



5-2018

REAL TIME STAND-OFF GAMMA-RAY IMAGING FOR PROCESS VERIFICATION OF SPECIAL NUCLEAR MATERIAL

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To the Graduate Council:

I am submitting herewith a thesis written by Mark Streett Quint entitled "REAL TIME STAND-OFF GAMMA-RAY IMAGING FOR PROCESS VERIFICATION OF SPECIAL NUCLEAR MATERIAL." I have examined the final electronic copy of this thesis for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Master of Science, with a major in Nuclear Engineering.

John D. Auxier II, Major Professor

We have read this thesis and recommend its acceptance:

Howard L. Hall, Jared A. Johnson

Accepted for the Council:

Dixie L. Thompson

Vice Provost and Dean of the Graduate School

(Original signatures are on file with official student records.)

REAL TIME STAND-OFF GAMMA-RAY IMAGING FOR PROCESS VERIFICATION OF SPECIAL NUCLEAR MATERIAL

A Thesis Presented for the
Master of Science
Degree
The University of Tennessee, Knoxville

Mark Streett Quint
May 2018

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ACKNOWLEDGEMENTS

Serving in Knoxville and the surrounding communities between June 2016 and May 2018 has been a privilege and an honor. I would like to thank my advisor Dr. John D. Auxier II for his guidance and mentorship throughout the process of earning the M.S. degree. Furthermore, Dr. Howard Hall and Dr. Jared Johnson have also provided insight into the realm of nuclear security and generously gave of their time to aid a student in need. Mr. Jon Garrison of the Radiochemical Engineering and Development Center (REDC) was perhaps the most essential member of my experimentation team throughout the course of this research. His valuable knowledge of chemical separations and first-hand expertise in conducting operations at the REDC was the lynchpin in reaping experimental data in the laboratory. I would also like to thank Dr. Ethan Hull and Dr. Matthew Kiser for their expertise on their unique gamma-ray imaging system that enabled us to make the measurements that made these experiments possible. Thank you as well to my parents and in-laws, the Quint Family and the Spaulding Family for their unrelenting support on base, on campus, and at home. Finally, and certainly most importantly, I would like to thank my loving wife Jennifer, and daughter Charlotte for their support and undying love. Without them, I would never be whole.

ABSTRACT

A novel method for gamma-ray imaging of isotope separations is tested and validated. Current methods of validating separation processes involve taking samples or gamma-ray counting before and after a process has occurred. The method presented offers individuals or agencies the ability to verify that a process has occurred by watching the process instead of the precursors and products of the process.

The goal of this method is to establish a means to evaluate the adherence to an approved isotope separation process quickly and safely with minimal interpretation needed by the user.

PREFACE

This material was funded in part by the U.S. Dept. of Energy National Nuclear Security Administration through the Nuclear Science and Security Consortium under Award Number DE-NA0003180.

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CHAPTER ONE

INTRODUCTION AND GENERAL INFORMATION

1.1 The Need for Stand-Off Detection of Radiological Materials

Accounting for radiological material has been a challenge for governments and agencies since the discovery of these useful isotopes. Useful isotopes in the pre-war era would have included industrially useful compounds including radium, however the greater multitude of isotopes available following the introduction of nuclear reactors have increased the complexity of the accountability task. While there are legitimate uses for these materials, there are countless nefarious uses for the same material, which can have devastating effects when used in even small quantities. While gram quantities are not necessarily themselves a direct threat to national security, the damaging social consequences of even a small mass of material cannot be overlooked.

To counter this threat, countless detection systems have become commonplace in facilities that handle radioactive material. Detection systems in these facilities range from simple hand-held devices to whole body counting systems. Furthermore, not only are the detection systems broad in scope, but their application is just as diverse. From hospitals to national laboratories across the world stand-off detection is used to ensure more unauthorized movement of radiological material.

The purpose of these systems is two-fold. Not only do they provide safeguards for public health and safety but perhaps more importantly, they help prevent unauthorized materials from leaving the facility.

1.2 The Need for Monitoring the Diversion Pathways of Special Nuclear Material

Diversion of Special Nuclear Material (SNM) can be accomplished in number of ways. SNM is available for diversion before and after it enters a reactor, however if a reactor is available a greater number of diversion pathways open to a malicious actor.

Diversion of Enriched Uranium

The effort required to enrich Uranium limits those who are able to accomplish this task. However, once the process of enriching the material to a low level is perfected, the difficulty of continuing enrichment to higher levels becomes much easier.

Piping in these enrichment facilities can be rearranged or changed in order to make a peaceful facility into a clandestine operation. Various technological advances in facility mapping have helped ensure no unauthorized changes have been made to the original operational intent of the enrichment process. Inspection groups can use that same facility mapping process to verify the existence of diversion pathways that have been created within the architecture of the building.

Diversion of Transuranic Elements and Post-Irradiation Uranium

Generally, if a country has established a means of creating used nuclear fuel, their scientific prowess is sufficient to enable them to reprocess this fuel. Such a situation would allow them to extract the SNM within. Given the potential dangers that transuranic elements and the remaining uranium-235 (^{235}U) pose to the non-proliferation of nuclear weapons, any covert operation involving the use of these isotopes is unacceptable to the international community.

Traditional methods of verifying the separation of industrially or commercially useful isotopes from the isotopes that present a proliferation concern would require either alpha particle or gamma-ray analysis. The challenge surrounding these analytic methods is that they require a stop of operations while the samples are analyzed prior to continuing the separation. Furthermore, these methods are not necessarily field deployable without significant resources.

Given the verification requirements and challenges presented by the current verification methods, a verification method which allows for an in-situ validation of separations would be an invaluable tool for the safeguards community. Not only would such a tool allow for greater deployability of verification teams, but also allow for more such teams to be fielded since a smaller logistical footprint is necessary for each unit.

CHAPTER TWO

LITERATURE REVIEW

The ability to use the principle of Compton scattering has been theorized and applied within the past half-century. In the past decade this Compton technology has been transformed from bulky, Dewar cooled high purity germanium crystals, to mechanically cooled, briefcase sized units capable of both detecting and subsequently localizing samples of gamma-ray emitting radioisotopes.

2.1 Using standoff gamma-ray detection to localize radioisotopes

While gamma-ray spectroscopy alone can be burdened by background radiation, gamma-ray imaging technology is potentially less sensitive to the same background noise [1].

Facility Verification and Isotope Localization

The use of facility mapping in recent years has been well documented and offers a great deal of progress in ensuring that facilities are used as intended. Some of the earlier work in contemporary facility verification dealt with simply ensuring that an “as approved” design matched the “as built” facility. Applications of this methodology were used on interior rooms, exterior facades, and aerial compound mapping [2]. Older methods might have involved comparing photographs of the facility over time, however modern methods allow for the use of laser range scanners with millimeters of resolution. These mapping systems can be tripod, motor vehicle, or aircraft mounted and offer excellent resolution using the laser range finding technology.

More modern methods of facility verification combine the use of laser mapping technology with radiation imaging. The integration of a laser scanned image with a gamma-ray image projection allows for much more detailed analysis of facility construction [3]. Such a methodology enhances the ability of safeguard teams to ensure that facility modifications are not clandestine in nature.

More recent work building from the previous example enhances the static imagery with a moving photographic imaging and gamma-ray detection system. This allows for a greater volumetric survey of the space and can produce near real-time imaging of the isotopes in the environment [4]. Use of “Commercial Off the Shelf” (COTS) technology has also made combining these technologies

when the appropriate integration software is applied. The use of commercially available High Purity Germanium (HPGe) detectors integrated with the Microsoft® X-Box® Kinect™ sensor is one such example of this COTS application.

Process Verification and Isotope Path Confirmation

Given the macroscopic application of the integrated photographic or laser image with the gamma-ray image described in the facility verification section above, a follow-on step would practical application of that technology would be the ability to verify processes with isotopic precision. Furthermore, the capacity for integrated software and hardware to produce more real-time readouts will make process verification practical for safeguards personnel. Such process verification would disable another proliferation pathway and potentially refuse an adversary the ability to divert Special Nuclear Material (SNM).

Aqueous reprocessing of used nuclear fuel is a standard method by which commercially and industrially useful isotopes are extracted for use by private enterprise or for use as new fuel for reactors. Because of the proliferation concern surrounding the disposition of specific isotopes, many safeguards have been enacted by the International Atomic Energy Agency (IAEA). At the Rokkashomura Reprocessing Plant (RRP) many such safeguards were designed into the facility design, however shortcomings still exist. The United States Department of Energy (USDOE) called for “innovative approaches in process monitoring and other safeguards” and suggested that using the facilities own instruments to monitor processes would violate the IAEAs policy on joint-use equipment [5].

To maintain accountancy of nuclear material within on significant quantity of plutonium (8kg), and to avoid the “joint-use” of equipment, the IAEA has been using destructive assay techniques at significant financial cost and time expenditure. To address these concerns, the use of online gamma-ray spectroscopy has been simulated to provide a near real-time accountancy of process streams. However, these streams were limited to reporting normal and off-normal conditions [6]. These same simulations were later used as the basis for several process experiments and while they did demonstrate the ability to distinguish normal and off-normal spectra, they still did not integrate any spatial isotope visualization into their experiments [7].

The most recent demonstration of an online process verification technique again involved gamma-ray measurements however compared results from before and after a process had occurred. Such a scenario would allow a user to verify input and output similarity and compare gross gamma-ray counts and peak ratios for isotopic analysis. Moreover, simple counting procedures like this one are cost effective and do not require significant logistical support for continuous operation

[8]. The drawbacks of such a system would include a lack of standoff from the actual facility architecture and no ability to watch the actual process itself.

