Implementation of a System of Gamma Imagers for Measuring Plutonium Holdup

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Abstract

The Surplus Plutonium Disposition (SPD) project is an effort to dilute and dispose of many tons of weapons grade plutonium. This work will take place in a shielded glovebox to protect workers from radiological dose. Accurate measurements of the holdup, or material left behind, in the gloveboxes is important for Nuclear Material Control and Accountancy, worker dose minimization, criticality safety, and process monitoring. Shielding around the gloveboxes presents an obstacle for the traditional Generalized Gamma Holdup (GGH) method. The uncertainties associated with that method, at around 28%, are also detrimental since the amount of holdup must be kept below an established limit with a high level of confidence.

Passive gamma-ray imaging provides a more complete depiction of the distribution of radioactive materials. Recent refinement of the method has demonstrated that quantitative information can be extracted from the images with a well-characterized system. A permanently installed system of gamma-ray imagers is currently being developed to measure plutonium holdup in the SPD gloveboxes. This paper will report on progress with the development of this system.

This application requires that the features of current commercially available instruments from H3D and PHDS, optimized for measurement performance in a mobile application, be translated into the constraints of the operational environment with fixed installations. The interaction between facility constraints and measurement capabilities will be addressed.

A series of measurements with plutonium sources in a mocked-up glovebox geometry with imagers mounted above the ceiling of the glovebox has been taken. Imager performance with respect to angular resolution, minimum measurement time for quantification, and signal-to-noise ratio with a multitude of sources will be evaluated for those measurements. A series of prototypes of user interfaces will be presented for display of the data to its various consumers for the purposes of Nuclear Material Control and Accountancy, worker dose minimization, criticality safety, and process monitoring.

1 Introduction

In special nuclear material processing, the presence of holdup, or material unintentionally left behind in equipment and ductwork, has broad consequences: Holdup material does not reach the expected location, resulting in a potential error in material accountancy; with sufficient quantities of fissile material, there could be a criticality hazard; holdup can be a significant source of radiological dose to personnel; and an expectation that unmeasured holdup will occur also presents an unwanted opportunity for small amounts of material to be diverted without detection. For these reasons, understanding and controlling holdup accumulation is important to the operation of a nuclear facility.

To assess holdup mass using passive γ-ray measurements, it is necessary to know the intrinsic efficiency of the detector, the distance to the source, the distribution of the source, the attenuating effects of intervening materials (including self-attenuation), and the contribution of backgrounds to the measured count rate. Assay using gamma rays has the advantage that detectors with high energy resolution are available and emission rates are high enough to use relatively small, transportable detectors. The Generalized Gamma Holdup technique [5] has been developed for this purpose and has enjoyed widespread use. This technique suffers, however, from uncertainties in correcting for attenuation, the distance and distribution of the deposit, and backgrounds from neighboring sources.

Gamma-ray imaging presents multiple advantages over the established gamma-ray assay methods by providing precise location and distribution information for deposits, and making it possible to distinguish between separate deposits in the field of view. Gamma-ray imaging instruments, which are now commercially available from PHDS Co. and H3D, Inc., use two imaging modalities: Compton reconstruction at higher energies and the coded-aperture technique at lower energies. To support holdup measurements for the Surplus Plutonium Disposition project, we are working to develop a system of these imagers and associated analysis tools. This system is intended to be retrofitted onto three gloveboxes for the purpose of plutonium oxide holdup localization and mass measurements.

A permanently installed system of imagers with automated data analysis tools would provide several advantages. By virtue of fixed installation, the measurement system would be available for measurement without setting up portable equipment, reducing worker dose and reducing time required to prepare for each measurement. Holdup measurements could be taken at any time when process material is not present. The position and orientation of the detectors would be known precisely and the distance to holdup locations would be fixed. Automated data analysis could further reduce the time and user intervention required for measuring holdup. This means that holdup measurements could be performed at a higher frequency with minimal incremental cost, and systematic errors due to poorly known detector and deposit geometries would be reduced.

