detector display. In an attempt to improve upon the current technologies, a solid-state surface barrier detector was investigated for the feasibility of using alpha spectroscopy to determine size and composition of radioactive particles [30]. The dependence of detector resolution on the spectral peak shape was investigated with samples of various thicknesses. A theoretical monolayer source, an experimental monolayer source, a semi-infinitely thick source, and a particulate source resulted in varying pulse height distributions as illustrated in Figure 2.19. The variation in pulse height distribution demonstrated that peak resolution was a function of the path length of the alpha particle within the material and also portrayed the difference in peak shape resulting from the larger and smaller particles in the particulate source. In addition, the particle size, particle shape, orientation, and the solid angle of the alpha particles were investigated using Monte Carlo calculations. The orientations of the alpha particles inside of the particulate were randomly selected along with the direction of the alpha
particles. If the angle between the direction of the alpha particle and y-axis was smaller than the designated angle than the alpha particle was traveling towards the detector. The distance to the surface of the particulate was calculated and the energy of the alpha particle leaving the spheroid was calculated. Negative energies of the alpha particles meant the alpha energy was completely deposited inside of the particulate and the alpha particles were never detected. The simulated alpha spectra showed various peak shapes for particulate size and shape; however, they were difficult to quantify. Although the elliptical shape of a particle was expected to be determined by detecting the particle from different angles, the simulated alpha spectra remained hard to quantify in a setting with a heterogeneous distribution and an unknown density. The method did prove acceptable for radioactive aerosols produced from the grinding machines in the manufacturing of nuclear fuels, however. The particles were equiaxial, well characterized, and consisted in a log-normal distribution; which allowed for a simple experimental method to be derived which allowed for the particle size to be determined [30].
Figure 2.19 The detection resolution as a function of source thickness and size [30].
CHAPTER 3
RESEARCH OBJECTIVES

An evaluation of the effectiveness of a ZnS(Ag) detector in the quantification of alpha-emitting radionuclides in aqueous solutions was conducted and the data was compared and contrasted with the PIPS CAM detector. The experimental results from the ZnS(Ag) detector and both experimental and previously published data for the PIPS CAM silicon detector were used to determine which instrument was best equipped for the direct detection of alpha particles from solution [18].

Several parameters were taken into consideration including solution concentration, the separation distance between the liquid and detector, solution density, detection surface area, and beta and gamma interference in the detectors. The absolute detection efficiency of the alpha particles from the liquid surface and pulse height spectra characterized with the ZnS(Ag) detector were compared to electroplated standards and the PIPS CAM detector.

The experimental pulse height spectra of the infinitely thick aqueous sources were compared to theoretical computer-generated spectra. The spectral shapes of the ZnS(Ag) and PIPS CAM detectors were discussed in relation to the theoretical expectations. The theory of alpha particle absorption in matter was explored and the effects of light propagation and collection were discussed. The effects of thin and thick scintillator thicknesses on the pulse height spectra were determined for the ZnS(Ag) detector.
Specific tasks conducted to meet the objectives include:

- Determined if the ZnS(Ag) detector was suitable for on-line alpha assay
  - Experimental data

- Determined if the PIPS CAM detector was suitable for on-line alpha assay
  - Experimental data
  - Published data

- Understood and explained the shape of the ZnS(Ag) pulse height spectra
  - Experimental data
  - Computer simulations
  - Theory of alpha particle interactions with matter
CHAPTER FOUR
MATERIALS AND METHODS

Apparatus

The energy spectra of aqueous alpha solutions were achieved by coupling a ZnS(Ag) detector to a multi-channel analyzer. The detection apparatus was configured using a Ludlum Model 43-1 ZnS(Ag) scintillation detector (Ludlum Measurements, Inc., Sweetwater, TX). The detector had an active face of 78.5 cm$^2$ with a layer of 0.8 mg/cm$^2$ aluminized Mylar® over a 3.5 mg/cm$^2$ layer of ZnS(Ag) scintillation crystals. A prefabricated 14 mg/cm$^2$ scintillation layer was later supplemented onto the detector. Using the Canberra Model 2006E preamplifier (Canberra Industries, Inc., Meriden, CT) or the Ludlum Model 297 Signal Splitter, the ZnS(Ag) detector was connected to the Bertan Associates Model 305 DC high voltage power supply (Bertan Associates, Inc., Syosset, NY) and the Ortec 572 amplifier (Ortec, Inc., Oak Ridge, TN) as shown in Figure 4.1. The amplified signal of the ZnS(Ag) detector was read by an Aptec-NRC MCArd analog to digital converter and multi-channel analyzer which transmitted the 511 channel energy spectra to the computer. The high voltage was set at 800 V when using the charge-sensitive preamplifier and 850 V when using the signal splitter. Details of the high voltage optimization are presented in Appendix A.
Figure 4.1 The experimental setup for the ZnS(Ag) detector.

A CAM 450AM PIPS detector (Canberra Industries, Inc., Meriden, CT), typically utilized for continuous air monitoring (CAM), was utilized. The detector had a 0.5 µm aluminum coating over the 450 mm² detection surface creating a light proof detector. A 1 µm polymer coating also created a detector surface which was resistant to chemical and physical properties. The detector was connected to a Canberra Alpha Spectrometer (Model 7401VR) and the data was recorded with an Aptec-NRC MCArd analog to digital converter and multi-channel analyzer as shown in Figure 4.2. The bias was set at 24.3V for the PIPS CAM detector. The original spectra for the PIPS CAM detector had 1024 channels, however, the data was re-binned to 511 channels using Microsoft Excel in order to directly compare the PIPS CAM data to the data from the ZnS(Ag) detector.

Figure 4.2 The experimental set up for the Canberra PIPS CAM detector.
Two detection set ups were required: one in the Pacific Northwest National Laboratory (PNNL) and one at Rich Laboratory of Clemson University. The PIPS CAM detector data was only acquired in PNNL. The Ludlum Model 43-1 ZnS(Ag) detector, Figure 4.3, was transported between the two locations which allowed for the same detector to be used in all measurements. The same models, though different pieces of equipment, of the Bertan high voltage, Ortec amplifier, and the Aptec-NRC MCArd analog to digital converter and multi-channel analyzer were used in both locations. The signal splitter was used in the data taken at Pacific Northwest National Laboratory and select experiments at Clemson, while the charge-sensitive preamplifier was used only at Clemson, Table 4.1. Differences in the pulse height spectra of data taken with the ZnS(Ag) detector in conjunction with the signal splitter and charge-sensitive preamplifier were explored in Appendix B. A Tektronix oscilloscope TAS 465 was used in conjunction with the Clemson apparatus in order to observe the electronic signal from the ZnS(Ag) detector and its associated electronics.

![Figure 4.3](image)

**Figure 4.3** The Ludlum Model 43-1 ZnS(Ag) scintillation detector clamped to a stand with bolts propping it over a PVC sample holding cup.
Table 4.1 The experiments which utilized the signal splitter or charge-sensitive preamplifier.

<table>
<thead>
<tr>
<th>Signal Splitter</th>
<th>Charge-sensitive preamplifier</th>
</tr>
</thead>
<tbody>
<tr>
<td>Solution Concentration</td>
<td>Energy Resolution and Spectral Shape</td>
</tr>
<tr>
<td>Solution Density</td>
<td>Absolute Detection Efficiency</td>
</tr>
<tr>
<td>Air Gap Separation</td>
<td>Detection Area</td>
</tr>
<tr>
<td></td>
<td>Minimum Detectable Concentration</td>
</tr>
<tr>
<td></td>
<td>Thin versus Thick Scintillation Layer</td>
</tr>
</tbody>
</table>

The ZnS(Ag) scintillator was covered by a Mylar® layer designed to make the instrument light tight; however, a small scratch in the surface of the Mylar® created a light leak. A black cloth was wrapped around the detector to minimize the effects of the light leak. The light shield corrected the problem as observed with the oscilloscope. The light shield was used in all of the experiments conducted at both Pacific Northwest National Laboratory and Clemson University.

The liquid samples were measured after pipetting the aqueous solutions into the sample holding cup which was placed at the center of the active face of the detector. The sample holding cups were made from polyvinylchloride (PVC). The large ZnS(Ag) detector face was masked to allow a smaller opening, a condition to mimic the companion experiments conducted with the PIPS detector. After 2 mL of liquid was placed into the PVC sample holding cup, the resulting thickness of the liquid source was 4.1 mm. The detector was placed on top of spacers that raised the detector face to 5.2 mm above the surface of the liquid as shown in Figure 4.3. The PNNL sample holding cup had an inner depth of 9.2 mm and an inner diameter of 23.9 mm. The inner diameter of the sample holing cup corresponded to the active surface of the Canberra PIPS CAM detector. The Clemson sample holding cup, also made of PVC, was created to mimic the PNNL sample holding cup with regards to the 5.2 mm air gap distance when filled with 2
mL of solution. The Clemson sample holding cup had an inner diameter of 23 mm and an inner depth of 9.8 mm. The 4.15 cm$^2$ Clemson sample holding cup was slightly smaller than the 4.48 cm$^2$ cup used at PNNL. A large diameter PVC cup was constructed at Clemson University to correspond with the large detection area of the ZnS(Ag) detector. The large sample holding cup had an inner diameter of 108 mm and inside depth of 5.6 mm. The arrangement provided an air gap of 5 mm when 30 mL of solution was present in the cup. The steel rim of the ZnS(Ag) detector was supported by the sides of the large cup. Large diameter shields, with inner diameters of 40.4 mm and 79.3 mm, restricted the detection area of the large detection cup for experiments which investigated the effects of solid angle.

A 6.35 mm Plexiglas shield (749.3 mg/cm$^2$) acted as an absorber between a beta source and the detectors and also acted as a platform for the alpha electroplated standard (EPS), Figure 4.4(a). In the absence of the beta source, the shield was used as a stage for the alpha electroplated standard in an attempt to maintain solid angle, Figure 4.4(b). The distance between the alpha electroplated standard and the detector was consistent for the experiments involving the charge-sensitive preamplifier, however, the distance could not be replicated for the signal splitter experiments.
Reagents and samples

All chemicals used at PNNL were of analytical grade. De-ionized water (18 MΩ-cm, Barnstead E-Pure, Dubuque, IA) was used in the preparation of the reagents. The actinide solutions were prepared by diluting stock solutions, $^{244}$Cm, $^{241}$Am, $^{239}$Pu, and $^{230}$Th, with 2% nitric acid and the activities of $2.1 \times 10^3 \pm 4.2$ Bq/mL, $6.9 \times 10^3 \pm 6.4$ Bq/mL, $5.0 \times 10^3 \pm 7.6$ Bq/mL, and $1.8 \times 10^4 \pm 12.2$ Bq/mL, respectively, were standardized with the Packard 2550 TR/AB liquid scintillation counter assuming 100% detection efficiency. The $^{241}$Am EPS had an activity of $259 \pm 3.9$ Bq. The samples of various densities were prepared by the addition of sodium nitrate salts to 2% nitric acid actinide solutions. The resulting density was confirmed gravimetrically and the activity of each sample was confirmed with the liquid scintillation counter.

The solutions and electroplated standards used at Clemson University included four actinide solutions, three NIST electroplated standards, and a plutonium electroplated source made at the Savannah River Site. The Clemson University actinide solutions
consisted of $^{244}$Cm, $^{239}$Pu, $^{238}$U/$^{234}$U (natural uranium), and $^{238}$U (depleted uranium) at concentrations of $220 \pm 3.1$ Bq/mL, $16200 \pm 26.8$ Bq/mL, $5137 \pm 15.6$ Bq/mL, and $11.3 \pm 0.16$ Bq/mL, respectively. The solutions were below pH 2 and the concentrations were standardized with the Wallac 1415 DSA liquid scintillation counter assuming 100% detection efficiency. The natural uranium solution was assumed to have an alpha energy of 4.51 MeV, a calculated average of the $^{238}$U and $^{234}$U alpha particle energies, while the total alpha activity was utilized. The NIST electroplated alpha standards included a $7151.7 \pm 93.0$ Bq $^{241}$Am, $1664 \pm 18.1$ Bq $^{241}$Am, and $489.9 \pm 6.4$ Bq $^{238}$Pu. The plutonium electroplated source was prepared at the Savannah River National Laboratory using NIST traceable sources. The source consisted of the isotopes $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{242}$Pu, and $^{241}$Am with the respective activities, 0.03, 264.07, 2.28, 0.37, 0.09, and 0.46 Bq. A high-activity $^{90}$Sr($^{90}$Y) source, $7.4 \times 10^8$ Bq, and a $6.88 \times 10^5$ Bq $^{137}$Cs source were also used to investigate beta and gamma interaction in the ZnS(Ag) detector.