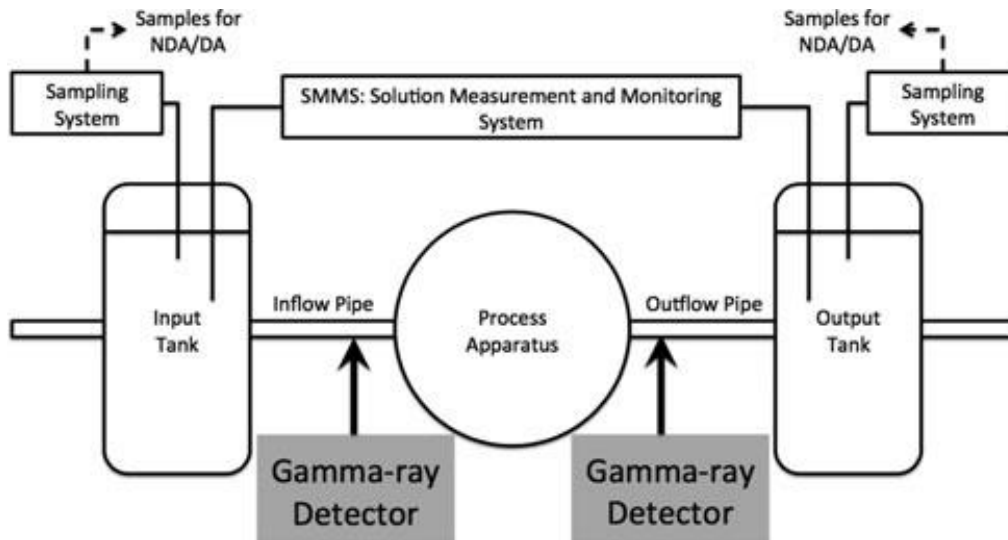


Figure 2.1: Gamma-Ray Detection Before and After Separation Process [8]

Based on the literature research, there is an apparent gap in the current state of process monitoring for safeguards purposes. Currently, methodology is only to evaluate a process before and after the actual process occurs. This can be accomplished either with destructive analysis in the post-process or online non-destructive analysis. However, what could offer greater certainty in safeguards would be a visualization of the actual process itself. Furthermore, if this process could be accomplished with an increased degree of standoff, less interaction with the facility architecture would be required by inspection groups.

CHAPTER THREE

MATERIALS AND METHODS

3.1 Equipment

The isotopes that we used in our experiments were both naturally occurring and synthetic. In general, the synthetic isotopes were substantially more active than their natural counterparts. The synthetic isotopes that were used originated at the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory (ORNL). The experiments were then conducted at the Radiochemical Engineering and Development Center (REDC) with the products of our separations used in applications across the United States in both science, medicine, and space exploration. While some of the experiments we observed are a weekly occurrence at the REDC, others were decades in the making. The einsteinium experiments we conducted were a decade in the making, and the separation was the first conducted using such a capable detection system as the one listed below.

While our experiments involved dozens of isotopes, we were able to execute each of the trials without changing much of our equipment. The most crucial device in each of our experiments was the gamma-ray detection device. Ensuring this device remained constant throughout the trials reduced uncertainty as we varied the parameters of the environment it was detecting.

Gamma-Ray Detector

For each of our four experiments, we used a high purity germanium (HPGe) detector produced by PHDS Co., of Knoxville, Tennessee. The Germanium Gamma-ray Imager (GeGI) was the fourth iteration (GeGI v4) of their marketable series of mechanically cooled HPGe detectors. The GeGI has the unique ability to both detect the presence of and identify the location of radiological species in a four-pi environment. These localized isotopes are then overlaid on a photograph taken directly in front of the detector face. All radiological signatures not forward of the detector face are visible on a white background rather than a rearward facing photograph.

The use of an individually portable detection system with such broad abilities would make such a detector invaluable to material verification teams operating in potentially restrictive environments. Normally, HPGe (high purity Ge) detection systems require the use of a Dewar of liquid nitrogen in order to cool the

germanium crystal below 80 degrees Kelvin. The advantage of the GeGI, and other systems using the same mechanical cooling technology, is that no bulky Dewar is necessary to ensure the sustained operation of the detector crystal. Contrastingly, while other gamma-ray detection systems, like Sodium Iodide (NaI), do not require cooling, their energy spectrum resolution cannot easily compete with that of HPGe systems.

During our experiments, we used two systems that reduced the portability of the GeGI, however increased the accuracy of our measurements. First, we used a tripod that allowed for the GeGI to remain stable for the duration of our samples. Secondly, we used lead collimators, with pinhole apertures between one and five millimeters, on the detector face of the GeGI (Figure 3.1).



Figure 3.1: The GeGI v4 with Gray and White Lead Collimator and Black Tripod

Glove Box Containment Systems

As our experiments involved isotopes that emitted significant and varied types of radiation all of our experiments were conducted in a glove box environment. The glove boxes contained all of the undesirable alpha and beta radiation while still allowing the experimentally useful gamma-ray radiation to penetrate the glove box plexiglass and subsequently, the detection system.

While the glove boxes did contain the undesirable alpha and beta radiation, there were some challenges involved in using these containment systems. Primarily, there have been dozens of other experiments conducted in the glove boxes that we used for our experiment. This reality has allowed unwanted background to collect in the boxes. While we attempted to minimize this with good

housekeeping procedures, our experiments will have elevated background radiation from both glove box contamination and our proximity to other active experiments in the surrounding glove boxes. The second challenge that glove boxes presented was the need to position the detector in a manner that enabled us to collect a photograph and minimally shielded gamma-ray emissions. This unobstructed placement requirement was also hindered by the need to ensure that the laboratory technician could safely and effectively perform the operation that we were witnessing. Figure 3.2 below shows the need to balance the ability of the detector to see the operation and the ability of the technician to access the gloves in the left most glove box.

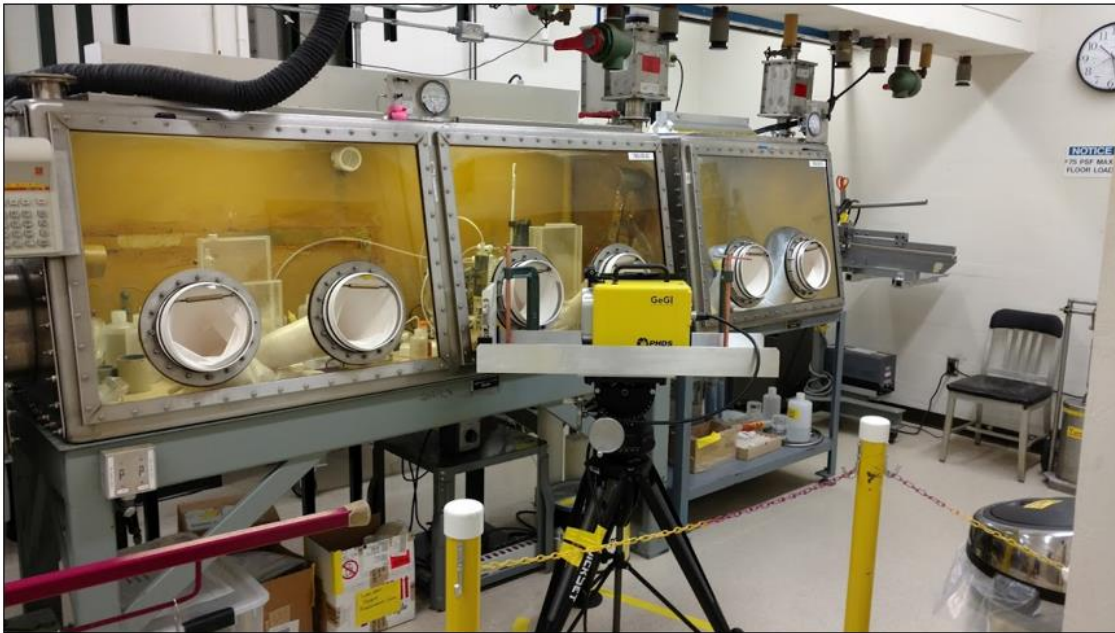


Figure 3.2: Typical Glove Box Setup for Chemical Separation

Columns for Species Separation

While we conducted three separations, only two different column resins were needed. Both the actinium – radium separation and the americium – neptunium – plutonium separation used BioRad® MP-1 100-200 mesh. Contrastingly, the einsteinium – berkelium separation used Dowex® 50W-X8 resin bed.

The physical columns for the experiments also varied based on the isotopes in question. The actinium – radium separation as well as the einsteinium – berkelium separation used a narrow diameter glass column. However, the volume of the material involved in the americium – neptunium – plutonium separation required the use of a larger diameter glass column. The differences and similarities regarding the columns may be seen in figure 3.3.



Figure 3.3: Setup of three unique experiments

3.2 Methodology for Species Separation

Column Control Operator

Each of our three experiments was executed by a trained laboratory technician using a set process algorithm. This individual controlled the pace of the experiment in order to meet the stated production timeline and goals. While the detector operator could make requests to move items around the glove box or wait to move to the next part of the chemical separation, the overall process was always under the control of the column operator who was conducting the separation for a specific customer. This level of rigidity was based on which isotopes were being separated, however because of the time restrictions placed on the medical isotope separation (actinium – radium), that experiment had little room for special requests.

Detector Operator

The detector operator was tasked with ensuring that the GeGI was collected data as designed. This involved saving the data collected at specified intervals to ensure a time progression could be shown following the completion of the experiment. The detector operator also was required to note the distance from the detector face to the column in order to compensate for the camera/detector parallax.