For the best spectroscopic imaging performance, the imaging system based on the PHDS Co. GeGI is attractive. That device is based on a high purity germanium detector, and has the advantages of large active surface area and superior energy resolution in comparison to the CdZnTe-based H420 imager from H3D, Inc. The size, 4-hour cool-down time, and mechanical complexity of the germanium instrument, however, are not fit for the limited spatial envelope and concept of operations envisioned for this application. Consequently, this work is focused on developing a system based on the H420 imager, which has acceptable energy resolution and approximately 19 square centimeters active area.

The measurement system described here is intended to measure holdup in a glovebox where plutonium oxide powder is diluted and repackaged. Activities taking place inside the glovebox include opening containers, weighing incoming material, sieving, grinding where necessary to reduce particle size, depositing in blend cans, mixing, and sealing containers. This process is currently being carried out with relatively small manual tools, but larger and more intricate equipment is anticipated to be in use by the time a system of imagers could be installed in a glovebox. While additional equipment should reduce touch time and worker dose, it also presents greater opportunity for holdup accumulation.

Holdup is likely to occur in outlet air filters, in crevices in equipment, on any horizontal surfaces, on gloves, and any location that is not readily accessible for cleaning. This means that the measurement system should be sensitive to sources in any of these locations. These surfaces are predominantly near the floor of the glovebox, but extend to the ceiling in some places, for example where conduit, ductwork, and electrical outlets hang down from the glovebox ceiling.

This paper introduces the envisioned measurement system and data analysis strategies, and describes the concept of operations that is being pursued.

1.1 Gamma Ray Imaging Methods

The gamma ray imagers being employed for this effort are based on a position-sensitive detector, a coded-aperture mask, and a read-out scheme that is suitable for imaging using both Compton and coded-aperture imaging methods. These two methods are complementary. Compton-reconstruction imaging relies on multsite events, is capable of imaging in all directions, and is sensitive at energies above around 250 keV for the detector material being considered here. Coded-aperture imaging provides superior angular resolution, but is limited to a narrower field of view, and, since it uses single-site interactions, is most sensitive at photon energies below 500 keV. More detailed descriptions of these methods are abundant in the literature (see, for example [3] and [2]) but are summarized here.

In Compton imaging, kinematic constraints are applied to the measured position and energy depositions for multi-site events. For each event, a determination is made for chronological order of the interactions, the axis defined by the first two interactions is found, the angle of the incoming photon to that axis is determined, and then a cone can be drawn that defines the incoming trajectories that could have produced that interaction pattern. The cone defines a circle (or annulus, if uncertainties are considered) that is the locus of intersection between the cone and the image surface. The image surface might be a plane or a spherical surface with some radius, centered on the detector. After the accumulation of many events, the overlap of these circles or annuli reveal the position of a source. This method is most sensitive for photon energies where multi-site events are abundant, that is, where Compton scattering prevails over photoelectric absorption. The angular resolution that can be achieved is typically around 20 degrees, dominated by the uncertainty of the interaction locations creating uncertainty on the direction between the sites . Iterative methods have been shown to improve precision for source localization to around 5 degrees, but resolution of sources still requires a minimum angular separation around 20 degrees.

Coded-aperture images are constructed from the shadow cast onto the detector by a patterned mask. In the typical application, the mask length and width are chosen to be double the size of the detector, so that as the source shines through the mask, it casts a shadowgram onto the detector from one quarter of the area of the mask. The phase of the shadowgram reveals the direction to the source. If, for example, the pattern on the detector corresponds to the central area of the mask, the source is determined to be located at the center of the field of view. The mask pattern must have the property that each phase of the pattern is distinguishable from all the other pattern phases. The contrast of the detected pattern varies with the material and thickness of the mask. The resolution of this method depends on the parameters of the mask and the separation between the mask and the detector, but for the H420 with the image plane one meter from the imager, spatial resolution is 9 cm over a two-meter-wide (90-degree total opening angle) field of view. To observe the shadow pattern requires that closed mask pixels stop the incoming radiation, and knowing where a radiation particle's initial interaction with the detector occurs. The method is thus most effective for lower photon energies where photoelectric absorption is the most probable mode of interaction.

