

USE OF GAMMA RAY IMAGING INSTRUMENTATION IN SUPPORT OF TRU WASTE CHARACTERIZATION CHALLENGES

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ABSTRACT

Over the last ten years or so the field of gamma ray imaging has developed and become established within the nuclear industry. This has played a useful role in the radiological characterization of redundant nuclear facilities and specific plant items. These instruments, operated remotely with little or no operator dose uptake, are able to offer systematic monitoring of extended areas of plant, and are becoming an established technique in situations where dose rates are high or in which there are physical access limitations. Gamma ray imaging can unambiguously identify, visualize and record permanently the origins of radioactivity within contaminated environments. The standard output from these devices is a picture showing the location, distribution and intensity of gamma radiation, represented by color, superimposed on to a still video image of the scene under investigation.

More recently, the data that such instrumentation produces has begun to be used in more novel ways and in a complementary manner with other established measurement and characterization technologies. This combination of established technologies, when used with the quantitative output from gamma imaging instrumentation, has provided the opportunity to solve specific plant characterization challenges in new and different ways from that previously possible. A step change in the rate of progress of a variety of waste characterization challenges has been achieved. Two examples are described in this paper.

The first example has addressed the challenge of determining the plutonium inventory of a number of discrete items of waste stored in crates and drums. These items have been stored outside for over thirty years on the Sellafield site in north-west England, UK, and are scheduled to be transferred to an engineered waste store for interim safe storage. The plutonium mass within each item is required, with sufficient accuracy and confidence, to guarantee that subsequent handling and any processing is performed safely. A complementary combination of measurements and analysis techniques has been used to accomplish this challenge.

Secondly, a gamma imaging survey, followed by neutron coincidence counting measurements, were performed within a redundant cell in the reprocessing area of UKAEA's Dounreay site in northern Scotland, UK. These data were used in conjunction with computer modelling techniques, plant-provided engineering drawings and plant-provided plutonium isotopic data to determine the plutonium hold-up within the cell process structures. The work has provided sufficient confidence in the total plutonium mass present to enable further, practical decommissioning plans to commence safely.

INTRODUCTION

Gamma imaging devices were originally developed as tools capable of remotely surveying gamma radiation in radioactive environments. Such instrumentation has been widely used within the nuclear industry over the last ten years, with a majority of its use falling in the field of decommissioning. The standard output from these devices, referred to as an 'overlay image', is a picture showing the location, distribution and intensity of gamma radiation, represented by color, superimposed on to a still video

image of the scene under investigation. An example overlay image can be seen in Fig. 4. An overlay image provides an easy to interpret visualization of the contamination present within a given environment. Development of a strategy for decommissioning is facilitated by the identification of radiation hotspots within the environment, as these allow clean-up teams to target effectively these areas for early waste removal by manual or remote means, decontamination or shielding.

The RadScan[®]: 800 gamma imaging device, produced by BNFL Instruments, is one of several gamma imaging instruments available. This instrument has been extensively used within the nuclear industry in the United Kingdom.

DESCRIPTION OF THE RADSCAN 800 IMAGING DEVICE

RadScan 800 is a real time, directional count rate meter offering low resolution gamma ray spectroscopy (LRGS). Gamma radiation is detected by a sodium iodide scintillator connected to a photomultiplier tube. This assembly is located within a tungsten alloy collimator which attenuates radiation from all angles of incidence apart from the forwards. It has a user-selectable 2, 3 or 4 degree aperture through which radiation is detected. A photograph of the system is shown in Fig. 1. The inspection head also contains a laser rangefinder and a color CCD camera with zoom lens. The whole assembly is mounted on a pan and tilt unit and is operated from a remote workstation via a single cable which can be in excess of 100 metres long. This enables use of the instrument with near-zero operator dose uptake.



Fig. 1 A photograph of RadScan 800

The field of view (FOV) of the gamma detector can be scanned manually, alternatively the instrument can be set to monitor an area automatically. During an automatic scan the system collects data from a grid of measurement points, essentially from equal intervals in pan (horizontal) and tilt (vertical). The scan regime employed ensures uniform coverage of the area being scanned in order to minimize the scan duration and to improve accuracy for any subsequent quantitative analysis. At each measurement point

the system stores the pan and tilt co-ordinates of the measurement, the range to the nearest line-of-sight object, a low resolution gamma spectrum from the FOV and the video image in compressed 'jpeg' format.