Radiation Control Technician (RCT)

During the course of our experiments, the team received dose from both neutron and gamma-ray radiation. Prior to each experiment, approval was required to execute these separations from the RCT overseers. To mitigate the radiation dose risks, thermo-luminescent dosimeters (TLDs) and electronic pocket dosimeters (EPDs) were worn throughout the operation. Radioactive waste was also created and was disposed of through appropriate channels when cleared by the appropriate RCT.

CHAPTER FOUR

EXPERIMENTS FOR THE VERIFICATION OF NUCLEAR MATERIAL PROCESSING METHODS

Three of our experiments began with a similar starting point in which a mixture of radioactive elements was separated into various fractions. These experiments had the with the goal of isolating an isotope of interest from the remaining fractions. While these radiochemical separations were of great interest to our research, the purpose of the separation itself was to isolate one or more isotopes for sale to a customer.

4.1 Einsteinium – Berkelium Separation Column

In order to test our hypothesis that our gamma-ray detection and visualization system, the GeGI, was able to witness, in real time, the separation of differing isotopes, we executed our first experiment using isotopes of significant activity. A mixture of einsteinium, berkelium, and californium was used to achieve the significant level of activity. The isotopic mixture was loaded on to the resin and the GeGI was allowed to collect gamma-ray signatures over the course of roughly 2.5 hours (9124 seconds). During this collection, the detector was able to identify the location of the isotopic mixture within the vial (figure 4.1), noting that the berkelium appeared well distributed within the liquid in the vial.

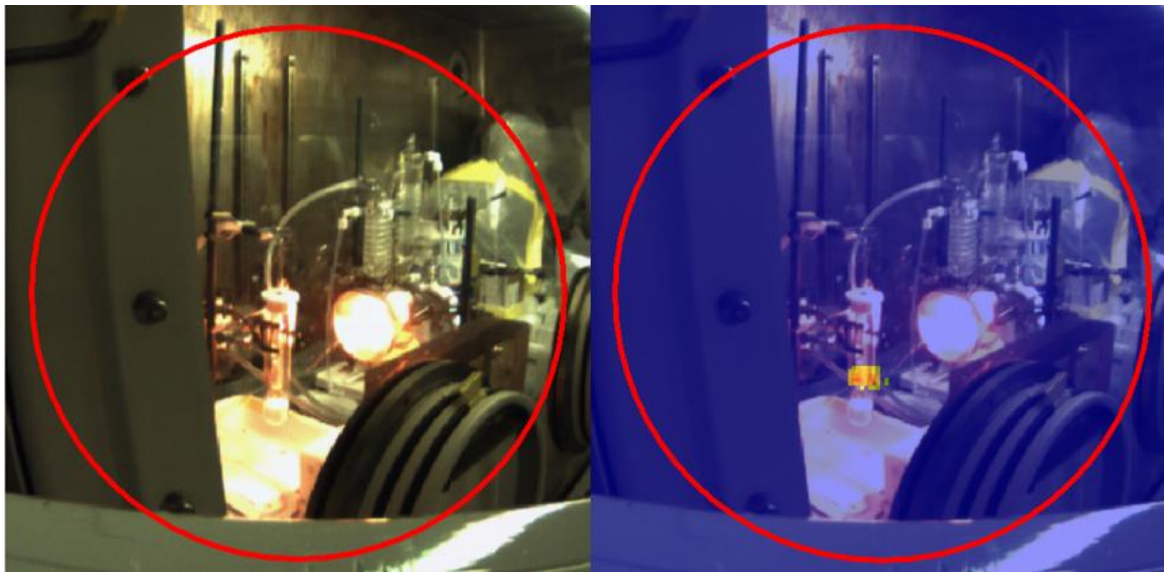


Figure 4.1: Initial Isotopic Mixture (^{253}Es – ^{249}Bk)

Following this initial collection, our team attempted a separation of the isotopic mixture with the goal of collecting a purified sample of ^{253}Es . While purifying the we were able to note the physical movement of the ^{249}Bk gamma-ray lines throughout the process (Figure 4.2). While we were unable to verify the presence of any isotopes remaining in the column, we could track the movement of those isotopes throughout the glovebox. The process took slightly over one hour (3976 seconds).

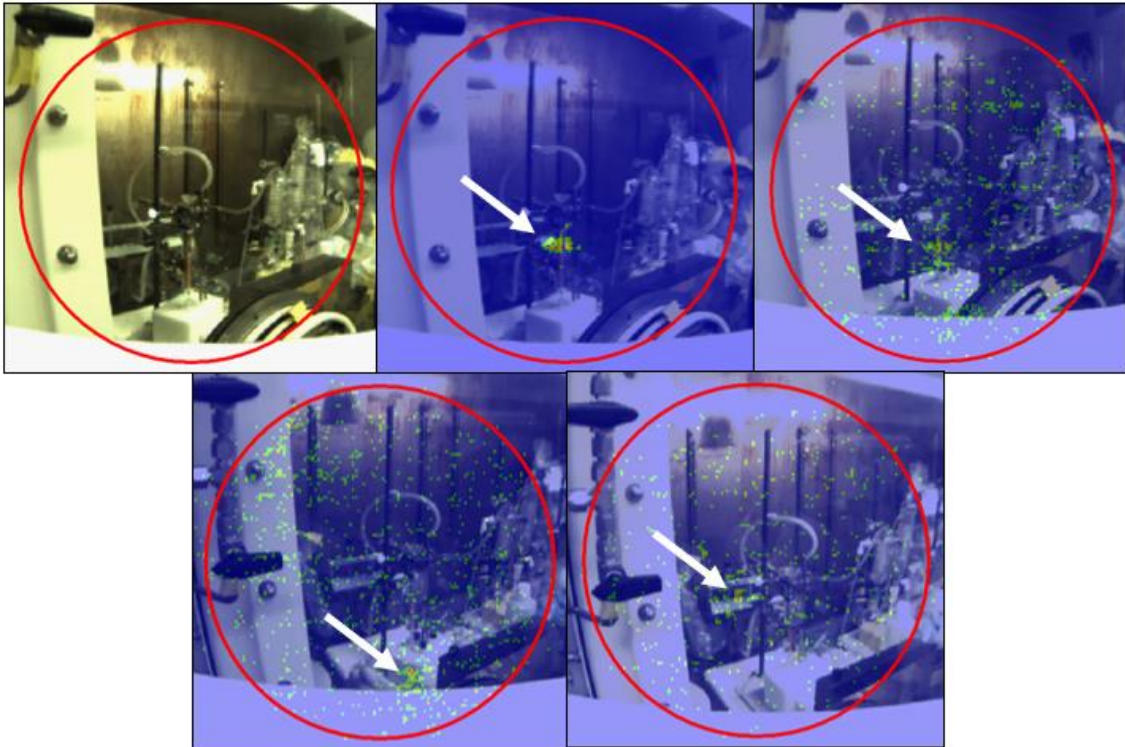


Figure 4.2: Tracking Movement of Isotopic Mixture

Figure 4.3 shows the prominent gamma-ray energies peaks emitted by the berkelium that allowed for position tracking the isotope as it moved around the glove box. Of note, during these experiments, the GeGI was able to identify both the presence and location of the berkelium isotope between 16 and 23 seconds after activation. This was possible for several reasons including proximity to the source, the high activity of the isotopes being observed, and finally higher energies of the gamma-ray emissions. Both einsteinium and berkelium have short half-lives making them significant sources of gamma-ray energy, however their SNM counterparts are much longer lived which presents a much lower activity scenario. Furthermore, the gamma-ray energies emitted by SNM are significantly lower than was studied in this experiment and will make imaging those isotopes a greater challenge using Compton imaging techniques.