Coded-aperture imaging provides inherent background subtraction when equal-time measurements are made in mask and anti-mask configurations. By inverting the mask and subtracting the shadowgram patterns recorded by the detector removes the unstructured signal from sources outside the field of view and unmodulated gamma rays that penetrate the opaque regions of the mask. In practice this is typically implemented using a mask that is antisymmetric upon ninety-degree rotation. The mask rotation intervals are set to provide equal or nearly-equal duration in each configuration for a measurement.

The width of the coded-aperture resolution, field of view, and sensitivity are competing design considerations. The resolution of the system is given by the mask pixel size divided by the spacing between the mask and the detector, multiplied by the distance to the image plane. The width of the field of view is given by number of elements in the mask along one direction multiplied by the resolution width. This is an indirect imaging technique where all of the counts in the detector are used to create the value for any single image pixel and this means that the signal-to-noise ratio in each pixel of the image depends on the total counts in the detector. This has practical limitations for the imaging dynamic range with strong sources dominating the noise across the image. It works best for point sources and reverts to the sensitivity of a pinhole camera if the whole field of view glows uniformly [3].

For application to measurement of gamma-ray radiation from plutonium, the strengths and weaknesses of these methods are complementary. Coded-aperture imaging provides superior sensitivity to the 129-keV emission, which is the most intense gamma ray, with better angular resolution, over a limited field of view. It is also sensitive to the more-penetrating radiations at 375 and 414 keV. Compton reconstruction is also sensitive at the higher energies, with a field a view that covers all angles, but with poorer angular resolution.

2 System Description

The envisioned holdup measurement system is embodied by four imagers placed on top of each glovebox, looking down onto the glovebox floor. The energy and location information from each interaction in each of the four imagers is provided to a central computer for image reconstruction so that the advantages of overlap-

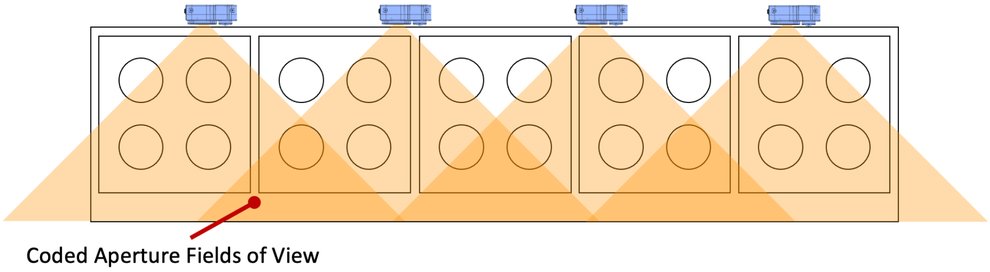

Figure 1: Proposed configuration of gamma ray imagers. Four imagers reside on the ceiling of the glovebox at regular intervals, viewing the floor of the glovebox. The coded-aperture fields of view overlap for areas near the center of the glovebox floor.

ping fields of view can be fully realized. Traditional decoding using correlation between the detected pattern and the mask pattern provides immediate two-dimensional images, and three-dimensional reconstruction can be achieved using a likelihood maximization technique that is currently being explored. We anticipate that by virtue of permanent installation and extracting a three-dimensional map of material location, this system would provide value to the facility through the generation of more frequent and accurate mass assessments, as well as providing improved awareness of material location and dose rates to users.

Each imager will be integrated into a light fixture to allow for retrofitting this technology into existing glovebox designs. In a typical glovebox, a glass plate is mounted to the top of the glovebox with a gasket to provide a containment barrier. A light fixture is then mounted above the glass plate to shine through it onto the floor of the glovebox. Regardless of the particular glovebox function or design, it is generally important for there to be a line of sight from the light fixture to the floor to provide adequate workspace lighting. Furthermore, in cases where the glovebox is shielded, the ceiling is often left unshielded because personnel are not exposed to radiation exiting through the top of the glovebox. This makes the light fixture a strategic location for placing a radiation detection system, providing an unobstructed view to the floor of the glovebox with only modest attenuation for gamma rays.