The effective activity, the aqueous radioactivity which contributed to a signal in the detector, was calculated based on the known activity concentration, the surface area of the detector, and the active depth. The active depth was an estimation of the solution thickness which emitted alpha particles that were capable of leaving the solution. The estimation of active depth allowed for a more realistic detected volume. The active volume of solution which the detectors were detecting was determined by multiplying the surface area of the cup, A, by the active depth, x. Equation 13 gives the relationship between the effective activity (EA), solution concentration (C), and active volume.

$$EA(Bq) = C \left( \frac{Bq}{ml} \right) A \left( cm^2 \right) x(cm)$$  \hspace{1cm} (13)
The maximum range of an alpha particle in solution was calculated using equations 1, 2, and 3 and was taken as the active depth, x, in solution. An alpha particle which originated below the maximum depth deposited all of the alpha energy into the aqueous matrix and did not leave the solution. The active depth of the radionuclide solutions, the maximum depth from which an alpha particle reached the surface, was dependent on alpha energy as the ranges of alpha particles through the solution was a function of alpha energy. There was a 35% difference in the alpha ranges through solution for $^{244}$Cm, the maximum alpha energy used, and $^{238}$U, the isotope with the least energy used—54.5 µm and 30.6 µm, respectively. The alpha ranges for the various energies and the solution effective activities used in these experiments are shown in Table 4.2.

Table 4.2 The active depths and solution effective activities for various radionuclide energies.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Alpha Energy (MeV)</th>
<th>Active Depth (µm)</th>
<th>PNNL Solution Effective Activity (Bq)</th>
<th>Clemson Solution Effective Activity (Bq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{244}$Cm</td>
<td>5.81</td>
<td>54.5</td>
<td>52.3 ± 0.10</td>
<td>5.36 ± 0.076</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>5.49</td>
<td>49.8</td>
<td>155.1 ± 0.17</td>
<td></td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>5.14</td>
<td>44.7</td>
<td>99.4 ± 0.13</td>
<td></td>
</tr>
<tr>
<td>$^{230}$Th</td>
<td>4.69</td>
<td>38.0</td>
<td>302.8 ± 0.21</td>
<td></td>
</tr>
<tr>
<td>$^{234}$U/$^{238}$U</td>
<td>4.51*</td>
<td>35.4</td>
<td>80.2 ± 0.236</td>
<td></td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>4.18</td>
<td>30.6</td>
<td>0.18 ± 0.002</td>
<td></td>
</tr>
</tbody>
</table>

*Average energy of the alpha particle of the two radionuclides

**Procedure**

Following each sample analysis, the sample holding cups were washed with two 1M nitric acid rinses and two distilled water rinses before being scrubbed dry. Prior to each experimental set, a background was taken with the detector in order to ensure that there was no significant contamination on the detector. The background was also subtracted from the upcoming data set.
CHAPTER 5
RESULTS AND DISCUSSION

The ZnS(Ag) and PIPS CAM detectors were characterized to determine the detector best equipped to quantify alpha radioactivity from aqueous solutions. The spectral shape and energy resolution of the pulse height spectra were evaluated and compared to theory. The absolute detection efficiency of each detector was determined with and without the presence of beta and gamma interference. The effects of solution concentration, detection area, solution density, and separation distance on the ZnS(Ag) detector were investigated and compared with measurements that were previously conducted with a PIPS CAM detector. Two detection layer absorber thicknesses of ZnS(Ag) were studied. A computer program was designed to model the experimental setup and the experimental and theoretical pulse height spectra were compared.

Energy Resolution and Spectral Shape

The pulse height spectra of an $^{241}$Am electroplated standard and an $^{241}$Am aqueous solution detected with a PIPS CAM detector are compared in Figure 5.1(a). The pulse height spectrum of the $^{241}$Am electroplated standard measured with the PIPS CAM detector was relatively narrow and characterized the finite energy distribution of an electroplated standard. The pulse height spectrum demonstrated a tailing effect due to the absorption through the air gap and the protective surface layer associated with the CAM. Despite the absorption layers, the alpha peak energy resolution for the $^{241}$Am electroplated standard was 3.1%. The PIPS CAM detector is renowned for its resolution due to the solid-state properties of the detector. The PIPS CAM detector allowed for all
the alpha energy to be deposited in the infinitely-thick active detection region. The leading edges in the $^{241}$Am aqueous solution and $^{241}$Am electroplated standard pulse height spectra from the PIPS CAM detector intercepted in Figure 5.1(a) at the maximum alpha energy that was deposited in the detector. The low energy noise tail present in the pulse height spectrum for the aqueous solution, Figure 5.1(a), was absent in the pulse height spectrum of the electroplated standard due to a higher LLD setting. The pulse height spectrum of the aqueous solution, an infinitely-thick source, illustrated a broad peak and alpha energy tailing as the alpha particles lost energy in the solution through inelastic scattering within the medium. An infinitely-thick source possessed a material thickness greater than the alpha range in that material—a property characteristic of liquid samples. The alpha particle which originated from the top of the liquid surface deposited the maximum alpha energy into the detector. Alpha particles located below the surface lost a significant amount of energy to the aqueous matrix. Alpha particles which originated deep in the solution never departed from the solution and deposited all of the alpha energy into the aqueous matrix. The aqueous solution had a high stopping power and was the leading contributor of alpha energy absorption for aqueous solutions.

The pulse height spectra of a $^{239}$Pu electroplated standard and a $^{239}$Pu aqueous solution obtained with the Ludlum Model 43-1 ZnS(Ag) detector in conjunction with the charge-sensitive preamplifier were compared in Figure 5.1(b). The energy resolution of the pulse height spectrum of the $^{239}$Pu electroplated standard taken with the ZnS(Ag) detector was 118%—much greater than that of the PIPS CAM detector. The spectral shape of the ZnS(Ag) pulse height spectrum for the $^{239}$Pu (5.147 MeV) electroplated standard is in good agreement to the pulse height spectrum for a 5.5 MeV alpha source in
Figure 2.6. The poor resolution of the alpha peak was attributed to scintillation properties of the detector. The ZnS(Ag) scintillation light intensity created was proportional to the energies deposited in the scintillator. The scattered light was partially absorbed in the scintillation layer and light pipe which resulted in further degradation of energy resolution. The ZnS(Ag) pulse height spectrum of the $^{239}$Pu aqueous solution, Figure 5.1(b), did not drastically differ from the pulse height spectrum of the electroplated standard. The poor resolution of the ZnS(Ag) detector created difficulties in distinguishing alpha peak characteristics. The leading edges of the pulse height spectra for the electroplated standard and aqueous solution did converge at the maximum energy as expected. The peak maximum of the pulse height spectrum for the $^{239}$Pu aqueous solution shifted to the lower energies relative to the pulse height spectrum of the electroplated standard. The pulse height spectrum for the aqueous solution had a greater number of lower-energy alpha particles as the particles from the solution have a decreased energy due to energy absorption in the solution matrix.
Figure 5.1 The pulse height spectra of a) $^{241}$Am and b) $^{239}$Pu samples for the PIPS CAM detector and ZnS(Ag) detector, respectively. Spectra corresponding to both aqueous radioactive sources and electroplated standards are shown.

The PIPS CAM detector was characterized with four aqueous solutions, $^{244}$Cm, $^{241}$Am, $^{239}$Pu, and $^{230}$Th, while the ZnS(Ag) detector was characterized with three aqueous solutions, $^{244}$Cm, $^{239}$Pu, and $^{234}$U/$^{238}$U. The PIPS CAM pulse height spectra in Figure 5.2(a) corresponds with the published graph, Figure 2.14(a), but with better
counting statistics. The differences in the resolution of the PIPS CAM and ZnS(Ag) detectors were observed in the leading edge of the pulse height spectra in Figure 5.2(a) and (b). The leading edges of the PIPS CAM detector were clear and quickly descended at the maximum alpha energies. The poor resolution of the ZnS(Ag) detector resulted in prolonged leading edges. The leading edges of the ZnS(Ag) pulse height spectra were quantified at the tenth maximum (FWTM) due to the broadening effect of the ZnS(Ag) detector. The spectral leading edges of the two extreme alpha energies, 5.8 MeV from the $^{244}$Cm solution and 4.7 MeV from the $^{230}$Th solution were separated by 95 channels for the PIPS CAM detector. While the leading edges of the ZnS(Ag) pulse height spectra were separated by 70 channels for the 5.8 MeV $^{244}$Cm solution and a 4.8 MeV $^{235}$U/$^{238}$U solution. The energy calibration for the PIPS CAM detector had a larger variation in the number of channels than the ZnS(Ag) detector.

The published PIPS CAM pulse height spectra, Figure 2.14(a), and the experimental data in Figure 5.2(a) are essentially the same data but plotted differently. The only real differences between the data are the count times; in Figure 2.14(a) the count time was 10 minutes, while the count time for the data presented in Figure 5.2(a) was 10 hours. The y-axis in Figure 2.14(a) consisted of counts normalized to activity concentration.

$$\text{NetCounts per Channel}_B^* = \frac{\text{Concentration}_A \left( \frac{Bq}{ml} \right)}{\text{Concentration}_B \left( \frac{Bq}{ml} \right)}$$

(14)

The net counts in each channel from solution B were normalized to the concentration of solution A, the control solution. The authors of the original work assumed that the units of concentration (Bq/mL) cancelled when normalized, but the active volumes of the
detected solutions were not the same for the different alpha energies which led to a misleading comparison. The y-axis in Figure 5.2(a) was created using the net count rate divided by the effective activity. The effective activity was calculated from the solution concentration using the active volume of the solution.

Figure 5.2 Spectra of the aqueous solutions recorded with the (a) PIPS CAM detector and (b) ZnS(Ag) detector demonstrated the spectral shape.
Absolute Detection Efficiency

In the absence of beta and gamma radioactivity, the absolute detection efficiencies of the PIPS CAM detector were 20% higher than that of the ZnS(Ag) detector, Figure 5.3. To find the absolute detection efficiency (%), the net count rate was divided by the effective activity of the solution (Bq) and multiplied by 100. The net count rate was calculated by dividing the counts in the alpha region of interest (ROI) by the live time and subtracting the background count rate. The calculated absolute detection efficiencies of the 100-511 ROI pulse height spectra measured with the PIPS CAM detector were $9.01 \pm 0.02\%$, $8.63 \pm 0.02\%$, $7.95 \pm 0.01\%$, and $7.15 \pm 0.01\%$ for $^{244}\text{Cm}$, $^{241}\text{Am}$, $^{239}\text{Pu}$, and $^{230}\text{Th}$ solutions, respectively. The absolute detection efficiencies are different from the counting sensitivities listed in Chapter 2 as the active depths were used in the above calculations. The absolute detection efficiencies of the ZnS(Ag) pulse height spectra, 20-511 ROI, were $7.55 \pm 0.14\%$, $6.08 \pm 0.02\%$, and $4.96 \pm 0.03\%$ for the $^{244}\text{Cm}$, $^{239}\text{Pu}$, and $^{234}\text{U}/^{238}\text{U}$ solutions, respectively.

![Graph showing absolute detection efficiency vs. alpha energy](image)

**Figure 5.3** The absolute detection efficiency for the PIPS CAM and ZnS(Ag) detectors for alpha radioactivity in aqueous solutions.
Beta and gamma radioactivity, prevalent in high-level waste, interfered with the alpha region of interest of both detectors despite minimal beta and gamma detection efficiencies. The absolute detection efficiencies were calculated as $(1.0 \pm 0.1) \times 10^{-2}\%$ and $(5.7 \pm 1.5) \times 10^{-5}\%$ for $2\,\text{mL}$ of $^{90}\text{Sr}^{(90)}\text{Y}$ beta and $^{137}\text{Cs}$ gamma solutions, respectively, for the ZnS(Ag) detector with a region of interest of 5-511. Although the absolute detection efficiencies for beta and gamma radioactivity were low, they remain significant due to the high activity levels present in HLW. The absolute detection efficiency of beta particles were determined to be higher than the absolute detection efficiency of gamma rays as beta particles lose energy more readily in absorption layers. Only a fraction of the incident gamma rays interacted in the thin ZnS(Ag) layer. The $^{90}\text{Sr}^{(90)}\text{Y}$ pulse height spectrum, measured with the ZnS(Ag) detector, was located at channels less than 50 while the $^{137}\text{Cs}$ gamma radioactivity was located at channels lower than 15, Figure 5.4. The beta and gamma pulse height spectra were located in the low-energy region of interest which allowed for a beta and gamma upper level discriminator (ULD) to be set at 55 channels. The beta and gamma pulse height spectra for the PIPS CAM detector in Figure 2.14(b) more severely spilled over into the alpha region of interest which resulted in the highest alpha detection efficiency. The beta and gamma pulse height spectra, found in channels as high as 500 (re-binned channel 250), significantly reduced the alpha detection region of interest of the PIPS CAM pulse height spectra, Figure 5.2(a), to channels above 550 (re-binned channel 275). The low beta and gamma counting sensitivity of the PIPS CAM detector in Table 2.3, $4.06 \times 10^{-4}$ and $4.69 \times 10^{-6}\%$, for $^{90}\text{Sr}^{(90)}\text{Y}$ and $^{137}\text{Cs}$, respectively, was calculated as having absolute detection efficiencies of $8.12 \times 10^{-2}\%$ and $9.38 \times 10^{-4}\%$, respectively. The absolute detection
efficiencies were found as the entire detection volume, 2 mL, was utilized in both the PIPS CAM and ZnS(Ag) experiments in the detection of beta and gamma radioactivity. Despite the absolute detection efficiencies, an alpha region of interest (275-511) was chosen for the PIPS CAM detector to minimize interference from beta and gamma radioactivity while maximizing the PIPS CAM alpha detection efficiency.