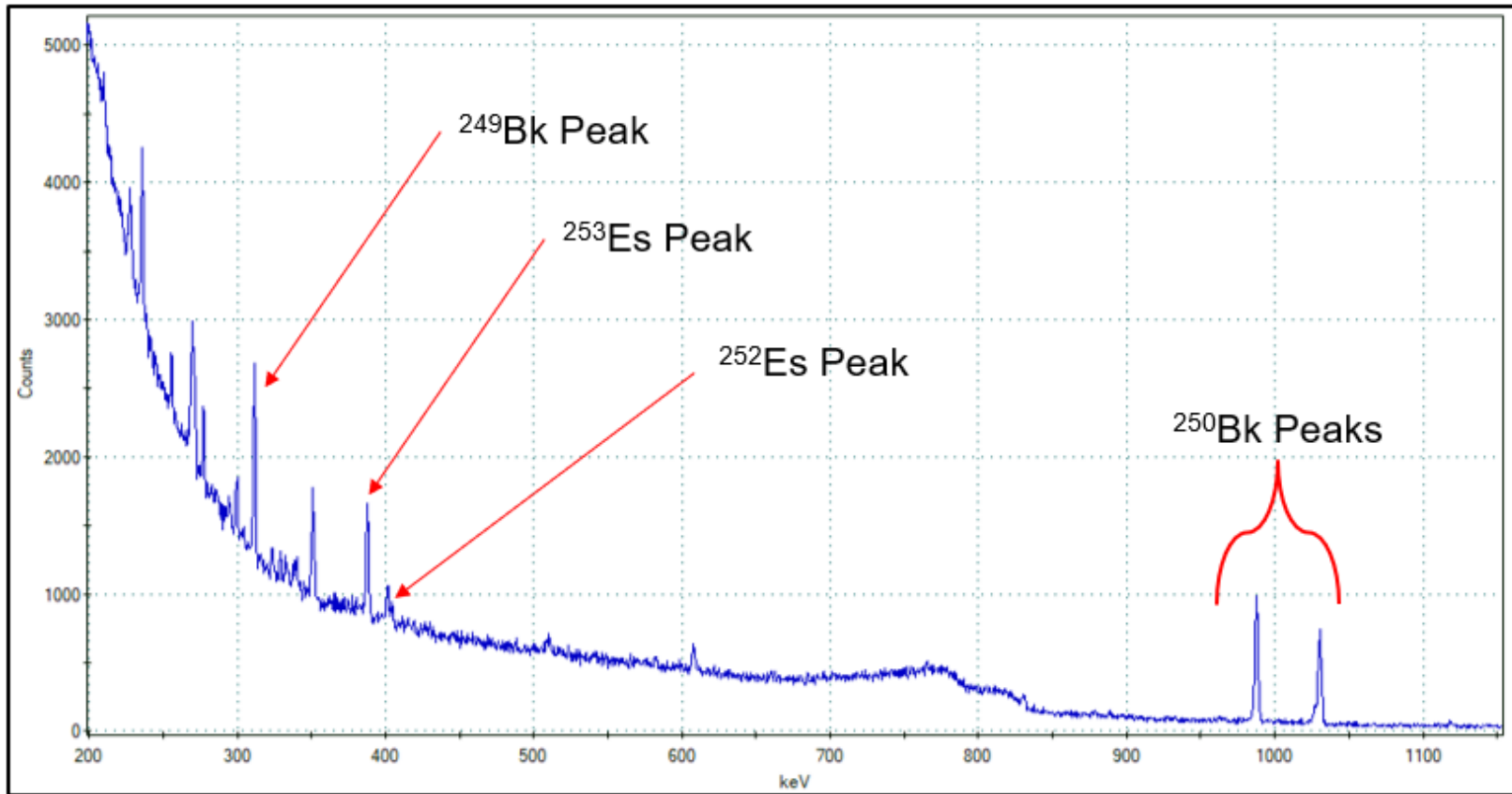


Figure 4.3: Gamma-Ray Spectrum for Es – Bk Experiment

While we were able to observe the movement of the mixture around a glovebox using high activity isotopes, we were unable to witness the separation of the isotopic mixture. This unobserved component of the process was one of the requirements to demonstrating the GeGI's ability to verify separation processes from a standoff distance, without the need for process interruption for classical alpha decay testing. Challenges faced during this experiment included a novice level of understanding of the GeGI's full spectrum of capabilities, a limited physical window from which to view the execution of the separation, and the limited reproducibility of this experiment due to a low quantity of einsteinium available.

4.2 Actinium – Radium Separation Column

One of the challenges that we noticed during our einsteinium – berkelium experiment was a lack of overall resolution after the separation began. To better test or detector's ability to isolate the movement of isotopes within a column we executed another separation involving small distances. We also began to move closer to the gamma-ray emissions expected of SNM by using lower activity sample with less energetic gamma-rays.

This separation would focus on medically useful isotopes and would only examine the final portion of the diagram in Figure 4.4.

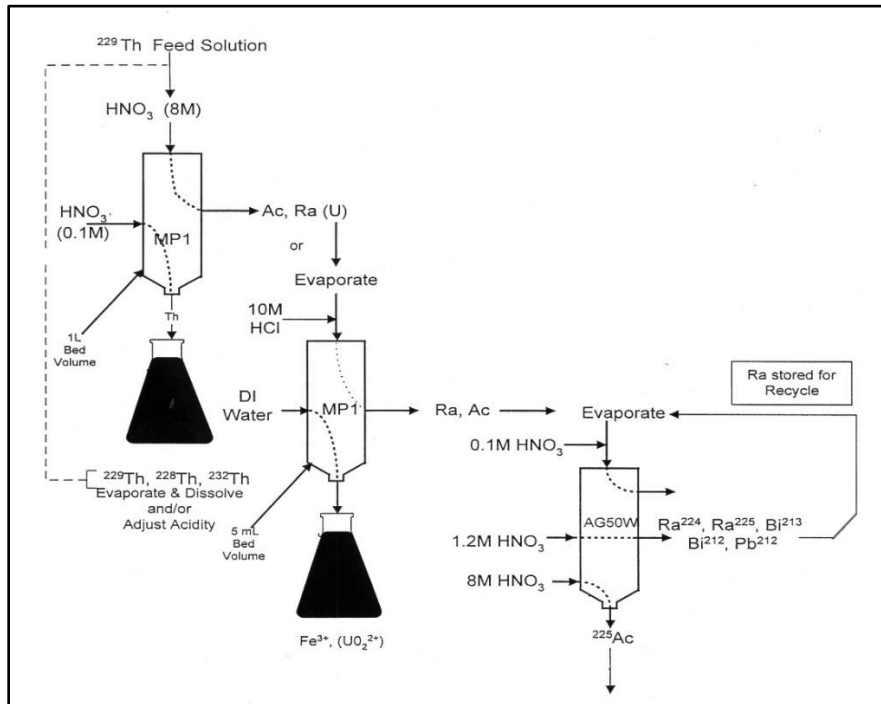


Figure 4.4: ORNL Ac – Ra Separation Flow Diagram [9]

In this process, the separation would attempt to purify a sample of actinium-225 (^{225}Ac) for use in the treatment of cancer. In order to accomplish this, the actinium daughters and other non-necessary isotopes would be removed allowing for the capture of purified ^{225}Ac to be collected. This particular experiment was very time sensitive as shipment of the collected actinium was scheduled for only a few hours after the start of the separation. Given this time constraint, accurate measurements were a necessity as the separation would not be available for some time and the process could not be slowed in any significant way.

In our experiment we were able to witness, from roughly a meter standoff distance, the detector's ability to expertly identify isotopes, track the movement of isotopes, and maintain excellent spatial resolution over the course of our experiment (Figure 4.5). The entire measurement took roughly one hour (3624 seconds) from our first measurement to our final measurement. The gamma-ray spectrum emitted by these isotopes is recorded in figure 4.6.