For a configuration of the system represented by Figure 1, a ninety-degree coded-aperture field of view, imagers placed at one-meter intervals, and a glovebox height of one meter, the entire glovebox floor is in view for at least one imager. Locations near the center of the glovebox floor are in view for two imagers.

In this configuration, portions of the glovebox near the ceiling are not in view for coded-aperture imaging, making Compton reconstruction the only viable method for localization. The poorer angular resolution afforded by Compton reconstruction and the relatively low event rate at 375 and 414 keV is mitigated, however, by the proximity of those regions to the detectors. The position resolution at the furthest locations in the Compton-only region is expected to be approximately 18 cm, in comparison to 12 cm resolution using coded-aperture imaging at the glovebox floor.

2.1 Advantages of a System of Imagers

The overlapping fields of view of the imagers provide several advantages over a series of independent imagers. It is for this reason that the data from each imager should be fed into a central data acquisition system. The stereoscopic capabilities of the system can then be taken advantage of and the raw data can be used for three-dimensional reconstruction using both Compton reconstruction and the coded-aperture method. The list-mode data streaming capability of the H420 accommodates this style of data integration.

As was mentioned before, the knowledge of distance from the source to the detector is a primary contributor to uncertainties for holdup measurements. While coded-aperture imagers have some inherent depth resolution because the mask shadow pattern is magnified at the detector, with the current mask designs each imager is not sufficiently sensitive to source distance on its own. However, three-dimensional location can be achieved by means of triangulation with two imagers, reducing the uncertainty on the source distance.

Measurements from multiple vantage points also provide extra opportunities to view the source with less attenuating material. In the case of a source next to a vertical plate, if one imager detects the source through the plate (which can be assessed by way of differential attenuation between the 129-keV and 414-keV emissions for example) and the other imager has an unhindered line of sight, the location of the source is further constrained and a more accurate mass assessment can be made using the imager with the unencumbered line of sight. A Maximum Likelihood Expectation Maximization has been implemented to realize these benefits, and is being presented at this conference [4].

Overlapping fields of view also provide assurance that sources will be detected if one of the imagers is not available. Sensitivity is expected to be reduced for source locations where the inactive imager has the most favorable line of sight. Locating the imager on the exterior of the glovebox and a simple mounting mechanism is intended to streamline the process for replacing inoperable imagers with spare units.

Greater system reliability and improved depth sensitivity with reduced ambiguities in mass map reconstruction can be achieved through the addition of more imagers at other light fixtures on each glovebox. The costs and benefits of adding additional imagers are being explored.

2.2 Requirements unique to the glovebox application

The design constraints for imagers permanently installed above a glovebox differ from typical commercial applications, where imager portability is desirable. We are exploring custom units that fit into the envelope set aside for the light fixture and do not contain unneeded capabilities. For instance, battery power is not necessary, and is a liability from the perspective of fire protection. A laser range finder is also not necessary for this application. For the sake of maximal imaging performance, it would be advantageous to have a rectangular field of view. This could be achieved with a rectangular mask and a rectangular detector, but the mask would no longer be inverted upon rotation, necessitating a linear motion to perform the mask/anti-mask imaging. These and other considerations are being discussed with the manufacturer for a design tailored to this application.

2.3 Outputs and Users

With the three-dimensional nature of the data that is collected with a system of imagers, and the increased measurement frequency that is envisioned, new data outputs will be possible. The data are expected to be useful for cleaning operations, nuclear material accountancy, criticality safety assurance, radiological dose minimization, and process monitoring. Each imager will, at a minimum, provide two-dimensional images of the distribution of holdup from Compton reconstruction and the coded-aperture technique. Those images will be useful for identifying deposits during cleaning operations and tracking material in process. Constraining the holdup locations would also be helpful for performing measurements with the GGH method and a collimated detector. Three-dimensional maps will be a necessary input for mass estimation, which serve nuclear material accountancy and criticality safety assurance. By propagating the holdup sources through a radiation transport model, it will be possible to produce pictorial dose rate maps that will serve to inform operational personnel and reduce dose.