![Graph](image)

**Figure 5.4** Beta particle and gamma ray interference in the ZnS(Ag) detector.

In the presence of beta and gamma radioactivity, the ZnS(Ag) detector had a 40% better detection efficiency relative to the PIPS CAM detector. The absolute detection efficiencies for the PIPS CAM detector of the reduced alpha ROI (channel 275-511) were $4.61 \pm 0.01\%$, $3.87 \pm 0.01\%$, $2.84 \pm 0.01\%$, and $1.32 \pm 0.01\%$ for $^{244}\text{Cm}$, $^{241}\text{Am}$, $^{239}\text{Pu}$, and $^{230}\text{Th}$, respectively, Figure 5.3. The absolute detection efficiencies for the ZnS(Ag) detector for the reduced region of interest (channel 55-511) were $7.19 \pm 0.13\%$, $5.37 \pm 0.02\%$, and $4.21 \pm 0.03\%$ for $^{244}\text{Cm}$, $^{239}\text{Pu}$, and $^{234}\text{U}/^{238}\text{U}$, respectively. The narrowed alpha regions of interests which minimized the spillover of the beta and gamma events resulted in lower detection efficiencies for both detectors. The absolute detection
efficiencies of the PIPS CAM detector were reduced more significantly than the ZnS(Ag) efficiencies because the beta and gamma ULD occurred at a higher channel.

In the absence of beta and gamma radiation, the PIPS CAM detector was better suited for gross alpha counting than the ZnS(Ag) detector due to decreased detection efficiency energy dependence. Higher-energy alpha particles were more efficiently detected with both detectors than lower-energy particles as shown in Figure 5.3. The slope of the efficiency calibration curve for the PIPS CAM, 1.71, however, was less than that for the absolute detection efficiency of the ZnS(Ag) detector, 2.26, in Figure 5.3. The PIPS CAM detector possessed a lower energy dependence than the ZnS(Ag) detector making it more suitable for the gross detection of alpha particles from a solution which contained several actinides. The PIPS CAM detector also appeared to be a more consistent detector as the linear fit fell within uncertainty for the data points and the regression estimate had a higher correlation coefficient. The increased uncertainty of the ZnS(Ag) detector was attributed the low-activity natural uranium ($^{234}$U/$^{238}$U) solution as total alpha activity was used in the calculation but the energy was assumed to be the average of the two main isotopes, $^{234}$U and $^{238}$U.

In the presence of beta and gamma radioactivity, the energy dependence of the PIPS CAM absolute detection efficiency increased while the ZnS(Ag) energy dependence stayed the same. The change of slope for the absolute detection efficiencies of the ZnS(Ag) detector, 2.26, was less than that for the PIPS CAM absolute detection efficiencies, 2.98, Figure 5.3. High-energy alpha particles, $^{244}$Cm and $^{241}$Am, were more easily detected with the PIPS CAM detector because a greater portion of the pulse height spectrum was located above the ULD of the beta and gamma region of interest. The
lower-energy alpha particles, $^{239}\text{Pu}$ and $^{230}\text{Th}$, were more greatly affected as the bulk of the pulse height spectra was located in the region below the ULD of the beta and gamma region of interest. The beta and gamma radioactivity were located low enough on the pulse height spectra of the ZnS(Ag) detector that the alpha region of interests for the various radionuclides were equally affected.

**Solution Concentration**

$^{241}\text{Am}$ and $^{230}\text{Th}$ solutions were used to determine the relationship between activity concentration and the net count rate of the ZnS(Ag) detector in conjunction with the signal splitter using the 4.48 cm$^2$ sample holding cup. A solution of $^{241}\text{Am}$, 13,896 Bq/mL, was diluted to 6,948, 4,627, and 2,084 Bq/mL. A $^{230}\text{Th}$ solution, 35,529 Bq/mL, was diluted to 17,765, 11,831, and 5,329 Bq/mL. A sample of nitric acid was used as a blank solution. The non-serial dilutions were conducted by diluting samples of the original solution to the appropriate concentration. The data in Figure 5.5 suggested a linear response of count rate with increased concentration. The $^{241}\text{Am}$ and $^{230}\text{Th}$ solutions have different slopes for net count rate versus solution concentration in Figure 5.5 due to differences in detection efficiency. The $^{241}\text{Am}$ solution possessed a greater slope than the $^{230}\text{Th}$ solution due to the higher detection efficiency of the high-energy alpha particles. The absolute detection efficiencies for the $^{241}\text{Am}$ and $^{230}\text{Th}$ solutions, 8.42 and 6.14%, were slightly higher than those previously presented due to differences between the signal splitter and preamplifier in the experimental set-up. The PIPS CAM detector demonstrated a similar linear relationship between count rates and the varied activity concentrations of $^{244}\text{Cm}$, $^{241}\text{Am}$, and $^{230}\text{Th}$ as observed in Figure 2.15. The PIPS
CAM detector also demonstrated differences in the absolute detection efficiency through the variation of slopes in Figure 2.15.

\[ y = 1.8796x - 0.0196 \quad R^2 = 0.9999 \]
\[ y = 1.0447x + 0.0918 \quad R^2 = 0.9997 \]

Figure 5.5 The net count rate of the ZnS(Ag) detector plotted against various concentrations of $^{241}$Am and $^{230}$Th solutions.

**Detection Area**

One of the advantages of the Ludlum Model 43-1 ZnS(Ag) detector over the PIPS CAM detector was the size of the detection area. The ZnS(Ag) detector in conjunction with the charge-sensitive preamplifier had a detection surface which was previously constricted for comparison with the PIPS CAM detector. The detection surface was uncovered to utilize the larger detection surface. The 11.3 ± 0.02 Bq/mL depleted uranium ($^{238}$U) solution was detected with surface areas of 4.15 cm$^2$, 12.8 cm$^2$, 49.4 cm$^2$, and 78.5 cm$^2$. The surface area of 4.15 cm$^2$ resulted in a minimum detectable concentration that was greater than 11.3 Bq/mL. A large sample holding cup and absorption shields were utilized to keep the surface area of the aqueous solution and the detection surface consistent. The absolute detection efficiency increased with increasing detection area as shown in Figure 5.6. The increased surface area of the aqueous solution
was the only way to increase the sample volume due to the small range of alpha particles in solution. The absolute detection efficiencies for the various detection areas, 12.8 cm$^2$, 49.4 cm$^2$, and 78.5 cm$^2$ were 3.29 ± 1.24%, 4.71 ± 0.34%, and 6.30 ± 0.24%, respectively for $^{238}$U. The absolute detection efficiency of the 78.5 cm$^2$ ZnS(Ag) detection area almost doubled the absolute detection efficiency of the 12.8 cm$^2$ detection area. The absolute detection efficiency for the largest detection surface had the least uncertainty as the associated count rates were higher. The extrapolation of Figure 5.6 also possessed uncertainty values of more than 1% for the 4.15 cm$^2$ detection area. The increased count rates resulted in increased absolute detection efficiency and counting statistics while uncertainty for the count rate was decreased.

![Graph](image)

**Figure 5.6** The Ludlum Model 43-1 ZnS(Ag) detector was used in the detection of three detection surface areas of depleted uranium solution.

**Minimum Detectable Concentration**

The minimum detectable concentration of the ZnS(Ag) detector decreased with increasing absolute detection efficiency and detection area. The minimum detectable
concentrations for the ZnS(Ag) detector in conjunction with the charge-sensitive preamplifier, Table 5.1, were calculated at 33.42, 50.39 and 79.38 Bq/mL for $^{244}$Cm, $^{239}$Pu, and $^{234}$U/$^{238}$U in a 4.15 cm$^2$ sample holding cup, respectively. The minimum detectable concentrations were calculated using equation 12, however, the $\varepsilon$ represented the effective absolute detection efficiency times the active volume instead of counting sensitivity. The gross background count for the 20-511 ROI was 54 counts in 600 seconds and the absolute detection efficiencies were 7.55, 6.08, and 4.96% for the $^{244}$Cm, $^{239}$Pu, and $^{234}$U/$^{238}$U solutions, respectively. The variations in the minimum detectable concentration for the radionuclides were due to the changes in the absolute detection efficiency for the different radionuclides. The minimum detectable concentrations of the PIPS CAM detector were published as 17.7, 23.0, and 35.0 Bq/mL for $^{244}$Cm, $^{241}$Am, and $^{239}$Pu, respectively [18]. The PIPS CAM minimum detectable concentrations were lower than those for the ZnS(Ag) detector due to the higher absolute detection efficiency of the PIPS CAM detector in the absence of beta and gamma radioactivity and higher background count rate attributed to suspected $^{241}$Am contamination present on the Mylar® surface of the ZnS(Ag) detector.

The minimum detectable concentration for the maximum surface area of the ZnS(Ag) detector used in these experiments, 78.5 cm$^2$, for 3600 seconds was the lowest at 0.28 Bq/mL for $^{238}$U, Table 5.1. The minimum detectable concentrations for the smaller areas, 49.4 cm$^2$ and 12.8 cm$^2$, were 0.60 Bq/mL and 3.31 Bq/mL, respectively. The projected minimum detectable concentration for the 4.15 cm$^2$ sample holding cup was calculated as 11.86 Bq/mL for $^{238}$U, greater than the solution concentration used. The minimum detection concentration decreased with increasing detection area.
The largest minimum detectable concentration of the ZnS(Ag) detector was 79.38 Bq/mL, a value over 10 times less than the maximum concentration allowed in the Saltstone facility at the Savannah River Site [1]. The MDC for the entire detection face of the Ludlum 43-1 ZnS(Ag) detector was 0.28 Bq/mL, over 1,000 times less than the maximum concentration. The ZnS(Ag) detector was shown to be capable of detecting alpha activity from HLW solutions.

**Table 5.1** The minimum detectable concentration (MDC) for the various areas and radionuclides.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Detection Area (cm²)</th>
<th>Time (s)</th>
<th>MDC (Bq/mL)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{244}$Cm</td>
<td>4.15</td>
<td>600</td>
<td>33.42</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>4.15</td>
<td>600</td>
<td>50.39</td>
</tr>
<tr>
<td>$^{234}$U/$^{238}$U</td>
<td>4.15</td>
<td>600</td>
<td>79.38</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>4.15</td>
<td>3600</td>
<td>11.86</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>12.8</td>
<td>3600</td>
<td>3.31</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>49.4</td>
<td>3600</td>
<td>0.60</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>78.5</td>
<td>3600</td>
<td>0.28</td>
</tr>
</tbody>
</table>

**Solution Density**

The density of the $^{241}$Am solution was increased with sodium nitrate salts in order to demonstrate the effects of the high salt content in HLW on the detection capabilities of the ZnS(Ag) detector in conjunction with the signal splitter. The normalized count rate over concentration as obtained with the ZnS(Ag) detector decreased as the solution density increased as shown in Figure 5.7. The normalized count rate over concentration, or relative efficiency as referred to in Chapter 2, assumed unity at 1.01 g/mL as both the count rate and concentration were normalized to the original 1.01 g/mL density solution. The increased solution density decreased the range of alpha particles in solutions resulting in decreased detection. Both the ZnS(Ag) detector, Figure 5.7, and the PIPS
CAM detector, Figure 2.16, demonstrated decreased count rates with the increased solution density. The ZnS(Ag) detector and the PIPS CAM detector had similar slopes of -0.56 and -0.57, respectively, as the relationship was dependent on the density of the solution and independent of the detector. The data taken with the PIPS CAM detector had greater uncertainty because several actinide solutions were plotted and the differences in absolute detection efficiency and alpha range were not taken into account. The actinide solutions in both experiments were in the absence of beta and gamma radioactivity.

The absolute detection efficiency of the ZnS(Ag) detector did not decrease despite the increase of the $^{241}$Am solution density. The absolute detection efficiency was calculated using the effective activity of the solution in the active volume of the sample. The active depth of the alpha particle, a function of density, decreased as the solution density increased as demonstrated by equations 1, 2, and 3. The active depth of the alpha particle in solution decreased from 49.9 µm for the original solution density of 1.01 g/mL to 39.8 µm for the 1.27 g/mL solution. Alpha particles were more greatly absorbed in the denser matrix. The decreased active depth reduced the volume and therefore, activity, exposed to the detector. The absolute detection efficiency did not have a declining relationship with the solution density because the reduced activities accounted for the density differences. The absolute detection efficiencies for the $^{241}$Am solutions of various densities were fairly constant as measured with the ZnS(Ag) detector, Figure 5.8.

The direct detection of a solution with the ZnS(Ag) detector provided a straightforward relationship between the detector and the chemistry of the solution. Components of liquid waste, such as salts and organics, would be treated as a single entity as they
effect detection through changes in density. Insoluble particles which float on the top of
the waste would have a negative impact on the method of direct detection along with
non-homogeneous mixtures. Although density effects could be correct for, it is often an
unknown parameter as the density of waste varies. A 30% decrease in count rates existed
between the 1.01 g/mL and 1.27 g/mL $^{241}$Am solutions. The decrease in the count rate
could be minimized by using an average density of HLW; however, uncertainty
pertaining to density could not be abolished.

\[ y = -0.5559x + 1.5544 \]
\[ R^2 = 0.9772 \]

**Figure 5.7** The normalized count rate over concentration of the ZnS(Ag) detector versus
the density of the $^{241}$Am solution.
Figure 5.8 The absolute detection efficiency of the ZnS(Ag) detector plotted against the increasing density of the $^{241}\text{Am}$ solution.