This quantitative data stored by RadScan 800 allows the instrument to be of much more value than would be offered by production of overlay images alone. Two applications where these data have been used to address waste measurement challenges are described.

MEASUREMENTS SUPPORTING HRGS ASSAY OF HISTORICAL STORED TRU-WASTES

Several historical items, comprising eight medium-sized crates (approximately 1 m width by 1 m length by 2 m height), two drums and a filter, have been stored in an outside compound on the Sellafield site for over thirty years. It was necessary, for criticality safety purposes, to provide information on the plutonium contents of the items so that they can be moved to an engineered waste store for interim safe storage. As the exact radionuclide inventory and physical contents of the crates were unknown, all working on the crates had to proceed without disturbing them. All measurements therefore were taken non-destructively and in-situ, outside in the compound.

A multi-staged method was developed for determining the fissile content of these waste items.

1. RadScan 800 would be used to produce two dimensional images of the gamma emitting material located within the crates. By combining the data from multiple deployments of RadScan 800 around each crate, the location of the radioactive material within could be determined in three dimensions.
2. Collimated, passive high resolution gamma spectroscopy (HRGS) measurements were performed. These enabled the radionuclides present in the items to be determined and to establish whether ^{235}U or PuF_4 was present. In addition, this technology allowed the plutonium and ^{241}Am isotopic compositions to be determined at different locations.
3. Gamma transmission measurements were collected in the regions of the items identified as containing fissile material. These measurements provided information on the gamma attenuating properties of the items and their contents.
4. Monte-Carlo based computer modelling of each item, incorporating the results of the three measurements described above, allowed gram quantity estimates of the contents of each to be determined.

The method was first used on one of the smaller crates, shown in Fig. 2. This figure clearly shows the limited access around the crate in the compound, several of the other crates can be seen in the distance in the image.

The first phase of measurements involved scanning the crate using RadScan 800 from the east, west and south. Regions of interest were set up in the system to allow differentiation between ^{241}Am (25 to 70 keV) and plutonium (150 to 250 keV). A third region of interest covering the full energy range of the system was used to ensure that no other isotopes were present in measurable quantities. For each deployment, the location of RadScan 800 was measured relative to the crate using a laser measuring device or tape measure, and care was taken to ensure that the relationship between the recorded pan and tilt angles and the plant geometry was known.



Fig. 2 A crated waste item in the compound

Analysis of the data from the three RadScan 800 deployments yielded the three dimensional locations of the ^{241}Am and plutonium hotspots present in the crate. The distribution is shown pictorially in Fig. 3.

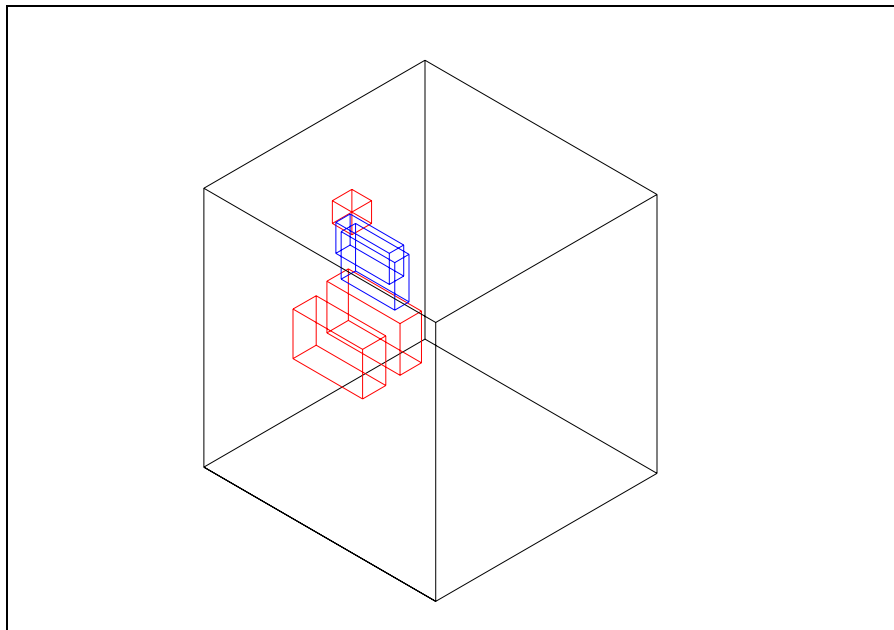


Fig. 3 View of crate showing ^{241}Am (red cuboids) and plutonium (blue cuboids) hotspots