For general operations, we envision a graphical user interface displaying the three-dimensional mass map overlayed onto a model of the glovebox and equipment. The user would pan, rotate, and zoom to navigate around the model. The two-dimensional coded-aperture images from each imager would also be displayed.

An ancillary page would display the two-dimensional Compton and coded-aperture images and the detected coded-aperture pattern for each imager. An energy histogram would also be displayed, with controls allowing the user to select the energy ranges for the image. The detected coded-aperture pattern will reveal any hot pixels or dead regions in the detector. This page would enable more detailed analysis and help the user understand the data that underlie the three-dimensional mass map.

For radiological dose minimization, the mass map overlay would be displayed next to a dose rate map. The dose rate map would be computed using radiation transport calculations based on the reported mass distribution and assumed equipment and shielding geometries. This will require that the software be updated or include options for variations in the position and orientation of equipment inside the glovebox.

A detector status page will display state-of-health information for each of the detectors. It would also plot trends for gross count rates and uptimes over periods of days to years. This information will support the maintenance of the system.

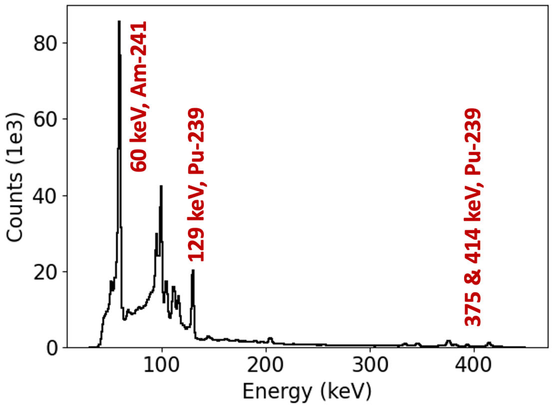

Figure 2: Measured energy spectrum from a plutonium triangle sealed source. The peaks at 60 keV from Am-241 and the series of x-ray peaks around 100 keV are the largest contributors to the spectrum, but do not provide a quantitative measure of the mass of the source.

Data collected by the system will be archived. Each imager produces a list-mode data stream that fills approximately 3 gigabytes per hour (one terabyte per week) at the maximum count rate, assuming 24-hour operation. On-board analysis tools produce images and logs summing to approximately 20 megabytes per one-hour measurement. To reduce the disk space required, we anticipate archiving the list-mode data during holdup measurements, which would be limited to one to two hours per day. This means there will be a maximum data storage requirement of 80 gigabytes per day of operation for twelve total imagers monitoring three gloveboxes. Mass maps can be expected to require four bytes per voxel. For cubic voxels with 2 cm length width and height, a three-dimensional map of the glovebox would contain approximately 2.4 million voxels, so the mass map data product can be expected to have a disk usage of around 10 megabytes. Consequently, archival of the list-mode data requires significant data-storage capability, and images, logs, and mass maps can be archived hourly without significantly affecting this requirement.

3 Mockup Measurements

A series of measurements has been undertaken with commercial-off-the-shelf imagers placed in a mockup of the glovebox geometry. The mockup was constructed from an extruded aluminum frame following the outer dimensions of a typical glovebox design on a 7-gauge stainless steel floor. A safety-glass plate with a thickness of 12.5 mm was mounted on the top of the mockup, and an imager was mounted above and parallel to the glass plate. A typical measured energy spectrum is shown in Figure 2.

For the measurements a set of sealed weapons-grade plutonium sources was used. Each source container is the shape of an equilateral triangular prism with a height of 11 mm and a base length of 57 mm constructed from stainless steel. Each source contains 1.8 grams of weapons-grade plutonium [1].

To test the position resolution of the system, four stacks of sources were arranged in a 2 by 2 array. A ten-minute measurement was taken with the four stacks in a close-packed array, and the separation between the stacks in both directions was increased in 2.5 cm steps. A ten-minute measurement was taken at each step to an inter-source distance of 20 cm. A photograph of the source configuration with a source-to-source distance of 17.5 cm is shown in Figure 3. The progression of radiation images is shown in Figure 4. The extent of the source distribution visibly grows at an inter-source distance of 12.5 cm and the sources are clearly resolved at 15 cm. A projection of the image onto the horizontal axis is shown in Figure 5 for the 13 pixel rows containing the sources. Fitting two Gaussian distributions with equal widths onto the final measurement yields a full-width half-max of 4.35 pixels, or 13.0 cm.