Air Gap Separation

The air gap between the ZnS(Ag) detector in conjunction with the signal splitter and the liquid surface was varied to determine the optimum distance. The distance between the detector and the surface of the $^{241}\text{Am}$ and $^{230}\text{Th}$ solutions exhibited an exponential relationship with the absolute detection efficiency, Figure 5.9. The monotonic decrease was expected given the variations in solid angle have no bearing on the calculation of the absolute detection efficiency and the absorber thickness of the air increased. The active depth of the solution was not calculated as a function of the air thickness and remained constant with increasing distance. Physically, however, the maximum depth of the alpha particles in solution decreased with increasing absorption thickness which resulted in less activity from the 4.48 cm$^2$ sample holding cup.
Figure 5.9 The absolute detection efficiency decreased as the air gap distance between the ZnS(Ag) detector and $^{241}\text{Am}$ and $^{230}\text{Th}$ solutions increased.

Contact between the Mylar® layer and aqueous solution would result in the highest absolute detection efficiency possible, however, due to the caustic properties of HLW the absence of an air gap was not practical. An air gap distance of 1.2 mm between the Mylar® surface and the $^{241}\text{Am}$ and $^{230}\text{Th}$ solutions resulted in absolute detection efficiencies of $12.94 \pm 0.06\%$ and $10.95 \pm 0.04\%$, respectively, and preserved the integrity of the Mylar® surface. The PIPS CAM detector demonstrated a similar exponential relationship of count rate as a function of increased air gap distance, Figure 2.18.

Thin versus Thick Scintillation Layer

The ZnS(Ag) detector lacked total alpha energy deposition due to the thin scintillation layer compared to total energy deposition in the PIPS CAM detector. The scintillation layer present on the ZnS(Ag) detector did not theoretically absorb the total alpha energy incident on the detector. The maximum range of a $^{239}\text{Pu}$ alpha particle through ZnS(Ag) scintillation crystals was estimated as 8.56 mg/cm$^2$ with equations 1, 2,
and 3. The estimated range was greater than the thin scintillation layer of the ZnS(Ag) detector, 3.5 mg/cm², however, shorter than a thicker ZnS(Ag) scintillation layer, 14 mg/cm². The thin detection layer of the ZnS(Ag) detector represented a ΔE detector as opposed to an E detector, with the remaining energy of the alpha particle deposited on the light pipe directly behind the ZnS(Ag) layer. The detector effectively detected every incident alpha particle but did not properly quantify the amount of energy that the alpha particle possessed. The maximum range of a $^{239}$Pu alpha particle in a PIPS CAM detector was calculated to be 27.5 µm—much less than the 120 µm depletion depth. The PIPS CAM detector possessed an infinitely thick detection layer relative to the range of an alpha particle in the media allowing the detector to quantify the total alpha energy deposited. The range calculations did not incorporate energy absorption in the air, Mylar®, or aluminized polymer layers.

The absolute detection efficiencies for the thick ZnS(Ag) scintillator layer were compared to the thin ZnS(Ag) layer in the detection of alpha radioactivity in aqueous solutions. The absolute detection efficiencies of the $^{244}$Cm, $^{239}$Pu, and $^{234}$U/$^{238}$U solutions detected using the two ZnS(Ag) scintillation layers were shown to be equivalent, Figure 5.10. The absolute detection efficiencies were expected to be the same for the two scintillation layers as the solid angle was maintained. Although the thin scintillation layer was a ΔE detector and did not quantify the total energy of the alpha particle, the ZnS(Ag) detector was able to detect each particle which was incident on the detection surface.
Figure 5.10 The absolute detection efficiency of the ZnS(Ag) for the thick, 14 mg/cm$^2$, and thin, 3.5 mg/cm$^2$, ZnS(Ag) scintillation layers for aqueous solutions.

The pulse height spectra of a $^{239}$Pu electroplated standard obtained with the thin and thick ZnS(Ag) scintillation layers were dissimilar. The ZnS(Ag) detector with a 14 mg/cm$^2$ scintillation layer resulted in a pulse height spectrum located in the lower energies and with greater peak resolution than the 3.5 mg/cm$^2$ scintillation layer produced, Figure 5.11. The peak maxima of the pulse height spectra were located at 184 and 230 channels for thick and thin scintillation layers, respectively. The peak energy resolutions for the pulse height spectra of the electroplated standard for the thick and thin layers were 63 and 118%, respectively.
The pulse height spectra of $^{244}\text{Cm}$, $^{239}\text{Pu}$, and $^{234}\text{U}/^{238}\text{U}$ aqueous solutions detected with the thicker scintillation layer on the ZnS(Ag) detector in conjunction with the charge-sensitive preamplifier had a different spectral shape and were located at lower channels than the spectra recorded with the thin scintillation layer. The leading edges of the pulse height spectra (FWTM) for the thick ZnS(Ag) scintillation layer, Figure 5.12, were 288, 246, and 211 channels for $^{244}\text{Cm}$, $^{239}\text{Pu}$, and $^{234}\text{U}/^{238}\text{U}$, respectively. The leading edges of the thin ZnS(Ag) scintillation layer, Figure 5.2(b), for the $^{244}\text{Cm}$, $^{239}\text{Pu}$, and $^{234}\text{U}/^{238}\text{U}$ aqueous solutions were 400, 363, and 330 channels, respectively. The leading edges of the pulse height spectra were located at lower channels and had more channels between the leading edges of the various solutions for the thicker scintillation layer. An increase in the energy dependence was expected for the thicker scintillation layers since all the alpha energy was deposited in the detector. The spectral shape of the pulse height spectra of the thick scintillation layer for the aqueous solutions, Figure 5.12, demonstrated better energy quantification as it had defined leading edges and closer
resembled the continuous energy distribution than that of the thin scintillation layer, Figure 5.2(b).

**Figure 5.12** The pulse height spectra for the thick, 14 mg/cm², scintillation layer on the ZnS(Ag) detector.

A light collection phenomenon was responsible for the location of the pulse height spectra from the thicker ZnS(Ag) scintillation layer in the lower channel region in comparison to the thin scintillation layer. The ZnS(Ag) crystals produced light intensity proportional to the incident alpha energy, however, the light was absorbed and scattered by the ZnS(Ag) crystals. The intensity of the light measured with the photomultiplier tube was decreased after scattering and absorption. The absorption of the scintillated light by the ZnS(Ag) was represented by the extinction coefficient. The peak emission wavelength of ZnS(Ag), 450 nm, or a photon energy of 2.76 eV corresponded to an estimated extinction coefficient of 0.025 from Figure 2.5. The intensity of the light, equation 9, transmitted by the ZnS(Ag) crystals was 94.2% for the 3.5 mg/cm² scintillation layer. The thicker scintillation layer, 14 mg/cm², transmitted 78.7% of the original light intensity created by the ZnS(Ag) crystal, significantly less than the thin
layer. The maximum recommended thickness of ZnS(Ag), 25 mg/cm², only transmitted 65.3% of the scintillated light.

**Computer Simulation**

A computer simulation was created in Visual Basic to demonstrate the finite thickness of the 3.5 mg/cm² ZnS(Ag) scintillation layer and the infinitely thick detection layer of the PIPS CAM detector. The Monte Carlo computer model simulated the experimental set-up, but the sample holding cup and detector face were approximated as a square with the same surface area as the round apparatus. The radionuclides were randomly distributed throughout the volume in the sample holding cup and the alpha particle directions were randomly generated. The active volume of the solution simulated an infinitely thick source by using the maximum range of the alpha particle in solution according to equations 1, 2, and 3. The computer code differentiated between the ZnS(Ag) detector and the PIPS CAM detector in physical variables for the differences in detector thickness and protective coatings over the detection surface. The ZnS(Ag) detector had 3.5 and 14 mg/cm² detection layers while the PIPS CAM had an infinitely thick detection depth. The ZnS(Ag) detector had a 0.8 mg/cm² Mylar® layer while the PIPS CAM detector possessed a 0.5 µm aluminum and varnish layer with a total absorption equivalent to 1.0 µm of silicon, 0.37 mg/cm², for the PIPS CAM detector. The protective layers were physical barriers which decreased the alpha particle energy while protecting the detection surface and excluding light from the apparatus. Differences between the scintillation and solid state detection properties for the detectors were not incorporated into the program.
The Visual Basic computer model was created to simulate the loss of alpha energy in various absorption layers and to display the spectral shape of the theoretical pulse height spectra. A combination of the Bethe equation, equation 5, and extrapolation from experimental data [31] were used to calculate the energy loss of the alpha particle for each layer of the detection system: solution, air, Mylar®, and ZnS(Ag). The alpha particle energy continuously decreased as it transmitted through the absorption layers. The stopping power, \(-dE/dx\), was a function of the alpha velocity which was dependent on alpha energy for the Bethe equation. The experimental extrapolation for stopping power in water was dependent only upon the alpha energy. The standard-known alpha energy was initially used to calculate the initial velocity of the alpha particle. A Visual Basic loop was created in order to calculate the stopping power for each section of the absorption layers. Each absorption layer was divided into 100 sections and the energy lost by the alpha particle was calculated over the dx of each absorption layer as shown in the flowchart, Figure 5.13. The alpha particle velocity was then computed for a given dx step and input back into the source code. The energy of the alpha particle as it left each absorption layer was returned to the worksheet. A histogram of the difference between the alpha energy entering and exiting the ZnS(Ag) layer produced the pulse height spectrum for the detection system. The PIPS CAM detection layer was programmed to absorb the total alpha energy. The program processed 19992 Monte Carlo trials for each run. A more comprehensive explanation of the computer simulation is detailed in Appendix B.
Figure 5.13 The flow chart of the Visual Basic computer simulation of the alpha energy deposition in various absorption layers.
The Monte Carlo computer model was benchmarked against published data and shown to be in good agreement for high alpha energies. The computer-simulated stopping power of water for alpha particles was plotted in Figure 5.4 from 0.15 to 200 MeV using the Bethe equation. The stopping power below 0.15 MeV was based on the experimental data of Ziegler from the NIST ASTAR database [31]. Table 5.2 displays the stopping power of water for higher-energy alpha particles from the computer-simulated data and values obtained from the NIST ASTAR database [31]. The NIST ASTAR values were derived using the Bethe equation for alpha values above 2 MeV and were fit to Ziegler’s experimental data below 2 MeV [31]. The percent difference between the computer-simulated and NIST ASTAR stopping power of water for alpha particles was less than 7.86% for alpha energies between 1 and 6 MeV. The computer-simulated stopping power values for low-energy alpha particles were not benchmarked against the NIST ASTAR database as the data were interconnected. The experimental data was incorporated into the computer simulation as the stopping power values for alpha energies below 0.11 MeV, as found with the Bethe equation, were negative as shown in Figure 5.14.
The computer-simulated stopping power of water for alpha particles.

Table 5.2 A comparison of the computer-simulated stopping power of water for alpha particles to theoretical values obtained from the NIST ASTAR database [31].

<table>
<thead>
<tr>
<th>Alpha Energy (MeV)</th>
<th>Computer-simulated Stopping Power of Water (MeV/cm)</th>
<th>Theoretical Stopping Power of Water (MeV/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>2.38 x 10^{3}</td>
<td>2.193 x 10^{4}</td>
</tr>
<tr>
<td>4.0</td>
<td>9.73 x 10^{2}</td>
<td>1.035 x 10^{3}</td>
</tr>
<tr>
<td>5.0</td>
<td>8.27 x 10^{2}</td>
<td>8.855 x 10^{2}</td>
</tr>
<tr>
<td>6.0</td>
<td>7.22 x 10^{2}</td>
<td>7.777 x 10^{2}</td>
</tr>
<tr>
<td>200</td>
<td>4.13 x 10^{1}</td>
<td>4.952 x 10^{1}</td>
</tr>
</tbody>
</table>

The theoretical pulse height spectra for the infinitely thick detection layer of the PIPS CAM detector and the finite scintillation thickness of the ZnS(Ag) detector were simulated for a point radioactive source with the computer modeling program. The theoretical alpha energy deposited in the 3.5 mg/cm^{2} ZnS(Ag) layer and an infinitely thick PIPS CAM detector are shown in Figure 5.15 for a $^{239}\text{Pu}$ point source. The pulse height spectrum for the PIPS CAM detector was a graph of the number of simulated alpha particles incident to the detection surface divided by the total counts for each
energy bin, Figure 5.15. The leading edge, 4.4 MeV, of the simulated pulse height spectrum for the PIPS CAM detector had a lower energy than the 5.147 MeV $^{239}$Pu alpha particles. The $^{239}$Pu alpha particles had a reduced energy and demonstrated tailing after the particle passed through 0.5 cm of air and the CAM protective coatings. The simulated pulse height spectrum for the $^{239}$Pu electroplated standard, Figure 5.15, demonstrated the finite detection ability of the 3.5 mg/cm$^2$ ZnS(Ag) detector as the bulk of the energy deposited in the scintillation layer was between 2.4 and 3.2 MeV. The leading edge of the pulse height spectrum was not located at the maximum alpha energy calculated to reach the detection layer after absorption through the air and Mylar® layer, 4.0 MeV. The alpha energy was not totally absorbed in the ZnS(Ag) layer which allowed for 4.0 MeV alpha particles to deposit only a fraction of their total energy. The maximum alpha energy at the detection surface of the ZnS(Ag) detector was lower than that of the PIPS CAM detector as the Mylar®, 0.8 mg/cm$^2$, was thicker than the aluminum and varnish coatings, 0.37 mg/cm$^2$.