A collimated HRGS detector was deployed to measure the passive gamma emissions from the crate. This was placed such that the entire crate was measured in a single acquisition. The positioning of the gamma detector was optimized by considering the three dimensional locations of the hotspots determined from

RadScan 800 so as to minimize the path length of the gamma rays from the object to the detector. In order to analyze the data acquired by the HRGS system to produce plutonium activities, the efficiency of the measurement is required. The efficiency of the measurement is dependent upon the location of the sources of radiation, and the amount and type of shielding present within the crate.

The amount and type of shielding present within the crate was assessed by taking gamma transmission measurements. This campaign of measurements was focussed only around the hotspot locations identified by RadScan 800. Transmission measurements were performed by deploying a ^{152}Eu source and the collimated HRGS detector on opposite sides of the crate. The transmission of ten key gamma lines from this source, spanning an energy range of 121 to 1408 keV, were measured. The measured transmission of these ten gamma lines was compared against the theoretical transmission through a matrix containing varying amounts of wood, steel and PVC. An iterative process was used to vary the relative and absolute quantities of each of these three materials until a best match between measured and modelled transmission was achieved across the full energy range. This best fit characterizes the amount of material present in the matrix; the known cross-sections of these materials were used to provide the gamma attenuation at any energy of interest.

The gram quantities of plutonium present within the crate were based upon analysis of the 413.7 keV line measured by the passive HRGS measurements. A number of geometrical models were constructed with varying assumptions about the distribution of plutonium and shielding within the crate. A detailed detector response supplied by the manufacturer was used within these models. The results demonstrated clearly that the determined mass of plutonium was dependent upon the assumed distributions of source and attenuator, with results varying from a few hundred grams up to a few kilograms of total plutonium. This variation in fissile content spans criticality safety limits for both the movement of the items to, and storage within, the engineered waste store. The penalty of not being able to demonstrate that the fissile content of an item is below 450 grams is the requirement for significantly increased effort in justifying the criticality safety associated with the movement and storage of each package. An alternative, but costly, option would be for the item to be dismantled, repackaged into smaller packages and re-measured to prove that each has less than 450 grams fissile content prior to consignment to the waste store. The combination of the gamma imaging and gamma transmission measurements was able to constrain the assumptions that had to be made in the modelling aspects of this work. In addition, the ^{239}Pu activity was assessed using a number of different ^{239}Pu gamma peaks to further confirm that the selected model had the appropriate levels of attenuation, and to examine the potential for bias due to the self-attenuation of the gamma signal within the plutonium.

A justified plutonium mass of 274 ± 51 grams at one sigma total measurement uncertainty was determined for this crate. As no gamma signal from any uranium gamma lines were detected during the HRGS measurements, it was deemed to contain only plutonium. A recommended upper bounding plutonium mass value of 427 grams (i.e. including three sigma uncertainty) was assigned to this crate, and this is the total fissile content value used in support of the safety case for removing the first item from the compound to the engineered waste store.

The strategy implemented for measurement and analysis of the items has been peer reviewed and accepted by BNFL Safety and Environmental Risk Management's criticality assessment experts as a rigorous and justified means of determining the fissile content. For those items with fissile content of less than 450 grams, simple over-packing and transfer to the engineered waste store will occur. For any with fissile content above this value the results of the individual measurement techniques will be used to support criticality safety during over-packing and transfer to the above store. This may take into account the distribution of the fissile mass and the presence of any thermal neutron poisons, for example plutonium in the form of ^{240}Pu . If this is not achievable, the item will have to be split into further smaller pieces such that criticality safety is always assured.