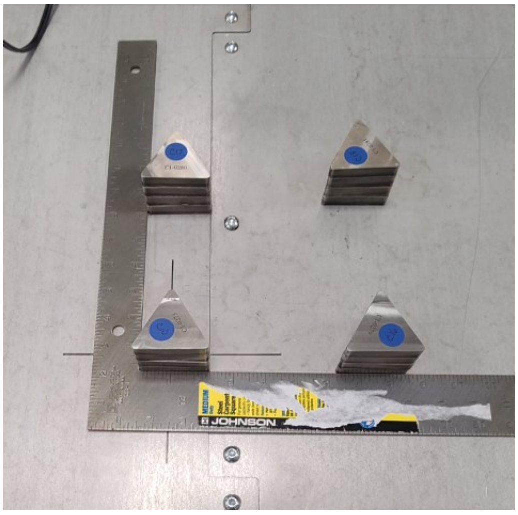

Figure 3: Photograph of the plutonium source configuration with a source-to-source distance of 17.5 cm.

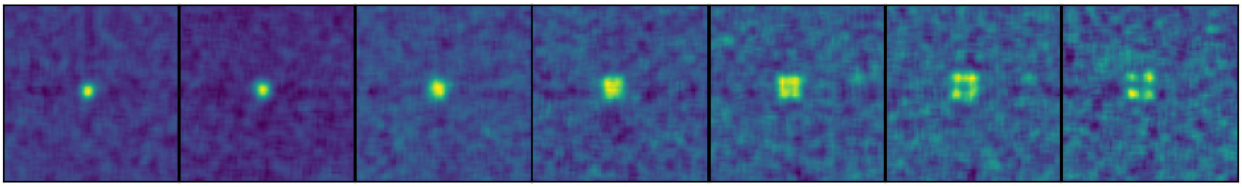

Figure 4: Coded aperture images for four sources with progressively widening spacing from 5 (left-most) to 20 cm (right-most) in 2.5 cm increments using the commercial-off-the-shelf imager. The sources were placed at a distance of 130 cm with a 1.3 cm glass plate in front of the imager.

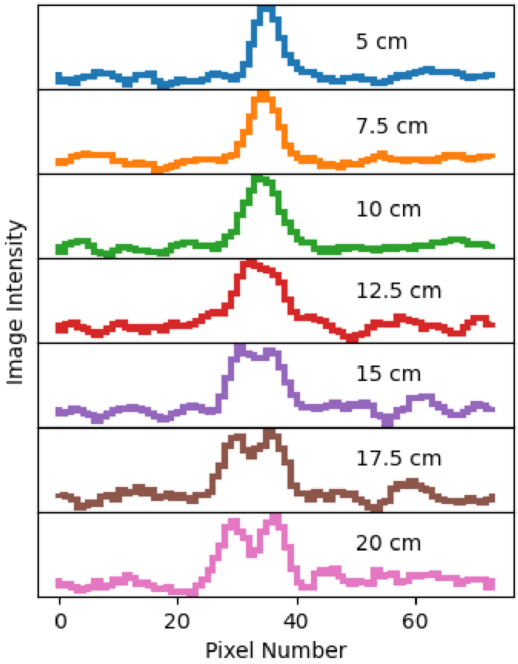

Figure 5: Projection of coded aperture images from Figure 4 onto the horizontal axis. The columns of sources are clearly resolved at a source-to-source distance of 15 cm.

4 Conclusions

A system of gamma-ray imagers designed for the purpose of measuring plutonium holdup in a glovebox is under investigation. By virtue of providing source localization, fixed detector geometry, and multiple views, this system is expected to improve on the accuracy of holdup determinations over that obtained with current techniques. Two other papers at this conference provide additional details for the development of quantitative coded-aperture imaging [6], and the likelihood maximization method for integrating data from multiple imagers [4].

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