![Graph](image)

**Figure 5.15** The computer-simulated pulse height spectra for the ZnS(Ag) finite scintillation layer and the infinitely thick detection surface of the PIPS CAM detector for a $^{239}$Pu point source.
The pulse height spectra for the infinitely thick detection layer of the PIPS CAM detector and thin scintillation thickness of the ZnS(Ag) detector were simulated for $^{239}$Pu aqueous solution with the computer model. The computer simulation of the PIPS CAM pulse height spectrum for the aqueous $^{239}$Pu solution was shown in Figure 5.16 and was consistent with the theoretical continuous energy distribution present at infinitely thick sources, Figure 2.19. The leading edge of the energy continuum represented the maximum alpha energy which entered the detection surface—4.4 MeV, a value which corresponded to the leading edge of the simulated pulse height spectrum for the $^{239}$Pu point source in Figure 5.15. The pulse height spectrum had higher probability of detection at high energies as most of the detected particles originated at the surface or directly underneath the liquid surface which has a larger solid angle. The computer-simulated pulse height spectrum for the 3.5 mg/cm$^2$ layer of ZnS(Ag), Figure 5.16, showed a narrow peak for the aqueous $^{239}$Pu solution. High-energy alpha particles passed through the ZnS(Ag) layer without losing all of their energy, thus creating a large $\Delta E$ peak. The large, narrowed peak was a summation of alpha particles which deposited energies greater than 2.4 MeV to the ZnS(Ag) layer. Because the detector was functioning as a $\Delta E$ detector, the amount of energy an alpha particle deposited into a thin layer of ZnS(Ag) did not have a linear relationship to alpha energy. The exact value of energy deposited in material was dependent upon the velocity of the alpha particle. The lack of linearity was demonstrated in the Bragg Curve, the average specific ionization, Figure 2.2. The stopping power of the water increased with decreasing alpha energy as shown with the Bethe equation, Figure 5.14. A 4.0 MeV alpha particle, the maximum energy alpha particle after absorption from air and Mylar®, deposited 2.0 MeV into a 3.5
mg/cm$^2$ ZnS(Ag) layer while a 3.0 MeV alpha particle deposited 2.3 MeV in the ZnS(Ag) layer as calculated with the Bethe equation. The pulse height spectrum of the 3.5 mg/cm$^2$ ZnS(Ag) detector of Figure 5.16 portrayed the total deposition of low-energy alpha particles in the ZnS(Ag) layer. The low-energy alpha particles, a property of the continuous energy distribution from aqueous solutions, have their total energy deposited in the ZnS(Ag) layer. The pulse height spectrum for the finite detection layer converged with the low-energy tail of the pulse height spectrum of the PIPS CAM detector in Figure 5.16 as it represented complete detection.

**Figure 5.16** The computer-simulated pulse height spectra for the ZnS(Ag) finite scintillation layer and the infinitely thick detection surface of the PIPS CAM detector for a $^{239}$Pu aqueous solution.

The computer-simulated pulse height spectra for the PIPS CAM detector of the point source and aqueous solution, Figures 5.15 and 5.16, were in good agreement with the experimental spectral shape of Figure 5.1(a). The computer-simulated and experimental data have similar pulse height spectra for the $^{239}$Pu theoretical point source and $^{241}$Am electroplated standard, respectively, as both demonstrated a defined leading edge and tailing. The computer-simulated pulse height spectra of the $^{239}$Pu aqueous
solution detected with the PIPS CAM detector, Figure 5.16, demonstrated a continuous energy distribution also present in the experimental pulse height spectrum of the $^{241}$Am aqueous solution with PIPS CAM detector, Figure 5.1(a).

The computer-simulated and experimental pulse height spectra for the ZnS(Ag) detector did not demonstrate the same spectral shape because the scintillation and light properties were not simulated in the computer model. The narrow peak of the computer-simulated pulse height spectrum for the $^{239}$Pu point source, Figure 5.15, did not resemble the broad alpha peak of the experimental pulse height spectrum of the $^{239}$Pu electroplated standard detected with the ZnS(Ag) detector, Figure 5.1(b). The computer-simulated and experimental pulse height spectra of the $^{239}$Pu aqueous solution with the ZnS(Ag) detector as shown in Figure 5.16 and Figure 5.1(b), respectively, did not show similar spectra shaping. The poor resolution of the ZnS(Ag) detector was created from propagation of the scintillated light—a property absent in the PIPS CAM detector and not simulated in the program.

The computer-simulated absolute detection efficiencies of the $^{239}$Pu aqueous solution for the 3.5 and 14 mg/cm$^2$ ZnS(Ag) detector and the PIPS CAM detector were compared to the experimental data. The ZnS(Ag) and PIPS CAM detectors had computer-simulated absolute detection efficiencies of $3.10 \pm 0.1$ and $4.87 \pm 0.2\%$, respectively, for the 0.6 – 5.5 MeV ROI in the computer-simulated pulse height spectra of the $^{239}$Pu aqueous solution, Table 5.3. The thick and thin scintillation layers had the same absolute detection efficiency, a result consistent to the experiments. The differences in the protective layers of the equipment caused differences of detection efficiency as the Mylar® on the ZnS(Ag) detector was thicker and absorbed greater alpha
energy. The solid angles of the simulated detection set up were consistent between the two detectors. The experimentally determined absolute detection efficiencies were 6.08 $\pm$ 0.02%, 6.07 $\pm$ 0.02%, and 7.95 $\pm$ 0.01% for the thin and thick ZnS(Ag) detectors and the PIPS CAM detector, respectively. The computer-simulated values of the absolute detection efficiency were lower than the experimental absolute detection efficiencies for the ZnS(Ag) and PIPS CAM detectors.

The computer-simulated pulse height spectra of the aqueous solutions revealed a high-count data point in the lower energy region. The probability of detection of the 0-0.4 MeV ROI in Figure 5.16 was 2.3 $\pm$ 0.1%, which was cropped out of the computer-simulated PIPS CAM pulse height spectrum. The pulse height spectra in the low-energy region of interest were not accessible in the experimental ZnS(Ag) data as the gain was too low. The low-energy region of the PIPS CAM spectrum was difficult to examine in the experimental data due to large noise peaks; however, the excess of counts in the low-energy region of the computer-simulated spectrum is inconsistent with the theory of continuous distribution. The low-energy alpha particles were attributed to inconsistencies of the Bethe equation and Ziegler extrapolation as seen in Figure 5.14 and also to unknown imperfections in the computer model. The computer-simulated absolute detection efficiency for the 0-5.5 MeV ROI for the ZnS(Ag) detector and the PIPS CAM detector were 3.85 $\pm$ 0.1 and 7.22 $\pm$ 0.2%, respectively, and are more consistent with the experimental values. The differences between the absolute detection efficiencies of the PIPS CAM detector and ZnS(Ag) detector were expected due to the differences in the thickness protective layers as demonstrated by Phoenix et al., in Figure 2.11 [15]. The thinner protective layer over a detector had higher count rates in the study.
Table 5.3 The absolute detection efficiencies (%) for $^{239}$Pu aqueous solutions for the ZnS(Ag) detector and PIPS CAM detector experiments and computer simulations.

<table>
<thead>
<tr>
<th>Detector</th>
<th>Experimental Absolute Detection Efficiency (ROI 0.6-5.5 MeV)</th>
<th>Simulated Absolute Detection Efficiency (ROI 0-5.5 MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thin ZnS(Ag)</td>
<td>6.08 ± 0.02</td>
<td>3.10 ± 0.1</td>
</tr>
<tr>
<td>Thick ZnS(Ag)</td>
<td>6.07 ± 0.02</td>
<td>3.10 ± 0.1</td>
</tr>
<tr>
<td>PIPS CAM</td>
<td>7.95 ± 0.01</td>
<td>4.87 ± 0.2</td>
</tr>
</tbody>
</table>

A channel shift to the higher energies occurred when the ZnS(Ag) scintillation thicknesses were increased in the computer-simulated pulse height spectra. The pulse height spectra for the $^{239}$Pu point source were simulated using both the 3.5 mg/cm$^2$ and the 14 mg/cm$^2$ ZnS(Ag) scintillation layers, Figure 5.17(a). In the computer-simulated spectra for the point source, the pulse height spectrum for the thicker scintillation layer was located at higher energies than the pulse height spectrum for the thinner layer. The thicker scintillation layer was capable of greater alpha energy deposition than the thinner scintillation layer and was able to register higher alpha energies. The peak resolution improved and the tailing from the air and Mylar$^\text{®}$ absorption layers was present in the pulse height spectrum for the point source as detected with the thicker scintillation layer. The simulated pulse height spectrum for the $^{239}$Pu aqueous solution, Figure 5.17(b), illustrates the same shift in channels. The thicker scintillation layer, 14 mg/cm$^2$, had a higher energy leading edge and closer resembled the continuous energy distribution. The leading edge of the pulse height spectra for the thicker scintillation layer was located at 3.6 MeV, lower than the maximum alpha energy, 4.0 MeV, which was incident to the scintillation layer. The 14 mg/cm$^2$ ZnS(Ag) detector was thicker than the 3.5 mg/cm$^2$ scintillation layer and had better energy detection capabilities. The 14 mg/cm$^2$ ZnS(Ag) layer did not have infinite detection capabilities in the computer model, despite
expectations based on the range-energy relationship. The lack of complete detection is believed to lie within imperfections of the model. Alpha particles were transmitted through the thicker scintillation layer on the ZnS(Ag) detector at 3.27%, a value inconsistent with theory and believed to be an unknown function of the computer simulation. The range-energy theory for alpha particles determined that no $^{239}$Pu alpha particle should have enough energy to be transmitted through the 14 mg/cm$^2$ ZnS(Ag) scintillation layer. The computer-simulated pulse height spectra for the thin versus thick scintillation layers were different from the experimental pulse height spectra, Figures 5.11 and 5.12, as the computer model did not simulate light propagation and collection properties.
Figure 5.17  The pulse height spectra for a (a) $^{239}$Pu point source and (b) $^{239}$Pu aqueous solution simulated for the thin, 3.5 mg/cm$^2$, and thick, 14 mg/cm$^2$, ZnS(Ag) scintillation layers.
CHAPTER 6
CONCLUSIONS

The ZnS(Ag) detector and PIPS CAM detector can both effectively detect alpha particles directly from the surface of aqueous solutions. The detectors are both applicable of detection of alpha radioactivity in high-level waste as the minimum detectable concentration for each is lower than the concentration expected in process streams. The ZnS(Ag) detector and the PIPS CAM detector would both make successful on-line alpha detectors, but under different conditions. The ZnS(Ag) detector had an advantage over the PIPS CAM detector as beta and gamma interactions were successfully discriminated against while still maintaining good alpha detection efficiency. The ZnS(Ag) detector also had the advantage of having a larger detection surface which further increased the detection efficiency and decreased the minimum detectable concentration. The PIPS CAM detector proved to be the better detector in the absence of beta and gamma radiation due to increased detection efficiency and decreased energy dependence.

The ZnS(Ag) and PIPS CAM detectors were affected by experimental systematic errors in the energy dependence and density effects on detection efficiency. The absolute detection efficiencies for the different alpha energies varied as much as 41% for the ZnS(Ag) detector and 71% for the PIPS CAM detector in the presence of beta and gamma radioactivity. The increased density of the solution, a physical property independent of the detectors, resulted in a decrease in the absolute detection efficiency. Density effects contributed an additional 30% variation in absolute detection efficiency between the 1.01 and 1.27 g/mL solutions. The combination of the effects from absolute
detection efficiency and density created total systematic errors of approximately 70% and 100% for the ZnS(Ag) and PIPS CAM detectors, respectively. The systematic error was maximized by taking the most extreme values; however, steps could be taken to reduce the error by calibrating the detection system. The average alpha energies and density of HLW could be incorporated into the system to improve the accuracy of the detectors.

The pulse height spectra of the ZnS(Ag) detector were obtained for two different detection thicknesses and compared to the PIPS CAM detector, computer simulations, and theory. The poor resolution of the ZnS(Ag) detector severely limited the capabilities of the detector to quantify alpha energy; however, the PIPS CAM detector had excellent resolution leaving room for possibilities of energy de-convolution based on the leading edge of the alpha pulse height spectrum. The thin scintillation layer produced a stronger signal than the thick ZnS(Ag) layer as the light intensity was not diminished from absorption and scattering in the ZnS(Ag) layer.
APPENDICES
Appendix A
High Voltage Optimization

The optimal high voltage was determined for the ZnS(Ag) detector and signal splitter which maximized the signal and minimized the instrumental noise. Ludlum Inc. recommended a voltage range between 500 V and 1200 V for the photomultiplier tube. The figure of merit, efficiency squared divided by the background, optimized the detection efficiency and background noise. The figure of merit of the ZnS(Ag) detector was determined using an $^{241}$Am electroplated standard for voltages ranging between 700 V and 1000 V in Figure A.1. The range was selected after the oscilloscope was used to observe the entire voltage range recommended by Ludlum Inc. The operating voltage (850V) and the amplifier gain (1000) were chosen in order to decrease the background noise and to maximize alpha detection when the signal splitter was in use. The high voltage of 800V was selected as the operating voltage when the charge-sensitive preamplifier was used and an Ortec amplifier gain of 20 was set.