MEASUREMENTS IN SUPPORT OF CELL CHARACTERIZATION AT DOUNREAY

A radiometric investigation was performed in an evaporator cell at the Dounreay site in order to determine the mass of plutonium contained within the pipes and vessels therein. A sudden shut-down of this plant left plutonium bearing liquors within pipes and vessels throughout the process. In order to achieve post-operational clean out (POCO) these liquors must be removed. The preferred method is to run the remaining liquors through the plant, as per its design intent, thus emptying all of the vessels by running their liquors through the in-cell pipework. In the period between shutdown and POCO, settling of the contents of some of the pipes within a particular cell occurred, resulting in them becoming blocked. These pipes need to be replaced before POCO can take place. In order to effect a replacement of the pipework within this particular cell its plutonium inventory was required for waste handling and safeguards purposes.

A three-staged method was developed for determining the fissile content of the cell.

1. Stage one comprised a RadScan 800 survey. This was used to produce overlay images showing the location and relative abundances of the plutonium held up in the vessels and pipes.
2. The second stage consisted of a series of neutron coincidence counting (NCC) measurements. The positioning of the neutron detectors for these measurements was identified from the results of the RadScan 800 survey.
3. Thirdly, the NCC data were analyzed to determine the quantity of plutonium present using a combination of laboratory calibration measurements, known plutonium isotopic compositions, RadScan 800 results and Monte-Carlo modelling.

RadScan 800 was deployed within the shielded cell and operated remotely. From this single deployment it was possible to scan the entire cell without the need for redeployment or repeated man-entry. This scan was used to identify all of the pipes and vessels within the cell that still contained plutonium. Regions of interest were set up in the gamma spectrum to enable images from ^{241}Am (25 to 70 keV) and plutonium (150 to 250 keV) to be generated. Fig. 4 shows a portion of the gamma imaging that was produced from this survey. The left image shows the plutonium distribution, and the right image the americium distribution, for the same area of plant.

The spectroscopic capability of the system, in providing a plot for both ^{241}Am and plutonium, shows that the two isotopes do not follow exactly the same spatial distribution, as might have been expected. The areas of enhanced americium activity may be an indication of the presence of older plutonium as americium is a decay product of ^{241}Pu and “grows in” to the plutonium over time. Fig. 5 shows a low resolution gamma spectrum collected during the scan from a plutonium hotspot. The most intense peak in this spectrum is the 60 keV peak associated with ^{241}Am , but also present are peaks centred at 100 and 210 keV, these are characteristic of plutonium.

At the relatively low gamma ray energies being measured in this deployment the RadScan 800 provides a very high signal to noise ratio (SNR). The SNR can be defined as the ratio of counts that the system is looking at in a given FOV as compared to the counts detected that have penetrated the system's collimator. This very high SNR means that the gamma ray images readily show the areas of plant that are free from significant residual plutonium and therefore do not necessitate further investigation. It also improves quantitative analysis when considering the relative intensities of the numerous hotspots of plutonium identified within the images.