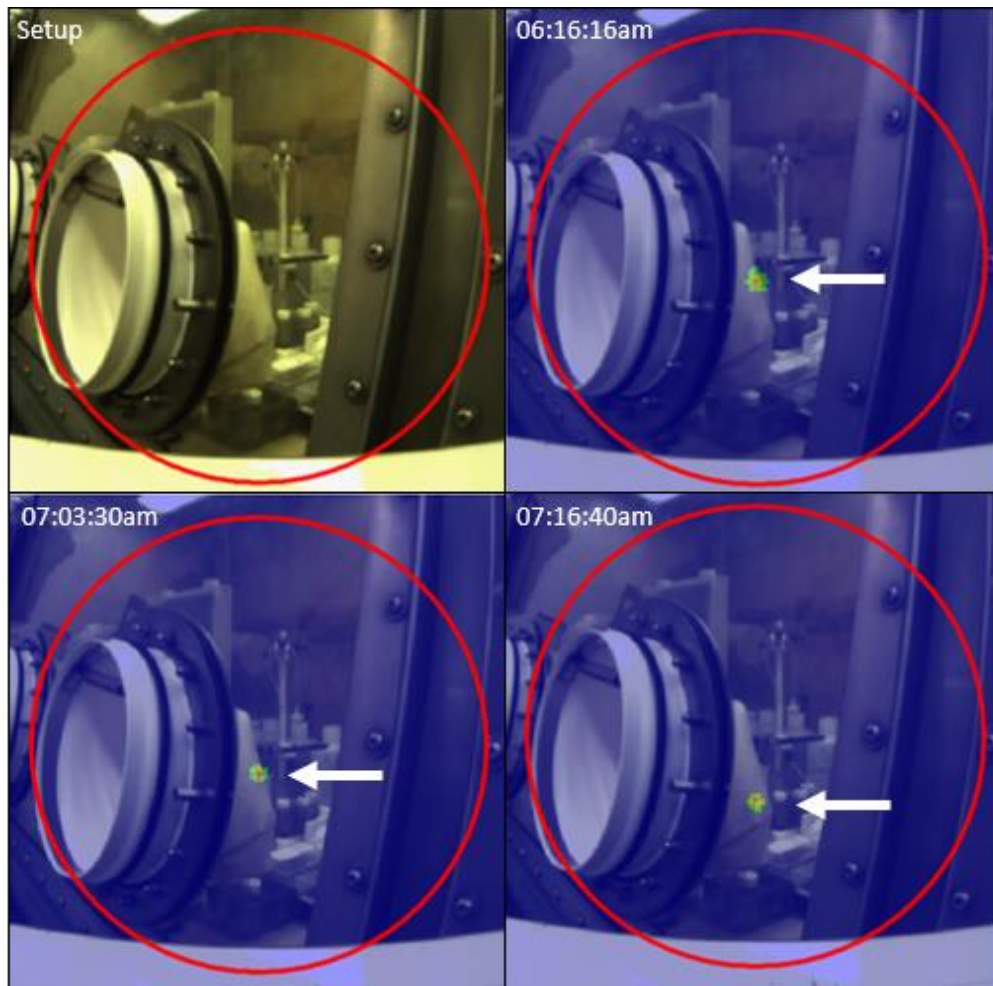


Figure 4.5: Improved Spatial Resolution of Detection

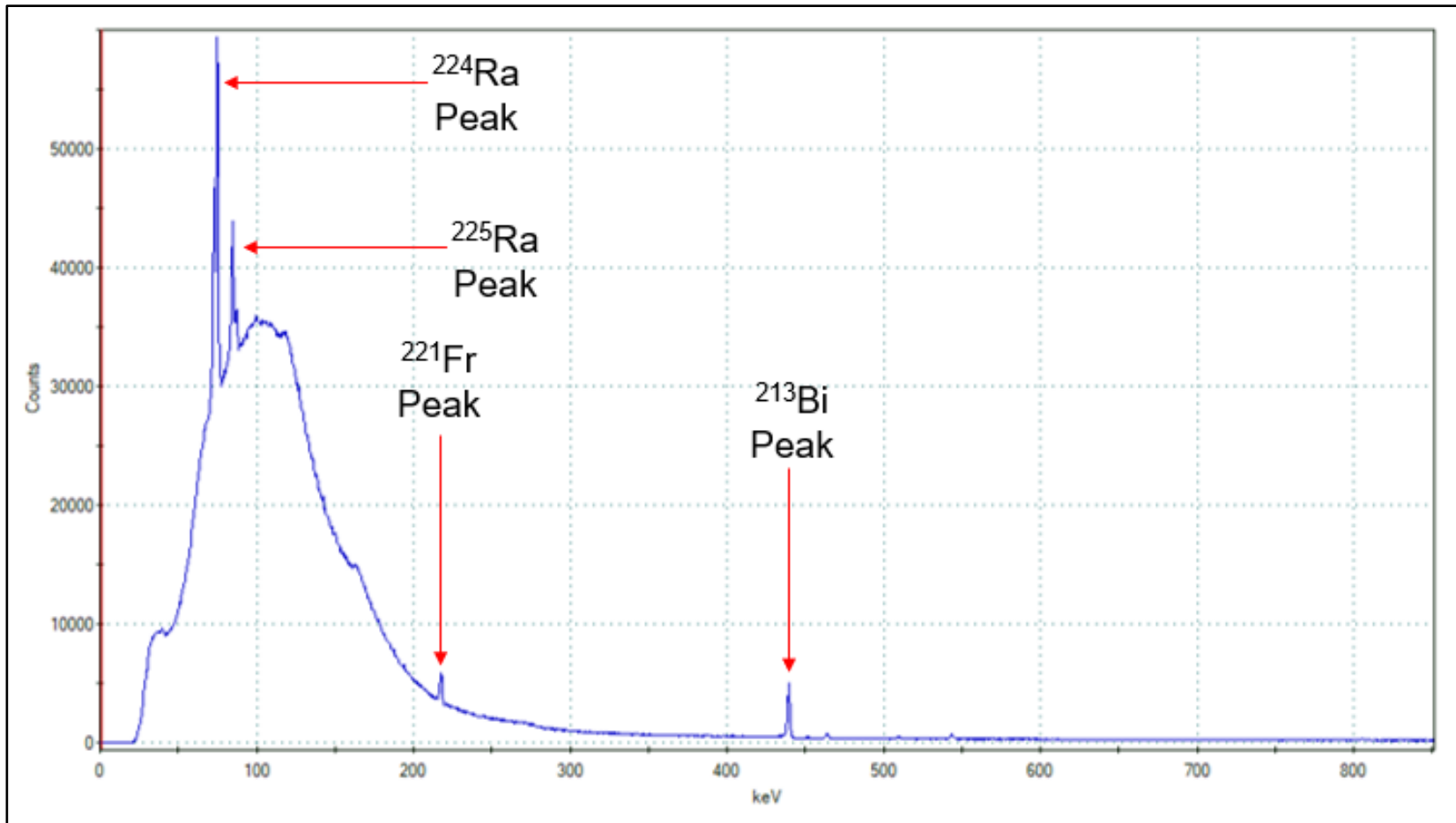


Figure 4.6: Gamma-Ray Spectrum for Ac – Ra Experiment

Furthermore, as verification of the detector's ability to identify isotopes both in front of and behind the detector, we moved the sample from the GeGI's forward field of view to the device's rear at a distance of roughly three meters. The GeGI was able to detect the movement and isolate the location even without a collimator or photograph available in that direction (Figure 4.7). One can see that the two gamma-ray peaks identified in the rear-view correlate with francium-221 (Auto:216) and bismuth-213 (Auto:440).

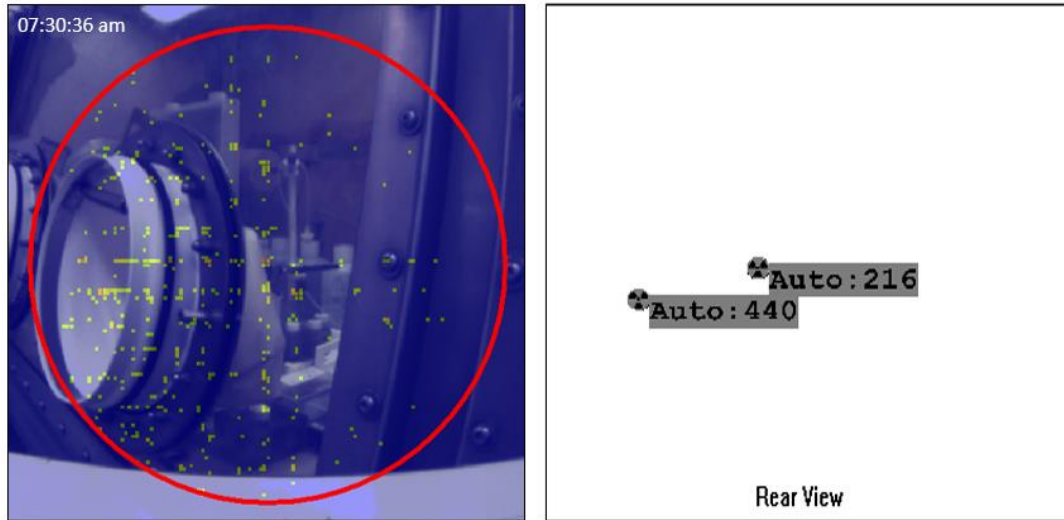


Figure 4.7: Catchment Vessel Transfer to Rear of Detector

4.3 Plutonium – Neptunium – Americium Separation Column

With our final experiment, we had the goal of putting all of the elements of previous experiments together into one final isotopic separation. The mass of isotopic mixture used was several hundred times smaller than industrial scale, however 1.96 grams of Plutonium 239 (^{239}Pu), 5.74 grams of Neptunium 237 (^{237}Np), and 13 milligrams of Americium 241 (^{241}Am) were on hand for this scaled experiment.

In the event this experiment was also conducted in an effort to date the Plutonium contained within the mixture, this could be accomplished through an application of the “Bateman Equations” (Figure 4.8) and known decay chains.

$$\frac{dN_i(t)}{dt} = \sum_j \left[\int_0^\infty \gamma_{ji}(E, t) \sigma_{f_j}(E, t) \phi(E, t) dE \right] N_j(t) + \sigma_{c_{i-1}}(E, t) \phi(E, t) N_{i-1}(t) + \lambda_{i'} N_{i'}(t) - \left\{ \left[\int_0^\infty (\sigma_{f_i}(E, t) + \sigma_{c_i}(E, t)) \phi(E, t) dE \right] + \lambda_i \right\} N_i(t)$$

Figure 4.8: Bateman Equation for Isotope Creation and Decay [10]

The known decay chains of plutonium isotopes are shown in Figure 4.9. The experimental setup for this separation is shown in figure 4.10 and contains all of the isotopes identified in the beginning of the section.