![Figure A.1](image)

**Figure A.1** The figure of merit for the ZnS(Ag) detector using the signal splitter.
Appendix B

Signal Splitter versus Charge-Sensitive Preamplifier

The signal splitter and charge-sensitive preamplifier were both used to split the input and output of the electronic signal. The comparisons and differences between the instruments and results were examined. Pulse height spectra taken with the signal splitter and charge-sensitive preamplifier were compared to determine the more appropriate instrument in the direct detection of alpha radioactivity from aqueous solutions.

Pulse Pile-up Background

Pulse pile-up is a phenomenon associated with high count rates when two pulses are close enough to be received as one pulse by the analysis system. Tail pile-up and peak pile-up are two types of pulse pile-up. Tail pile-up occurs when pulses are superimposed upon the long duration tail or undershoot from the preceding pulse. Tail pile-up creates artificially larger or smaller pulses as shown in Figure B.1. When the tail is negative, a smaller pulse is created, while pile-up on a positive tail results in a larger pulse. Detection systems affected by pulse pile-up records only one pulse instead of two, resulting in a smaller total area in a given spectrum. Pulses, as shown in Figure B.2, are combined into a pulse with amplitude equal to the summation of the two original pulses. Tail and peak pile-up affects the $^{55}$Fe pulse height spectral peak shape as shown in Figure B.3. The dash line represents a low-activity scenario where pile-up is absent and the dark line signifies pile-up due to high-activity [6].
Figure B.1 Tail pile-up occurs whenever a tail or undershoot from a previous pulse is present. The effect of the pile-up is displayed on the pulse height spectra to the right as the cross-hatched area [6].

Figure B.2 Two signal pulses which are close together form into a single distorted pulse as peak pile-up. The dashed lines represent possible overlapping scenarios [6].
Figure B.3 A pulse height spectra of $^{55}\text{Fe}$ demonstrates the effects of peak and tail pile-up. A low counting rate, where pile-up is negligible, is represented by the dashed line. The solid line represents a high counting rate and displays effects due to both tail and peak pile-up [6].

Detection Dead Time and Recovery Time Background

Radiation detectors often fail to properly process incident radioactivity during large count rates due to the dead time or recovery time of the detector. Electronic pulses are not formed during the dead time of the detector and do not reach their full magnitude during the recovery time of the detector. The dead time of a counting system is the minimum time which separates two events in a detector and allows the events to be recorded separately. The recovery time of the detector is defined as the time interval which the detector needs to return to its original state and to produce a pulse of the initial amplitude. The pulses of a Geiger Mueller tube demonstrated dead time and recovery time in Figure B.4 [6].
Figure B.4  The electronic signal pulse shape which is signature for dead time in a Geiger-Mueller counter during high count rates [6].

An event is lost by the detector if it occurs too rapidly in time after the preceding event as dead time follows each event in the detector. The dead time is set by either the detector or the associated electronics. Losses in a detector are significant when the detection system undergoes high counting rates. Two models of dead time behavior are the paralyzable and nonparalyzable responses [6]. If an event occurs during the dead time of a paralyzable detector, the count is not recorded and the dead time is prolonged as shown in Figure B.5. If an event occurs during the dead time of a nonparalyzable detector, the count is not recorded and the dead time is not prolonged. The top figure in Figure B.5 demonstrates a paralyzable detector where only three counts are recorded for the six events. The bottom figure represents a nonparalyzable detector which is able to record four counts as the dead time is not extended. Although a nonparalyzable detector is not able to record a count rate equal to the true event rate, the detector detects an increasing count rate when the interaction rate is increasing. The paralyzable detector is only capable of detecting a rising interaction rate to the point where the count rate levels
off and then begins to decline, Figure B.6. The dashed line represents when the recorded count rate, $m$, is equivalent to the true interaction rate, $n$, in the ideal detector [6].

Figure B.5 The recorded events in a detector, paralyzable versus nonparalyzable models [6].

Figure B.6 The true count rate versus the observed count rate for nonparalyzable and paralyzable detectors [6].

Errors remain high in the counting system even with attempts to correct for dead time. The value of $\tau$ varies and the detector does not necessarily follow the nonparalyzable or paralyzable model. Dead time losses greater than 30 or 40% have a large effect on the count rate and a different detection system is recommended [6].
**Instrumental Components**

The sole contents of the signal splitter were a resistor and capacitor. The circuit, shown in Figure B.7, was responsible for redirecting the input and output currents. The signal splitter lacked amplifying capabilities and simply passed the signal to the amplifier. The \((1.5 \pm 0.3) \times 10^{-3} \ \mu F\) capacitor, used as a filter, allowed alternating current from the detector to pass into the amplifier; however, it blocked the direct current from the high voltage supply and detector. The ability of the filter to block DC circuit was due to the capacitive reactance of the capacitor. The capacitive reactance was the measure of the opposition or resistance of the capacitor to current. The capacitive reactance naturally increased at low frequency and decreased for high frequency signals. The steady signal from DC current had zero frequency which resulted in an infinite capacitive reactance or total opposition [32].

![Figure B.7](image)

**Figure B.7** The schematic diagram of the signal splitter.

The Canberra preamplifier Model 2006E was a charge-sensitive preamplifier which shaped and amplified the output signal from the detector before it was further amplified by the amplifier. The charge-sensitive preamplifier had several components including a DC filter, integrator, P/Z differentiator, and buffer as shown in Figure B.8. The DC filter, a \(1.0 \times 10^{-3} \ \mu F\) capacitor, separated the DC high voltage and detector from the rest of the circuit and signal output. The operational integrator integrated the
incoming charge pulse. The potential difference across the feedback capacitor was proportional to the accumulated charge from the detector input. The differentiator, a pole/zero (p/z) cancellation circuit, was responsible for providing a 50 µs tail pulse and the optimum overload performance. The buffer applied a high voltage gain to the signal to allow the signal to pass through a long cable without significant pulse degradation. A gain setting favorable to high signal gain and low noise was applied to the electronic pulse signal [33].

![DC filter diagram](image)

**Figure B.8** The schematic diagram of the Canberra preamplifier Model 2006 [33].

The charge-sensitive preamplifier provided gain to the detector signal which supplied a stronger electronic pulse output than the signal splitter and reduced the gain needed from the amplifier. The amplifier was capable of amplifying the signal; however, it also amplified the instrumental noise. In order for the electronic pulse from the signal splitter to be strong enough to be used by the multi-channel analyzer, the amplifier needed to boost both the signal and the noise by a factor of 1,000, the maximum setting. In the presence of the charge-sensitive preamplifier, the instrumental noise levels remained small as the amplifier was set to 20, the minimum setting.
Experimental Results

The pulse height spectra obtained with the ZnS(Ag) detector connected to the signal splitter were different from the spectra obtained when the detector was connected to the charge-sensitive preamplifier while the detection capabilities varied at high count rates. The absolute detection efficiency of the detector in conjunction with both the signal splitter and charge-sensitive preamplifier remained consistent for low count rates; however, characteristics in the pulse height spectra differed. Figure B.9 depicted the count rates from the ZnS(Ag) detector in conjunction with the charge-sensitive preamplifier and the signal splitter. The count rates were equivalent at low count rates and began to differ when high-activity electroplated standards were detected. The aqueous solutions possessed low activities and were primarily consistent. Despite similarities in detection, the pulse height spectra taken with the ZnS(Ag) detector and signal splitter were different from those taken with the ZnS(Ag) detector in conjunction with the charge-sensitive preamplifier. The differences in pulse height spectra were present regardless of count rate. The pulse height spectra obtained with the ZnS(Ag) detector and signal splitter displayed two distinctive properties. The pulse height spectra possessed overall good peak energy resolution, however, experienced decreased energy resolution when the spectra shifted to the low-energy region of interest in the presence of high alpha or beta activity.
Figure B.9 The count rate of the ZnS(Ag) detector in conjunction with both the charge-sensitive preamplifier and the signal splitter for the detection of several electroplated standards.

The pulse height spectra for a $^{239}$Pu solution and $^{239}$Pu electroplated standard, both at a 5.2 mm air gap distance and detected with the 14 mg/cm$^2$ ZnS(Ag) detector in conjunction with the signal splitter, demonstrated similar pulse height spectra in Figure B.10 despite the differences in source medium. The pulse height spectrum for the $^{239}$Pu aqueous solution was only slightly broader than the pulse height spectra of the corresponding electroplated standard. The pulse height spectrum obtained with the signal splitter appeared to offer better energy resolution; however, the pulse height spectrum lacked characteristics commonly associated with alpha particles. The ZnS(Ag) detector and signal splitter pulse height spectrum for the electroplated standard in Figure 5.10 did not resemble the computer-simulated pulse height spectrum for a point source in Figure 5.17(a) as tailing was not present. The pulse height spectrum for the aqueous solution taken with the ZnS(Ag) detector and signal splitter, Figure B.10, was not consistent with the theoretical pulse height spectrum for the thick scintillation layer as simulated in...
Figure 5.17(b). The experimental pulse height spectrum lacked the continuous energy distribution of the aqueous solution. The pulse height spectrum did not resemble the finite scintillation layer in Figure 5.17(b) as the low-energy tail was not present. ZnS(Ag) was reported to scintillate for alpha energies as low as 0.1 MeV in experiments conducted at Saint Gobain Crystals, Inc. which suggested the expectation of the low alpha energy tail [34].

The pulse height spectra for several aqueous solutions, $^{244}$Cm, $^{239}$Pu, and $^{234}$U/$^{238}$U, taken with the 14 mg/cm$^2$ ZnS(Ag) detector and signal splitter demonstrated a narrow peak resolution in Figure B.11. The pulse height spectra obtained while using the signal splitter did not reveal the broad alpha peaks present in Figure 5.12, the pulse height spectra obtained while using the charge-sensitive preamplifier. The leading edges of the pulse height spectra obtained with the ZnS(Ag) detector and signal splitter were separated by 17 channels for the 5.8 MeV $^{244}$Cm solution and a 4.8 MeV $^{234}$U/$^{238}$U solution. The
leading edges of the pulse height spectra obtained with the ZnS(Ag) detector and charge-sensitive preamplifier in Figure 5.12 were separated by 77 channels for the 5.8 MeV $^{244}$Cm solution and 4.8 MeV $^{234}$U/$^{238}$U solution. The differences in the channel numbers of the various solutions demonstrated that the ZnS(Ag) detector and signal splitter had a small, non-linear energy dependence.

![Figure B.11](image)

**Figure B.11** The pulse height spectra for $^{244}$Cm, $^{239}$Pu, and $^{234}$U/$^{238}$U aqueous solutions using the thick scintillation layer on the ZnS(Ag) detector and the signal splitter.

The charge-sensitive preamplifier and the signal splitter were both utilized to separate the signal for the ZnS(Ag) detector from the high voltage input. The instruments connected the high voltage input and the signal output to a single electrical port on the ZnS(Ag) detector probe. The sole purpose of the signal splitter was to split the high voltage and signal pulses, while the charge-sensitive preamplifier was a more sophisticated device affecting the electric signal from the ZnS(Ag) detector. Although a charge-sensitive preamplifier was not required for gross alpha detection of low count rates, it proved necessary for accurate count rates and pulse height spectra. After
contacting several electronic experts, the properties behind the instruments which caused the variations in the energy dependence of the pulse height spectra remained unknown.

The pulse height spectra measured with the 3.5 mg/cm² ZnS(Ag) detector and signal splitter shifted the pulse height spectrum to the lower energies in the presence of high activities, an effect not present with the charge-sensitive preamplifier. The phenomenon was illustrated in Figure B.12 by the shift and broadening of the ²⁴¹Am and ²³⁸Pu pulse height spectra. The alpha energies for the ²⁴¹Am electroplated standards were 5.479 MeV and ²³⁸Pu had a similar alpha energy of 5.487 MeV. The lowest activity electroplated standard, 489.9 ²³⁸Pu, had the best resolution, 9.2%, and the peak maximum was located in the highest channel, 434. The 1644 Bq ²⁴¹Am electroplated standard had a peak resolution of 11.7% and the peak maximum channel number at 424. The highest activity electroplated standard, 7151.7 Bq ²⁴¹Am, had the poorest resolution, 28.7%, and the peak maximum was located in the lowest channel, 365. The alpha peaks were expected to be located at the same channel since the alpha energies were approximately the same. A difference of 69 channels for the peak maxima of the pulse height spectra for the lowest and highest activities indicated that the ZnS(Ag) detector in connection with the signal splitter did not produce pulse height spectra properly at high count rates. The pulse height spectra for the ²⁴¹Am and ²³⁸Pu electroplated standards detected with the charge-sensitive preamplifier did not show a shift in channels or peak broadening in Figure 5.13. The greater peak maximum of the 7151.7 Bq ²⁴¹Am pulse height spectrum in Figure 5.13 was attributed to differences in solid angle. The 7151.7 Bq ²⁴¹Am electroplated surface was 0.5 mm closer to the detector than the lower activity ²⁴¹Am and ²³⁸Pu electroplated standards. At a separation distance of 1.5 mm, the difference in solid
angle was significant. The pulse height spectra of the ZnS(Ag) detector and charge-sensitive preamplifier were present in the same channels and had the same resolution, as was expected since the alpha energies were nearly the same and the energy resolution was poor.