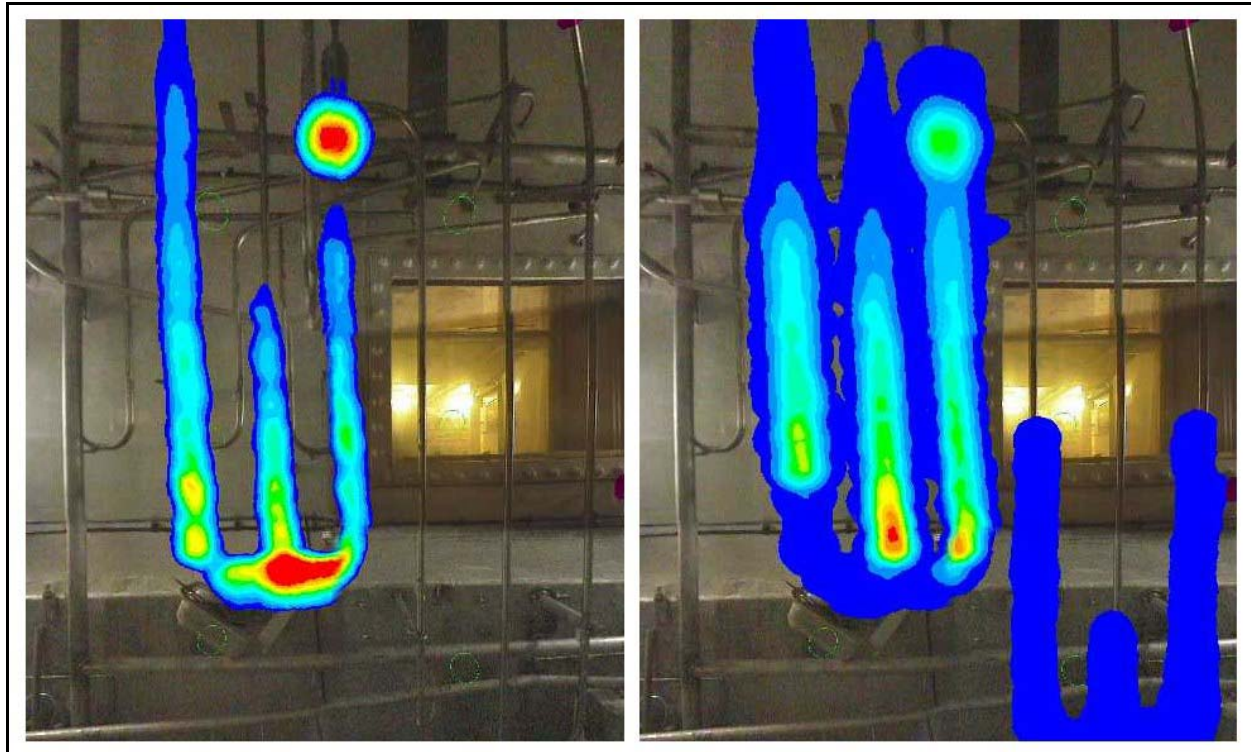


Fig. 4 An overlay image for plutonium (left) and ^{241}Am (right) for a portion of the cell