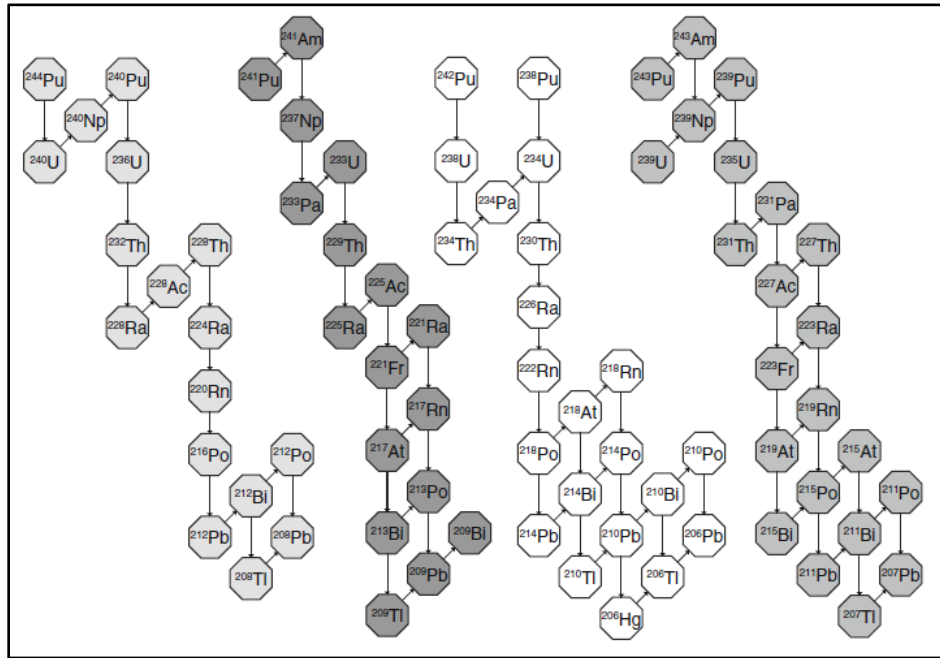


Figure 4.9: Plutonium Decay Chains

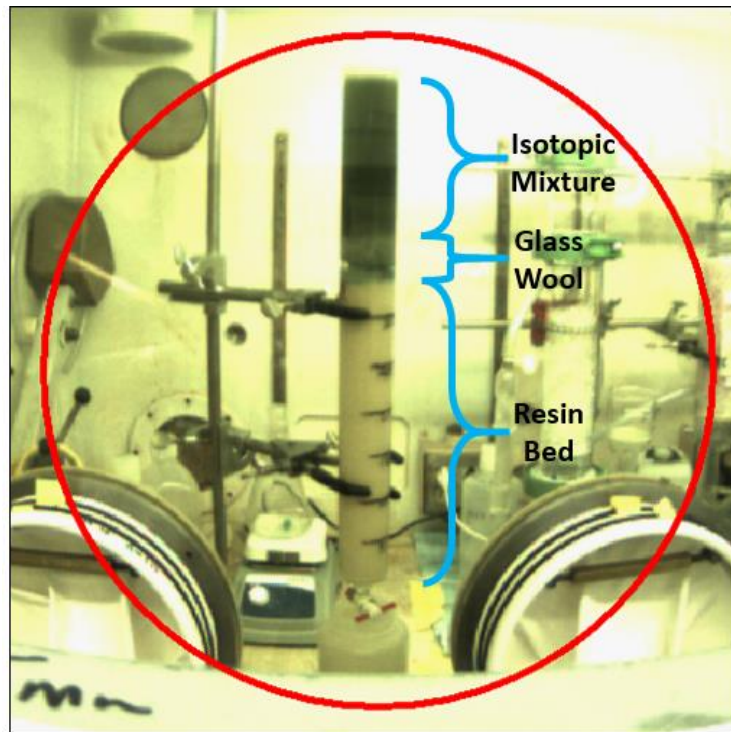


Figure 4.10: Loaded Column Prior to Separation Attempt

This experiment however was not naturally aged plutonium therefore no aging calculations would be worthwhile in this scenario. Our isotopic mixture was doped with its americium and neptunium instead of allowing the negligible mass of plutonium-241 (^{241}Pu) to decay into ^{241}Am and subsequently ^{237}Np .

This separation would attempt to show evidence to determine the plausibility of using a gamma-ray detection system to provide a means of standoff detection and process verification in nuclear safeguards. Success for this exercise would be based on our ability to show that in real time we could witness the extraction of ^{241}Am from the other isotopes which pose a proliferation concern. Figure 4.11 shows the relative location of all isotopes prior to the separation phase of the experiment. The gamma-ray spectrum emitted by these isotopes is recorded in figure 4.12.

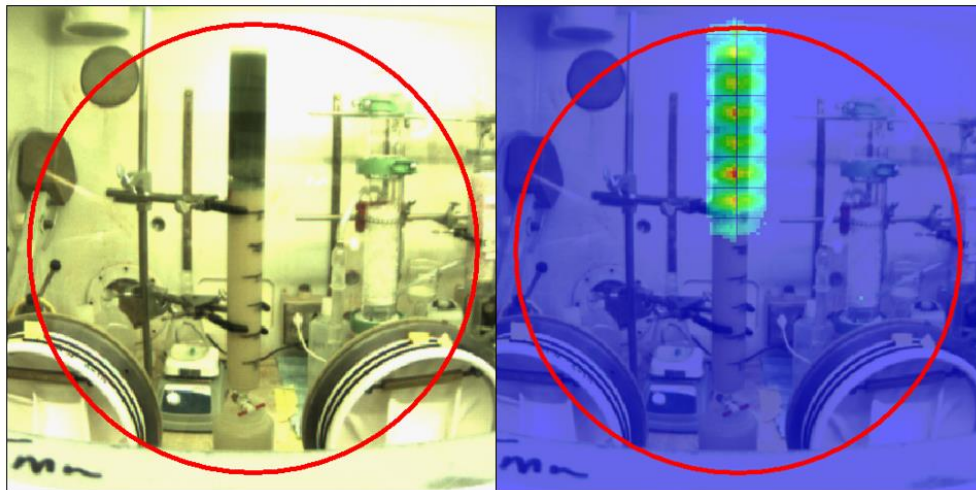


Figure 4.11: Isotopes Locations Within the Column Prior to Extraction

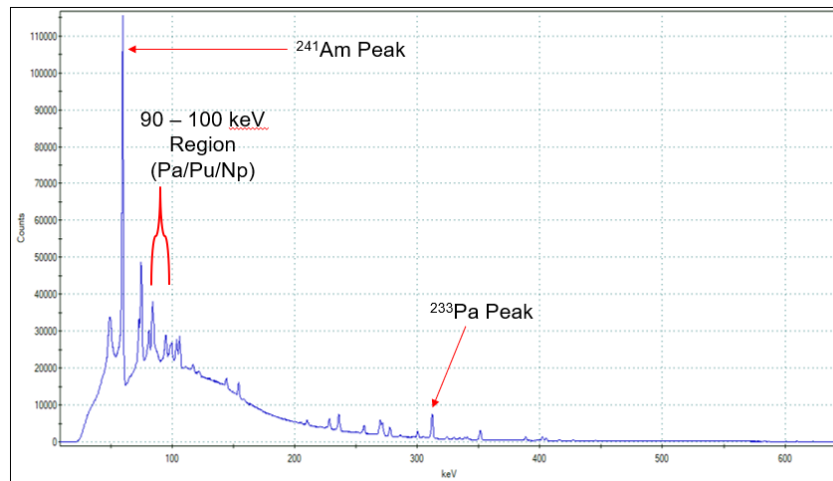


Figure 4.12: Gamma-Ray Spectrum Prior to Extracting Isotopes

As our separation progressed, we noticed the successful extraction of the ^{241}Am while verifying the ^{237}Np and ^{239}Pu remained in suspension (Figures 4.13 and 4.14). The overwhelming 59.5 keV peak shown in the previous figure is beneficial in ensuring the successful extraction of the ^{241}Am .

Subsequent alpha particle analysis showed that 99.23% of the ^{241}Am was extracted without the removal of SNM isotopes. The gamma-ray spectrum emitted by these isotopes without the ^{241}Am contamination is recorded in figure 4.15.