Figure B.12 The pulse height spectra for 1644 Bq and 7151.7 Bq $^{241}$Am electroplated standards and a 490 Bq $^{238}$Pu electroplated standard using a ZnS(Ag) detector and signal splitter.

Figure B.13 The pulse height spectra for 490 Bq $^{238}$Pu EPS, 1644 Bq $^{241}$Am EPS, and 7151.7 Bq $^{241}$Am EPS detected with a ZnS(Ag) detector and charge-sensitive preamplifier.
Recovery time in the electronic signal from the signal splitter was responsible for the pulse height spectral shift at high count rates. Recovery time was absent in the electronic pulse from the ZnS(Ag) detector, signal splitter, and amplifier in Figure B.14(a) for the low activity, 489.9 Bq, $^{238}$Pu electroplated standard. The filter capacitor inside the signal splitter had sufficient time to recover between electronic pulses. The electronic pulse from the ZnS(Ag) detector, charge-sensitive preamplifier, and amplifier, Figure B.14(b), also demonstrated a similar pulse for the 489.9 Bq $^{238}$Pu electroplated standard which was free of recovery time. The electronic pulse of the 1644 Bq $^{241}$Am electroplated standard detected by the detector, signal splitter, and amplifier exhibited recovery time in Figure B.15(a). The electronic pulses to the right of the primary pulse were smaller and amplified with time. The energy output overlapped with the previous signal energy if too little time had lapsed. The collection of energy occurred at the capacitor in the signal splitter. The count rate of the alpha particles, $615.6 \pm 1.4$ cps, was large enough for the capacitor in the signal splitter to stop functioning properly. The electronic pulses entered the capacitor at a fast rate, not allowing for the capacitor to discharge fully after each signal. Although the same electroplated standard was detected by the detector, charge-sensitive preamplifier, and amplifier in Figure B.15(b), recovery time was not present in the electronic pulses. Recovery time remained absent in the electronic pulses of the highest activity $^{241}$Am electroplated source, 7151.7 Bq, detected with the ZnS(Ag) detector, charge-sensitive preamplifier, and amplifier in Figure B.16(b). The detector, signal splitter, and amplifier electronic signal for the detection of the highest activity, 7151.7 Bq, $^{241}$Am electroplated source was observed in Figure B.16(a). The electronic signal demonstrated an even more exaggerated recovery time
pattern. The recovery time for the 7151.7 Bq $^{241}$Am electroplated source was 120 µs as compared to only 80 µs for the 1644 Bq $^{241}$Am electroplated standard. The higher count rates overwhelmed the capacitor inside the signal splitter and intensified the problem. The recovery time did not paralyze the detector as the small electronic pulses were still counted. The tail located on the 7151.7 Bq $^{241}$Am pulse height spectra in Figure B.12 represented the electronic signals which were reduced to lower energies due to the effects of recovery time. The low-energy electronic pulses caused by the effects of recovery time were responsible for the shift in the peak maxima of the $^{241}$Am pulse height spectra, Figure B.12. While the ZnS(Ag) detector was connected to the charge-sensitive preamplifier, recovery time was absent from the electronic signals which resulted in pulse height spectra which did not shift at high count rates, Figure B.13.

![Figure B.14](image)  
**Figure B.14** The electronic signal from the ZnS(Ag) detector and (a) signal splitter and (b) charge-sensitive preamplifier demonstrated the pulses created by a 489.9 Bq $^{238}$Pu electroplated standard.
Figure B.15 The electronic signal from the ZnS(Ag) detector and (a) signal splitter and (b) charge-sensitive preamplifier demonstrated the pulse created by a 1644 Bq $^{241}$Am electroplated standard.

Figure B.16 The electronic signal from the ZnS(Ag) detector and (a) signal splitter and (b) charge-sensitive preamplifier demonstrated the pulse created by a 7151 Bq $^{241}$Am source.

The absolute detection efficiency of the ZnS(Ag) detector in connection to the signal splitter and charge-sensitive preamplifier were consistent for low count rates as shown in Figure B.1 and Table B.3. The absolute detection efficiencies for the instruments fell within uncertainty for the 489.9 Bq sample—35.6 $\pm$ 0.5% and 34.8 $\pm$ 0.5% for the signal splitter and charge-sensitive preamplifier, respectively. The absolute
detection efficiencies of the ZnS(Ag) detector coupled to the signal splitter and charge-sensitive preamplifier were 37.4 ± 0.4% and 35.9 ± 0.4% for the 1644 Bq sample, respectively. The absolute detection efficiencies of the 489.9 Bq $^{238}$Pu electroplated standard and 1644 Bq $^{241}$Am electroplated standard were expected to be similar for the similar alpha energies, 5.487 MeV and 5.479 MeV, respectively. The recovery time present in the electrical signal from the signal splitter did not have an effect on the absolute detection efficiency. The absolute detection efficiencies of the charge-sensitive preamplifier for the $^{238}$Pu and $^{241}$Am electroplated standards were close and just fell out of agreement. The absolute detection efficiencies for the highest count rate detected by the ZnS(Ag) detector in connection to the signal splitter and charge-sensitive preamplifier were not consistent with the absolute detection efficiencies for the lower count rates due to differences in solid angle. The absolute detection efficiency for the 7151.7 Bq $^{241}$Am electroplated standard was high for the ZnS(Ag) detector and signal splitter, 41.3 ± 0.5%, for the 115-511 channel region of interest. The absolute detection efficiency for the ZnS(Ag) detector in connection with the charge-sensitive preamplifier was artificially large due to solid angle effects at 39.1 ± 0.5% for a region of interest between 20 and 511 channels. The regions of interest varied between the pulse height spectra of the signal splitter and charge-sensitive preamplifier due to differences in the location of the alpha region of interest and the noise peak. The noise peak of Figure 5.11 was located in the higher channels which indicated that a LLD at 115 was necessary for the signal splitter. The noise peak of Figure 5.2 dictated a LLD of 20 was necessary. The absolute detection efficiencies of the alpha electroplated standards were summarized in Table B.1.
Table B.1 The absolute detection efficiencies of the ZnS(Ag) detector connected to both the signal splitter and the charge-sensitive preamplifier.

<table>
<thead>
<tr>
<th>Electroplated Standard</th>
<th>Activity (Bq)</th>
<th>Count Rate (cps) Signal Splitter</th>
<th>Count Rate (cps) Preamplifier</th>
<th>Abs. Detection Efficiency (%) Signal Splitter</th>
<th>Abs. Detection Efficiency (%) Preamplifier</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}$Pu</td>
<td>$489.9 \pm 6.4$</td>
<td>$174.6 \pm 0.8$</td>
<td>$170.7 \pm 0.8$</td>
<td>$35.6 \pm 0.5$</td>
<td>$34.8 \pm 0.5$</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>$1644 \pm 18.1$</td>
<td>$615.6 \pm 1.4$</td>
<td>$590.4 \pm 1.4$</td>
<td>$37.4 \pm 0.4$</td>
<td>$35.9 \pm 0.4$</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>$7152 \pm 93.0$</td>
<td>$2957 \pm 3.1$</td>
<td>$2793.7 \pm 3.1$</td>
<td>$41.3 \pm 0.5$</td>
<td>$39.1 \pm 0.5$</td>
</tr>
</tbody>
</table>

In the presence of high beta activity, the alpha pulse height spectrum shifted to lower energies for the signal splitter while a shift did not occur for the charge-sensitive preamplifier. The beta particle radiation field was created with a $^{90}$Sr($^{90}$Y) source at a distance less than 10 cm from the ZnS(Ag) detector face. The 7151.7 Bq $^{241}$Am electroplated standard was placed upon the 0.635 cm Plexiglas shield and platform illustrated in Figure 4.4, above the $^{90}$Sr($^{90}$Y) source. The peak maximum of a pulse height spectra for the 7151.7 Bq $^{241}$Am electroplated standard in the absence of the beta radioactivity was located at channel 401 in Figure B.17, when the ZnS(Ag) detector was connected to the signal splitter. The pulse height spectrum shifted to lower energies in the presence of the high beta radioactivity which resulted in a peak maximum at channel 101 while the pulse height spectra of the ZnS(Ag) detector and charge-sensitive preamplifier did not shift in Figure B.18. Although the instrument set up in Figure 4.4 was utilized, the solid angle between the electroplated standard and the ZnS(Ag) detector connected to the signal splitter did not remain consistent with and without beta radioactivity due to an increased distance between the detector face and the electroplated standard. The pulse height spectra of the ZnS(Ag) detector in conjunction with the signal splitter did not shift despite solid angle or changes in alpha energy in the absence of beta and gamma radioactivity which permitted the spectra to be compared.
Figure B.17  The pulse height spectra of the 7151.7 Bq $^{241}$Am EPS on top of the Plexiglas shield with and without the 7.4 x $10^8$ Bq $^{90}$Sr($^{90}$Y) source detected with the ZnS(Ag) detector and signal splitter.

Figure B.18  The pulse height spectra of the 7151.7 Bq $^{241}$Am EPS on top of the Plexiglas shield with and without the 7.4 x $10^8$ Bq $^{90}$Sr($^{90}$Y) source detected with the ZnS(Ag) detector and charge-sensitive preamplifier.

Combined alpha and beta radiation demonstrated greater increased noise in the electronic signals in the ZnS(Ag) detector and signal splitter than the charge-sensitive preamplifier. The oscilloscope trace of the ZnS(Ag) detector in connection with the signal splitter and amplifier, Figure B.19(a), and the ZnS(Ag) detector in connection with the charge-sensitive preamplifier and amplifier, Figure B.19(b) revealed differences in
the noise baseline. The baseline of the signal splitter oscilloscope trace, Figure B.19(a) demonstrated an enlarged baseline greater than one volt due to noise and lower energy deposition in the ZnS(Ag) detector from beta interactions. The electronic signal from the noise and beta particles overwhelmed the capacitor inside of the signal splitter which caused recovery time problems. The electronic signal from the detected alpha particles did not properly charge the capacitor. The electronic pulses reduced the amplitude of the pulses during combined alpha and beta detection. The decreased pulse amplitude and therefore energy explained the shift of the pulse height spectrum of Figure B.17. The beta signal interference was absent in the electronic signal of combined alpha and beta radioactivity, Figure B.19(b), for the ZnS(Ag) detector and charge-sensitive preamplifier detection system. The charge-sensitive preamplifier was able to maintain a flat baseline in the presence of beta radioactivity. The absence of recovery time in the electronic signal of the ZnS(Ag) detector in connection with the charge-sensitive preamplifier explained the consistency of the location of the pulse height spectra with and without beta interference in Figure B.18.

Figure B.19  The electronic signal for the ZnS(Ag) detector, amplifier, and (a) signal splitter and (b) the preamplifier. The 7151.7 Bq \(^{241}\text{Am}\) electroplated standard was set on top of the 0.635 cm Plexiglas sheet placed above a 7.4 x 10^8 Bq \(^{90}\text{Sr}\left(\text{Y}\right)\) source.
Appendix C
Visual Basic Computer Model

A computer model was designed in Microsoft Excel using Visual Basic to predict the pulse height spectra for the detection of alpha particles from an infinitely-thick source with a finite thickness detector. The infinitely thick radioactive source, the aqueous solution in a sample holding cup, was simulated by randomly sampling a point \((x,y,z)\) inside of the theoretical box, Figure C.1. The cylindrical sample holding cup and circular detection surface from the experimental set up were simplified to a square cup and square detection surface with a surface area equivalent to that of the experimental apparatus. The coordinate system was placed at one of the corners of the square at a distance, \(z_1\), from the top of the liquid surface. The distance, \(z_1\), was selected as the active depth of the liquid which determined the volume of liquid exposed to the detector. The active depth, \(z_1\), was calculated using equations 1, 2, and 3 and was an important parameter in the computer simulation. If the distance was too large, complete absorption for the majority of the simulated points resulted in poor counting statistics for the pulse height spectrum.
All random numbers were generated using the Microsoft Excel random number generator. The simulation consisted of 19,992 generated alpha particles. The randomly generated origin of each alpha particle, \((x_o, y_o, z_o)\), represented a spherical moving coordinate as shown in Figure C.1. The x and y components, the origin of the alpha particle, were randomly generated between 0 and 2.04 cm. The z component was
generated between 0 and the active depth, the maximum range the alpha particle traveled in solution.

\[
0 < x_0 < 2.04 \text{ cm} \\
0 < y_0 < 2.04 \text{ cm} \\
0 < z_0 < z_1 (\text{cm})
\]  \hspace{1cm} (14)

The direction of the alpha particle was randomly generated in all directions. Spherical coordinates, \( \theta \) and \( \phi \), were generated as

\[
0 < \theta < 2\pi \\
0 < \phi < \pi
\]  \hspace{1cm} (15)