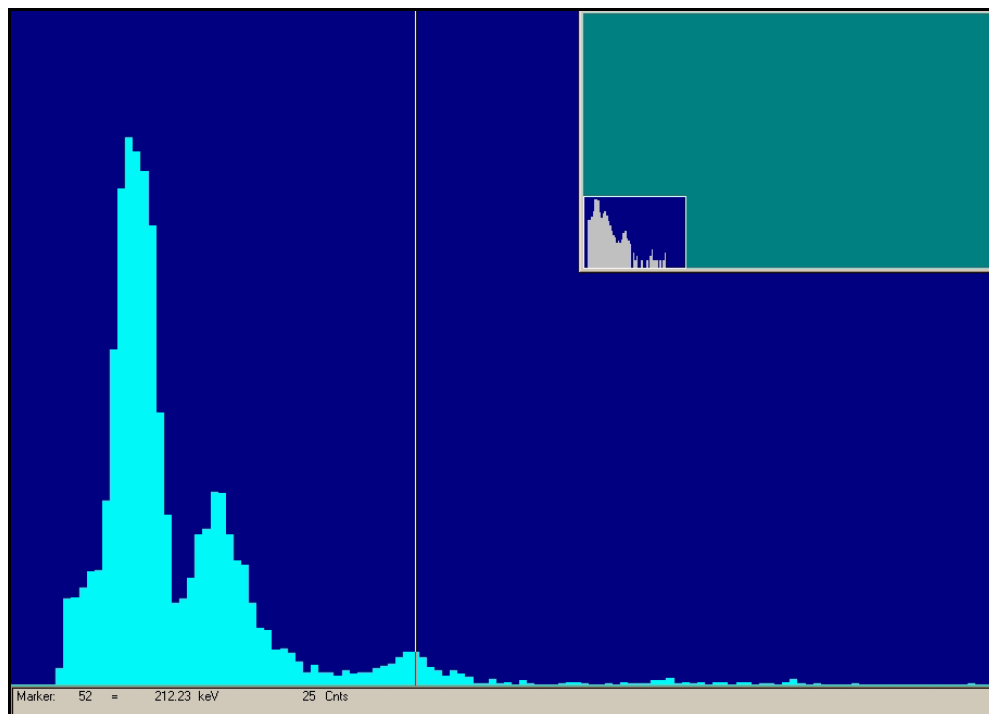


Fig. 5 A low resolution gamma spectrum from one of the hotspots in the cell

The RadScan 800 survey results were used to locate the items of interest and the concentrations of plutonium activity within the cell. The cell structures to be monitored were decided upon, as were the locations for deployment of the NCC equipment. Polythene moderated ^3He neutron detectors were deployed, surrounding each of the selected vessels in turn. Typically six detector modules were used, with each containing two 0.5 m long ^3He neutron detectors inside a polythene moderator case. Suitable counting electronics measured the neutron coincidence count rate from the deployed modules. Counting times were typically 4 to 16 hours. These long counting times were used for convenience. Shorter counting times may have produced acceptable results for some of the measurements, however, cell access was restricted to two suited entries per day and so maximum use of the available time was made.

MCNP models were created, using engineering drawings of the cell for the geometrical input, to simulate the NCC measurements. The models generated the neutron detection efficiencies for each of the experimental arrangements. The efficiencies were ascertained in terms of the neutrons detected per neutron emitted from the plutonium. These were calculated for the distributed activities determined from the RadScan 800 survey results. The absolute detection efficiencies of the series of NCC measurements varied between 2.6 and 7.6 %.

It is essential to know the plutonium isotopic composition present within a cell structure in order to interpret the neutron coincidence counting measurements to determine its plutonium content. The relative abundances of the plutonium isotopics within the plant vessels were known from the plant's historical records and knowledge of the last feed material to go through the plant. Had this not have been available, this information could have been measured using in-situ HRGS.

Five deployments of the NCC equipment were made focusing on five areas of the cell. The quantity of plutonium present in each of the areas was determined. There were 103.0 ± 0.4 grams of plutonium associated with the "w" shaped pipework visible to the left of the lead glass window in Fig. 4, and 14.3 ± 0.1 grams associated with the product hold-up vessel as can be seen by the hot spot above the right hand arm of the "w" shaped pipework. The errors quoted are at one sigma confidence and reflect only statistical counting uncertainties. Sources of systematic uncertainty were considered, taking into account variations in the distribution of the plutonium, and the effects of background neutron fields, and were estimated to be about 40 % at one sigma.

The results generated have been used as the declared waste inventory for waste handling purposes, and have provided the fissile masses for declaration to Euratom. Previous attempts to measure the fissile content of the cell, using total neutron counting (TNC) techniques alone, resulted in a much increased total measurement uncertainty. The improved accuracy afforded by incorporating the results of a gamma ray imaging survey have given the decommissioning team much greater confidence in the assay results, and the ability to better plan and demonstrate the safety of all operations within this cell.

CONCLUSIONS

This paper has showcased two applications where gamma ray imaging has supported waste characterization challenges. The ability to support the measurements described can only fully be achieved by the provision of high quality, quantitative data from the gamma imaging device. In particular the follow characteristics have made gamma ray imaging an invaluable technology for assisting with waste characterization challenges:

- Spectroscopy has proven to be essential. In the first instance spectroscopy provides a simple tool to help in the initial interpretation and characterization of an environment. The capability to analyze specific portions of the gamma ray spectrum relating to individual radionuclides has enabled more quantitative analysis to take place.

- Positional information, pan and tilt angle and range, has been used to support other gamma and neutron measurements by determining the three dimensional positioning of the radioisotopes. This information has reduced significantly the total measurement uncertainty associated with the analysis of other measurement techniques by constraining some of the assumptions that would otherwise have had to be made.
- By being able to remotely assess and image a complete environment it is possible to focus subsequent measurements to only those plant items or areas containing significant amounts of activity. This reduces the dose uptakes and costs associated with detailed characterization.
- A very high SNR gives a high tolerance against the effects of off-axis radiation, this is the ability to detect one source in the presence of other sources. This is invaluable when interpreting the data produced as the presence of false artifacts in an image is misleading. The very high SNR in these instances has proven to be essential for quantitative analysis.

Finally, in addition to these specific benefits, the distribution of activity visualized by the overlay image is in itself a useful aid in planning and implementing practical tasks.