Figure 4.13: Beginning of Americium Extraction

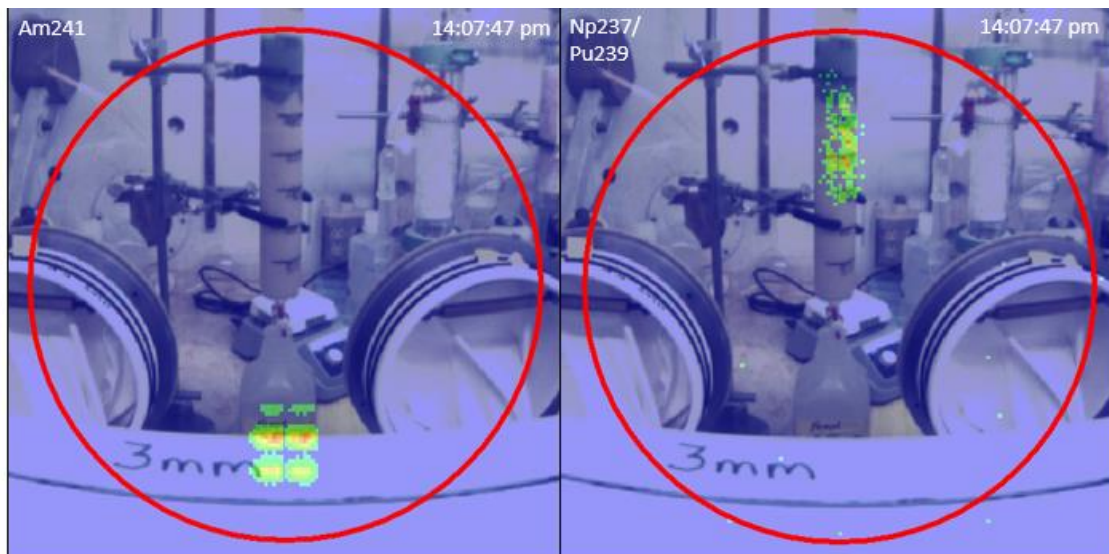


Figure 4.14: Verification of ^{241}Am Extraction and SNM Suspension

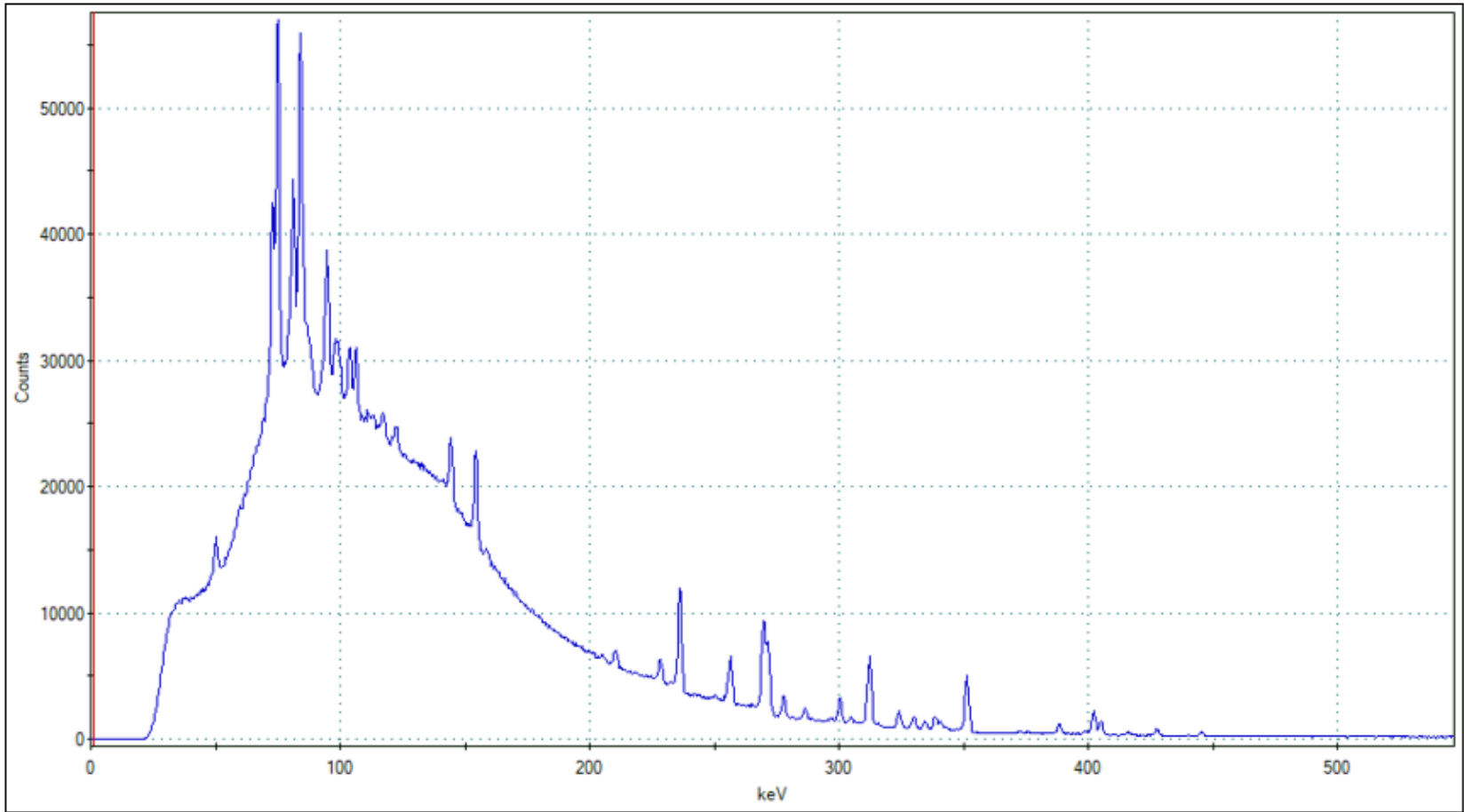


Figure 4.15: Gamma-Ray Spectrum Following Extraction of ^{241}Am

Of note, during these separations, there was ingrowth of protactinium-233 (^{233}Pa). There were both benefits and drawbacks to this ingrowth. The primary benefit was that the ^{233}Pa could help show where its parent ^{237}Np was radiating from. However, it did offer a challenge of creating several conflicting gamma-ray energies that could mask both ^{237}Np and ^{239}Pu since those isotopes are far more long-lived than ^{233}Pa which has a half-life of only 27.4 days. With only gram quantities of both ^{237}Np and ^{239}Pu , the non-conflicting gamma-ray lines were hidden within the gamma-ray background. Figure 4.16 shows the spectrum used to locate the ^{237}Np and ^{239}Pu .

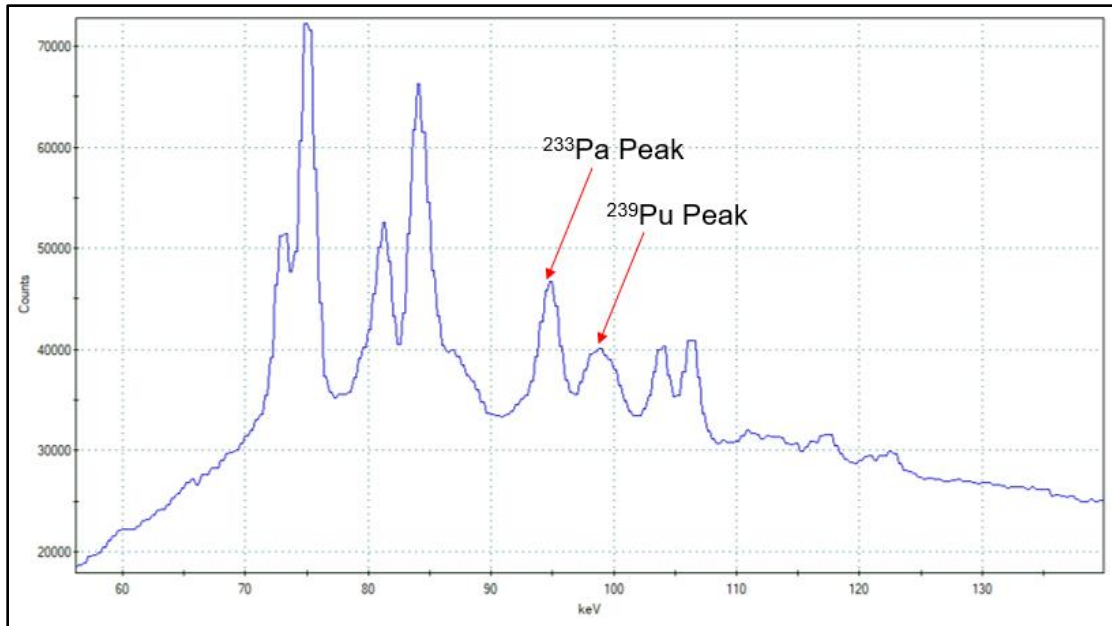


Figure 4.16: Gamma-Ray Spectrum of ^{237}Np (via ^{233}Pa) and ^{239}Pu

The lower energy gamma-ray emissions of the $^{237}\text{Np}/^{233}\text{Pa}$ and ^{239}Pu , coupled with their relative closeness in energy (95.5 keV and 99.0 keV) did challenge the detector in localization. Figure 4.17 shows the $^{237}\text{Np}/^{233}\text{Pa}$ is located towards the top of the column and the ^{239}Pu is located towards the bottom. Figure 4.18 shows the detector did note the center of concentration of each isotope correctly, however did require some interpretation of the $^{237}\text{Np}/^{233}\text{Pa}$ location because of the “smear” of $^{237}\text{Np}/^{233}\text{Pa}$ counts shown in the ^{239}Pu region of the column.

The anticipated order of extraction on this separation was ^{241}Am , ^{239}Pu , and $^{237}\text{Np}/^{233}\text{Pa}$ respectively. After completion of the entire separation, low background alpha particle spectroscopy and gamma-ray spectroscopy showed that the ^{241}Am was extracted without notable contamination. Furthermore, the ^{237}Np and ^{233}Pa were extracted together. Contrastingly, while much of the ^{239}Pu was extracted without contamination from the ^{237}Np and ^{233}Pa , there was an

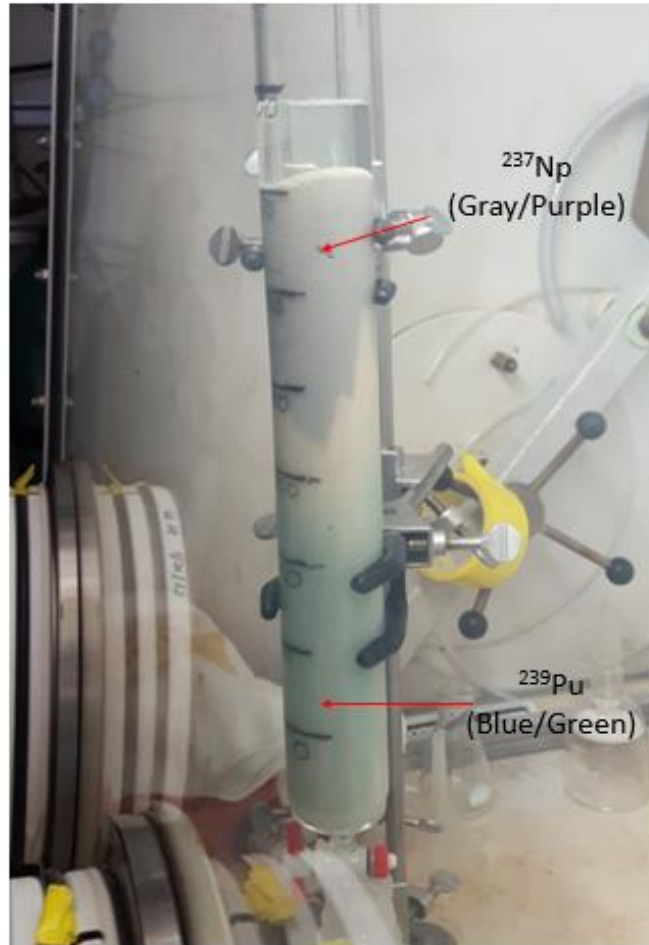


Figure 4.17: Column Showing Bands of Color and Isotope Locations

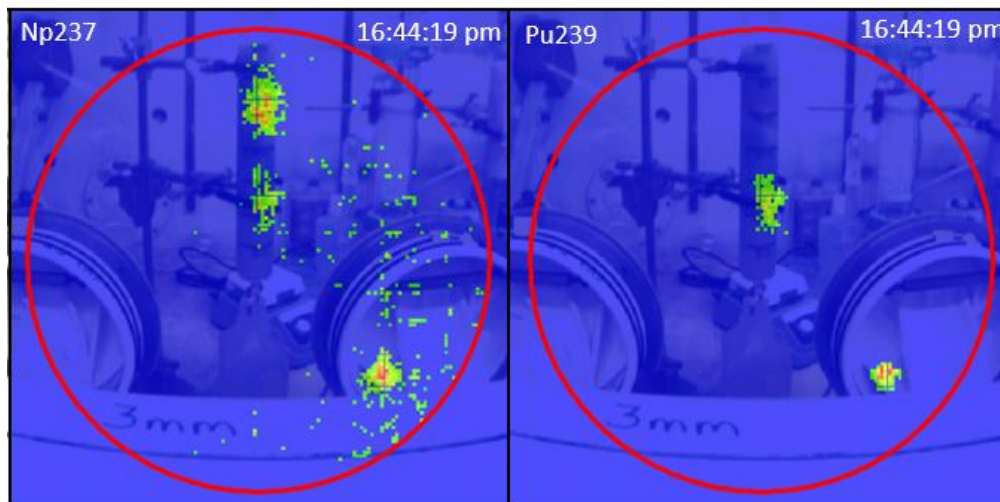


Figure 4.18: SNM Suspension in Separate Column Locations

equal amount of the ^{239}Pu that remained with the ^{237}Np and ^{233}Pa . This situation suggests an efficient extraction of the ^{241}Am , and purity of the ^{239}Pu extracted prior to the ^{237}Np and ^{233}Pa coming off the column.