The spherical coordinate of \( r \) represented the distance the alpha particle traveled and was a function of the vector \( z \) and \( \phi \). The standard spherical coordinate equations

\[
x = r \sin \phi \cos \theta \\
y = r \sin \phi \sin \theta \\
z = r \cos \phi
\]  \hspace{1cm} (16)

were utilized where \( x, y, z, \) and \( r \) were variables [35]. The vertical distance, \( z_1 - z_0 \), and the known \( \phi \) were used to determine the total distance the alpha particle travels through the water absorption layer, \( r_1 \). The variables, \( r_2, r_3, \) and \( r_4 \), were calculated for each absorption layer. The distance of the vector, \( r_1 \), between the surface of the liquid, \( z_1 \), and the original point, \( z_0 \),

\[
r_1 = \frac{z_1 - z_0}{\cos \phi}
\]  \hspace{1cm} (17)

was found. The distance to the surface of the liquid from the origin of axes at the corner of the box in Figure C.1 was always equal to the active depth, \( z_1 \). The \( z \) components of the box, the thickness of the air, Mylar\textsuperscript{®}, and ZnS(Ag) layers were known. The calculated \( r \) and randomly generated \( \phi \) were then used to determine the \( x \) and \( y \) at the
Mylar® surface using equation 16. For example, the distance, $r_2$, from the original point, $z_o$, to the surface of Mylar®, $z_2$, was determined when $\varphi$ and $\theta$ were known as shown in equation 17. The coordinates, $x_2$ and $y_2$, were the intersection points at the air-Mylar® surface while $x_o$ and $y_o$ were the coordinates for the original randomly generated point, equation 18. The edges of the sample holding cup collimated the alpha radiation onto the surface detection area.

$$
\begin{align*}
    x_2 &= r_2 \sin \varphi \cos \theta + x_o \\
    y_2 &= r_2 \sin \varphi \sin \theta + y_o
\end{align*}
$$

The directional alpha particle vectors were generated in $4\pi$ and a method to eliminate the alpha particles which were never incident on the detection surface was created. A series of “If Statements” were designed in Microsoft Excel using binary systems. A zero was placed in the column if the statement was false and a one was input into the spreadsheet if the statement was true. The “If Statements” defined the parameters of the detection geometry. If a false, or zero, was present for a generated point, the point was cancelled and was determined to never be incident with the detection surface. A false was output if the randomly generated $\varphi$ was above $\pi/2$ meaning that the alpha particle was generated into the negative direction. The distance the alpha particle traveled in water was compared to the maximum range of alpha particle in solution. If the alpha particle traveled a distance that was greater than the maximum range, a false was recorded. If the computed values of $x_2 + x_o$ or $y_2 + y_o$ were greater than 2.04 cm, then the alpha particle left the box and a false was recorded. If $x_2 + x_o$ or $y_2 + y_o$ were negative then the alpha particle left the box and never reached the theoretical detection surface. The original coordinates, $x$ and $y$, were added to the intersection values to take into account the moving coordinate system.
The simplified Hans Bethe equation,

\[
\frac{dE}{dx} = C_1 n \left[ \ln \left( \frac{C_2 \left( \frac{v}{c} \right)^2}{I \left( 1 - \left( \frac{v}{c} \right)^2 \right)} \right) \right] \quad \text{MeV cm}^{-1}
\]  

(19)

was used to simulate the energy lost by the alpha particles above 0.15 MeV in the various materials. The extrapolated equation from experimental data conducted by Ziegler, equation 20, was used to calculate the stopping power for alpha particle energies between 0.001 MeV and 0.15 MeV where E was in MeV [31]. Alpha energies less then 0.001 MeV were assumed to be 0 MeV.

\[
\frac{dE}{dx} = -29096E^2 + 11329E + 320.86
\]  

(20)

The constants in Table C.1 were called upon in the Microsoft Excel worksheet, Figure C.2, by the Visual Basic source code in the assessment of equation 19. C1 and C2 were constants present in equation 8 while n and I were found using equations 6 and 7, respectively. The value for the speed of light in vacuum, 3 x 10^8 m/s, was a known constant.

<table>
<thead>
<tr>
<th>Constants</th>
<th>HNO₃ Solution</th>
<th>Air</th>
<th>Mylar®</th>
<th>ZnS(Ag)</th>
<th>Al</th>
<th>Si</th>
</tr>
</thead>
<tbody>
<tr>
<td>C₁</td>
<td>2.03x10⁻³⁰</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C₂</td>
<td>1.02x10⁶</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>n</td>
<td>3.00x10²⁹</td>
<td>3.92x10²⁶</td>
<td>4.08x10²⁹</td>
<td>1.15x10³⁰</td>
<td>7.84x10²⁹</td>
<td>6.99x10²⁹</td>
</tr>
<tr>
<td>I</td>
<td>92.6</td>
<td>98.1</td>
<td>69.2</td>
<td>258.0</td>
<td>163.3</td>
<td>174.7</td>
</tr>
<tr>
<td>c</td>
<td>3 x 10⁸</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Figure C.2  The Microsoft Excel worksheet referred to by the Visual Basic source codes.

The velocity of the alpha particle was not a constant and was changing through the simulation. The original velocity of the alpha particle was dependent upon the alpha particle energy of the radionuclide. The particle energy was converted to joules from MeV. The original velocity of the alpha particle was found using the non-relativistic kinetic energy equation as the alpha mass was too large to be relativistic. The original velocity was used as a starting point for equation 19. The alpha particle continuously lost energy as it traveled through the material which resulted in a constantly decreasing velocity. A loop was formed to recalculate the velocity of the alpha particle 100 times in each absorption thickness as demonstrated in Source Code C.1. The loop was only employed if the length of the distance traveled, r, was above zero. The length of the alpha particle was zero if the point was cancelled with “If Statements,” and the alpha particle was determined not to reach the detection surface.
The code ran for each of the four absorber thicknesses: liquid, air, Mylar®, and ZnS(Ag). Source Code C.1 modeled the loss of alpha particle energy in the solution, and output the energy which the alpha particle had as it left the solution. The alpha particle energy was then input into Source Code C.2 as the initial energy of the alpha particle as it began to transport through air. Source Code C.2 simulated the loss of alpha energy as it traveled through air. Source Code C.3 and Source Code C.4 modeled the Mylar® layer and ZnS(Ag) layer, respectively. Source Code C.4 output the final energy of the alpha particle as it exited the ZnS(Ag) layer into the Microsoft Excel worksheet. The change of energy, energy deposited in the scintillation layer, was found by subtracting the final energy of the particle after having exited the ZnS(Ag) layer from the energy of the alpha particle after it left the Mylar® layer.

The histogram function in Microsoft Excel was used to determine the number of counts in each energy bin. The efficiency or “incident counts over total counts” was found by dividing the counts in each bin by the total number of counts. Using the computer model, the pulse height spectra or counts per energy bin versus energy bin, were simulated.
Source Code C.1. The computer code for the simulation of the energy of the alpha particle as it left the aqueous solution.

Sub Loopysolutionwater()
    For tmp = 15 To 20006
        L = Cells(tmp, 1)
        EmeV = Cells(5, 1)
        EJ = Cells(7, 1)
        Cells(7, 3) = EJ
        dL = L / 100
        M = Cells(8, 1)
        c = Cells(6, 4)
        Cells(7, 4) = c
        n = Cells(1, 3)
        I = Cells(1, 6)
        C1 = Cells(9, 1)
        C2 = Cells(10, 1)

        If L = 0 Then
            Cells(tmp, 5) = EmeV
        End If

        If L > 0 Then
            For Intervals = 1 To 100
                If EmeV > 0.15 Then
                    v = Sqr(Abs(2 * EJ / M))
                    B = v / c  'unit-less
                    term1 = ((C1 * n) / (B ^ 2))
                    term2 = (C2 * B ^ 2) / (I * (1 - B ^ 2))
                    term3 = Log(term2) - B ^ 2
                    dEdx = term1 * term3
                    dE = dEdx * dL
                    EmeV = EmeV - dE
                    EJ = EmeV * 1000000 * 1.60217646E-19
                ElseIf (0.001 < EmeV) And (EmeV <= 0.15) Then
                    dEdx = -29096 * EmeV ^ 2 + 11329 * EmeV + 320.86
                    dE = dEdx * dL
                    EmeV = EmeV - dE
                ElseIf (EmeV <= 0.001) Then
                    EmeV = 0
                End If
            Next Intervals
            Cells(tmp, 5) = EmeV
        End If
    Next tmp
End Sub
**Source Code C.2.** The computer code for the simulation of the energy of the alpha particle as it left the air absorber layer.

```vba
Sub Loopsolutionair()
    For tmp = 15 To 20006
        L = Cells(tmp, 2)
        EmeV = Cells(tmp, 5)
        EJ = EmeV * 1000000 * 1.60217646E-19
        dL = L / 100
        M = Cells(8, 1)
        c = Cells(6, 4)
        n = Cells(2, 3)
        I = Cells(2, 6)
        C1 = Cells(9, 1)
        C2 = Cells(10, 1)

        If L = 0 Then
            Cells(tmp, 6) = EmeV
        End If

        If L > 0 Then
            For Intervals = 1 To 100
                If EmeV > 0.15 Then
                    v = Sqr(Abs(2 * EJ / M))
                    B = v / c   'unit-less
                    term1 = ((C1 * n) / (B ^ 2))
                    term2 = (C2 * B ^ 2) / (I * (1 - B ^ 2))
                    term3 = Log(term2) - B ^ 2
                    dEdx = term1 * term3
                    dE = dEdx * dL
                    EmeV = EmeV - dE
                    EJ = EmeV * 1000000 * 1.60217646E-19
                    ElseIf (0.001 < EmeV) And (EmeV <= 0.15) Then
                        dEdx = -29096 * EmeV ^ 2 + 11329 * EmeV + 320.86
                        dE = dEdx * dL
                        EmeV = EmeV - dE
                        ElseIf (EmeV <= 0.001) Then
                            EmeV = 0
                    End If
                Next Intervals
            Cells(tmp, 6) = EmeV
        End If
    Next tmp
End Sub
```
Source Code C.3. The computer code for the simulation of the energy of the alpha particle as it left the Mylar® absorber layer.

Sub LoopysolutionMylar()
    For tmp = 15 To 20006
        L = Cells(tmp, 3)
        EmeV = Cells(tmp, 6)
        EJ = EmeV * 1000000 * 1.60217646E-19
        dL = L / 100
        M = Cells(8, 1)
        c = Cells(6, 4)
        n = Cells(3, 3)
        I = Cells(3, 6)
        C1 = Cells(9, 1)
        C2 = Cells(10, 1)

        If L = 0 Then
            Cells(tmp, 7) = EmeV
        End If

        If L > 0 Then
            For Intervals = 1 To 100
                If EmeV > 0.15 Then
                    v = Sqr(Abs(2 * EJ / M))
                    B = v / c "unit-less"
                    term1 = ((C1 * n) / (B ^ 2))
                    term2 = (C2 * B ^ 2) / (I * (1 - B ^ 2))
                    term3 = Log(term2) - B ^ 2
                    dEdx = term1 * term3
                    dE = dEdx * dL
                    EmeV = EmeV - dE
                    EJ = EmeV * 1000000 * 1.60217646E-19
                ElseIf (0.001 < EmeV) And (EmeV <= 0.15) Then
                    dEdx = -29096 * EmeV ^ 2 + 11329 * EmeV + 320.86
                    dE = dEdx * dL
                    EmeV = EmeV - dE
                ElseIf (EmeV <= 0.001) Then
                    EmeV = 0
                End If
            Next Intervals
            Cells(tmp, 7) = EmeV
        End If
    Next tmp
End Sub
Source Code C.4. The computer code for the simulation of the energy of the alpha particle as it left the ZnS(Ag) scintillation layer.

Sub LoopysolutionZnS()
    For tmp = 15 To 20006
        L = Cells(tmp, 4)
        EmeV = Cells(tmp, 7)
        EJ = EmeV * 1000000 * 1.60217646E-19
        dL = L / 100
        M = Cells(8, 1)
        c = Cells(6, 4)
        n = Cells(4, 3)
        I = Cells(4, 6)
        C1 = Cells(9, 1)
        C2 = Cells(10, 1)
        
        If L = 0 Then
            Cells(tmp, 8) = EmeV
        End If
        
        If L > 0 Then
            For Intervals = 1 To 100
                If EmeV > 0.15 Then
                    v = Sqr(Abs(2 * EJ / M))
                    B = v / c  'unit-less
                    term1 = ((C1 * n) / (B ^ 2))
                    term2 = (C2 * B ^ 2) / (I * (1 - B ^ 2))
                    term3 = Log(term2) - B ^ 2
                    dEdx = term1 * term3
                    dE = dEdx * dL
                    EmeV = EmeV - dE
                    EJ = EmeV * 1000000 * 1.60217646E-19
                ElseIf (0.001 < EmeV) And (EmeV <= 0.15) Then
                    dEdx = -29096 * EmeV ^ 2 + 11329 * EmeV + 320.86
                    dE = dEdx * dL
                    EmeV = EmeV - dE
                ElseIf (EmeV <= 0.001) Then
                    EmeV = 0
                End If
            Next Intervals
            Cells(tmp, 8) = EmeV
        End If
    Next tmp
End Sub
REFERENCES

[1] Correspondence with David Hobbs, Savannah River National Laboratory.


[34] Correspondence with Saint Gobain Crystal, Inc.