As described previously, this isotopic mixture was doped with significant amounts of ^{237}Np which altered the state of secular equilibrium that would most likely exist in a properly aged plutonium sample. While industrial scale operations would offer different challenges to measurement, resolving the physical locations of the ^{237}Np and ^{239}Pu could be less challenging when using more non-conflicting gamma-ray energies.

CHAPTER FIVE

CONCLUSIONS AND RECOMMENDATIONS

5.1 Applications to In-situ Verification of Radiochemistry Processes

In our limited experiments, we were able to demonstrate the capability to monitor a process by visually witnessing, in real time, the separation of specific isotopes from a mixture of SNM. Such a system could enhance security of process monitoring by providing inspection teams the ability to watch the process and not simply the before process and after process comparison. Such a novel system could also allow inspectors with lesser levels of training to observe processes with greater confidence and less need to collaborate with outside resources.

While the application of this process monitoring method is useful when evaluating outside entities and governments on the truthfulness of their process outcomes, this methodology would be equally useful as our own government expands its production goals with plutonium purification and casting for the first time in decades. Given the potential loss in personnel experience that would come with a long absence in industrial scale plutonium production here in the United States, process verification methods such as this one could prove invaluable as scientists rediscover these skills or attempt different purification methods.

No matter what the application is however, the user is more result would available in real-time as the experiment is occurring and would involve much less data interpretation as currently available methods. Furthermore, visualization of processes could help refine procedures and reduce waste, as we better understand the physical processes occurring by pairing our current analysis methods with the visual data provided with this method.

5.2 Applications to Facility Verification

As discussed earlier in this document, significant progress has been made in both static and mobile versions of the stand-off gamma-ray facility verification systems. These roving systems however, appear to have been tested using sources emitting higher energy gamma-rays or more active gamma-ray sources which tend to be easier for Compton scattering based imagers to isolate and localize. Integrating the system tested in our experiments with a facility

verification scenario involving more than one isotope would help verify that these systems would be useful in real-world settings.

These facility verification inspections would involve verifying that known facility plumbing has not been removed and not additional plumbing has been added. Any added plumbing would typically be tested to ensure it is not diverting any material from the originally designed intent of the facility. However, by integrating our imaging technique with the facility mapping techniques developed by others, a greater level of confidence is obtainable. In fact, verifying that pipes are carrying the exact isotopes that they are permitted to be carrying would be possible. This added level of verification would integrate plumbing verifications with isotope diversion prevention.

5.3 Future Work

While our experiments met with some successes, there were many areas identified that need greater study. The challenges presented by SNM and other weakly emitting sources requires greater study. As the SNM we tested tended to produce gamma-ray energies below 200 keV, and also tended to have greater half-lives than the other non-SNM isotopes we experimented with, our technique requires refinement. While industrial scale manufacture of SNM would produce greater quantities of gamma-rays for collection, the stand-off required would be greater. An experimenter could potentially overcome this issue through the use of pinhole zoom techniques (changing focal length of the detector) or by sacrificing the detector itself to work inside the contaminated area. Either of these techniques would require more time and funding to determine the feasibility of either method.

Furthermore, deconvolution of the gamma-ray emissions below 100keV would prove useful when working with SNM and other sources that emit lower energy gamma-ray signatures. A deconvolution method might provide a better means to isolate peaks that are less prominent than are idea to work with. However, conducting this experiment in an environment of lesser background gamma-ray emissions would be useful. Conversely, collecting data on the background gamma-ray spectrum and then subtracting that data from the experimentally collected spectrum could offer a less elegant, yet equally effective means of reducing unwanted interference.

Undoubtedly, if more research is conducted into this method more challenges and opportunities for employment of this technology and technique will present themselves. While the financial and man-hour cost for this research may be higher than simply continuing use of our current verification methods, it does present the opportunity to improve our separation and verification methods,

better secure sensitive radiological material, and reduce the time necessary to identify potential security threats. Each of these benefits warrants additional attention, however which requirement is most important will allow us to focus our effort on a specific area of research.

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APPENDIX

Appendix A: Control Data Sheet for GeGI Used in Experiments

Technical Quality Assurance Verification
(PHDS Co. form TQAV051215)



3011 Amherst Road
Knoxville, TN 37921
(865) 202-6253
www.phdsc.com

Quantity	Value	Initials	Date
System Number	GeGI 413	MRK	11/13/17
Mass (kg)	15 kg	MRK	11/13/17
Standby/Operating Power (W)	78 W / 94 W	MRK	11/13/17
Internal Battery Number/Type	2 x PH3059HD25 (Li-Ion)	MRK	11/13/17
Operating Battery Life	50 minutes (single battery; > 90 minutes on 2 batteries)	MRK	11/13/17
Charger function	OK	MRK	11/13/17
Samlex AC Power Setting	120 VAC / 60 Hz	MRK	11/13/17
Transition Line -- Battery	OK	MRK	11/13/17
LED Function	Green, Yellow, Blue OK	MRK	11/13/17
Master On/Off switch	OK	MRK	11/13/17
Temperature cycle integrity	OK: 05/04/16	MRK	11/13/17
Crystal Dia./Thickness/Active area	90 mm / 10 mm / 55 cm ²	MRK	11/13/17
NxN Strips/Pitch/Gap	16 x 16 / 5.0 mm / 0.125 mm	MRK	11/13/17
Crystal number/N/(cm3)/type	<070115> S2 C1 / mixed typeness (n)	MRK	11/13/17
Depletion Bias/Operation Bias	V _{dep} = -950V; V _{op} = -1200V	MRK	11/13/17
Cooler serial number	MT8-08	MRK	11/13/17
Cooler card number/generation	#50032217002	MRK	11/13/17
Cooler Set Temp/Diode Temp	73K / 79K	MRK	11/13/17
Cooler Max Power	85 W	MRK	11/13/17
Approx. cooler time to date	~30 months	MRK	11/13/17
Power Controller number	New power control board; ID not populated (Imager32)	MRK	11/13/17
Firmware (Power Control) version	20170227-0845	MRK	11/13/17
SPECT32LT number/FPGA rev.	#41 P3	MRK	11/13/17
Ion pump current (RT/Cold)	40 μA / 0.05 μA	MRK	11/13/17
FPGA Version (Mother / Daughter)	Mother: 2015042852 Daughter: 2010111250	MRK	11/13/17
Pin Hole distance (mm)	-65 mm	MRK	11/13/17
Camera number	6436	MRK	11/13/17
Cart Fit (Tripod)	Fit OK on PHDS Pinhole Imaging Plate	MRK	11/13/17
External Battery	OK on ORNL Battery	MRK	11/13/17
Ae ² + Be +C = FWHM (All Events)	A = 1e-9; B = 0.0014; C = 1.2339	MRK	11/13/17
Pixel total FWHM 122/356/662 keV	1.40 keV / 1.62 keV / 1.80 keV	MRK	11/13/17
Composite Flood-Compton file	OK: f 11 10 17 f8	MRK	11/13/17
Location 10 uCi, 1 m, 100 sec	OK: f 11 13 17 f2	MRK	11/13/17
Compton/Sci/Tactical/Rearview	OK: f 11 13 17 f1	MRK	11/13/17
Detector Configuration Depths	7 - 16	MRK	11/13/17
Compton event ep /interpolate sep	1-16 / 1-16	MRK	11/13/17
Compton position uncertainty	(0.5 mm, 0.5 mm, 0.5 mm)	MRK	11/13/17
Set Serial Number (control board)	OK: 413	MRK	11/13/17
Teamviewer Installed	OK on tablet	MRK	11/13/17
Delete Non-Essential Dell Apps	N/A (no laptop)	MRK	11/13/17
Imager32 (version) / settings file	3.16.11.0904/ "G413 Settings 11 13 17.ini"	MRK	11/13/17
PC Serial Number	Tablet: 013804534753	MRK	11/13/17
PC numbers / passwords	Tablet: PHDS033015S; no password	MRK	11/13/17
PC updates off / Power settings	OK on tablet	MRK	11/13/17
Screen Capture file	"G413-Screenshots-20171113.pdf"	MRK	11/13/17
PDF manuals (Imager32, GeGI)	On tablet (and hard copies delivered)	MRK	11/13/17
Repairs/upgrades/NRE	New power control board and accessible Li-Ion batteries	MRK	11/13/17

VITA

Mark S. Quint has served as an Officer in the United States Army for the past twelve years. He is a 2007 graduate of the United States Military Academy at West Point, New York with a B.S. in Mechanical Engineering. Major Quint has served in a variety of staff and leadership roles in the Army and has been stationed both at home and abroad.

Major Quint is married to the former Jennifer Spaulding of Winslow, Maine. They have one daughter, Charlotte, and he and Jennifer are expecting their second child in May 2018.