The Use of X-RADIOGRAPHY Information with GAMMA SPECTROSCOPIC Analysis of WASTE DRUMS – 24478

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ABSTRACT

Gamma Spectroscopy of waste containers traditionally relies on assumptions or measurement (using radionuclide sources), of the internal contents and heterogeneity both in terms of the waste matrix and radionuclides. Mirion and VJT have previously investigated the feasibility of using alternative means to determine the internal waste matrix properties by combining X-Ray CT with gamma spectroscopy and ISOCSTM modelling. This can substantially improve assay accuracy, without relying upon *a priori* knowledge or external radionuclide sources.

Building on the results and conclusions previously presented using a carefully constructed 5-gallon calibration test pail loaded with controlled heterogeneous materials, we now extend that work to 55-gallon waste drums which represent a wider application field and potentially greater benefits. X-Ray CT scans were conducted with carefully selected interrogation energy, producing a voxel map of the heterogeneous material density as well as recognition of specific object locations and shapes within the drum. This data was coupled with SuperISOCS modelling of the contents. In this paper we illustrate the accuracy improvement that can be achieved by modelling the drum contents with X-Ray – inferred knowledge of specific internal objects, for a simple far-field ISOCSTM drum assay, compared with a traditional approach of assuming the entire contents (matrix and activity) are uniform. Lastly, noting that many plant operators already have an installed RTR system, which are easily adaptable to provide CT data, and a gamma scanner operated independently, we highlight how a simple software upgrade can allow operators to benefit from these advanced technologies, including outlining the principles of the density voxel concept for TGS.

INTRODUCTION

Accurate radiological characterization of sealed containers by Non-Destructive Assay (NDA) methods is dependent on the nature and distribution of both the radiological materials and the physical matrix contents. This coupled with varied waste streams and project dependent methods of loading waste into the containers, often leads to high variability and unknowns, which in turn can lead to large uncertainties in the NDA results. Several methods exist for reducing these uncertainties. Here we explore the possibility and benefits of X-Ray Computed Tomography (X-Ray CT) data as one input to improving the quality of the result.

Waste characterization based on gamma spectrometry can be performed with uncollimated detectors in a simple far-field geometry to more complex and accurate systems such as a Tomographic Gamma Scanner (TGS) that uses a collimated field-of-view as well as rotation and 3D-scanning of the drum to improve accuracy and reduce uncertainty. The accuracy of the activity determination of each radionuclide depends on accurate knowledge of (i) the waste matrix composition and (ii) radionuclide spatial distribution within each container. For the radionuclide activity distribution (with usually generates the largest uncertainty component in the Total Measurement Uncertainty), the simplest assumption is of a uniformly distributed activity throughout the sample. More sophisticated detection techniques provide additional information and can improve the accuracy. For the radioactive emissions, such techniques can use collimated scanning approaches such as with the Segmented Gamma Scanner (SGS) which segments a drum into vertical disks, or an Advanced Sectorial Gamma Scanner (ASGS) [1] which scans in vertical disks and wedge shaped 'pie' slices. The TGS [2] takes this a step further and scans the drum laterally and vertically enabling the matrix and radionuclide distribution to both be visualized and quantified as a 3-dimensional voxel map. Similarly, several techniques exist for the determination of the waste matrix composition.

A-priori data either from Acceptable Knowledge (AK) or visual inspection are the most straightforward sources of information about the drum contents, but the former can be questionable if the provenance is not reliable, and the latter can be outright prohibitive. A simple weighing is a means to calculating an average density which can then be assumed to apply to the entire volume of the container. SGS and TGS systems use high-activity radionuclide transmission sources to measure gamma attenuation properties of the contents and provide a more detailed and reliable estimation of the impact of the contents on the measured activity. However, such techniques rely on adequate penetration through the matrix which can be limited by source activity/content density and low energy gamma emissions, and management of the transmission source has inherent burdens. Since X-Ray Real Time Radiography (RTR) systems are routinely used for visual examination of drums in a separate stage prior to the radiological measurement, the concept of using that information instead is the focus of this paper.

A significant aspect of the activity and uncertainty determination in the methods described above is the choice of calibration. Source-less calibration is now an established technique that avoids the need for radioisotope calibration sources which can be prohibitive to produce and manage on the grounds of cost, safety and technical concerns over their representativeness of real waste material. However, this also depends on accurate knowledge of the waste container matrix make-up and radionuclide distribution. Mathematical modelling supports gamma spectrometry using source-less calibration tools such as ISOCSTM. Advanced services are now available, for example ISOCSTM variants as reported previously encapsulated in the AIGS (Advanced-ISOCS Gamma Spectrometry) service.

While these reduce measurement uncertainties, they still require knowledge of the contents and are not a substitute for robust a-priori data either from Acceptable Knowledge (AK) or drum inspection by X-Ray or other means.

The range of techniques and tools available require careful consideration of the goals of the measurement, and a balance between the cost and complexity of the method applied and the cost savings realized through more accurate activity determination (and/or reduced uncertainty determination) and reductions in measurement time. Given the previously mentioned routine use of RTR the work in this paper aims to quantify the benefit of using this information in the calibration and activity determination for drums.

TECHNOLOGY DEVELOPMENT ROADMAP

In addition to the above core radiometric technologies and systems, real-time radiography is widely used for performing quality checks on the contents of sealed waste containers. Mirion and VJT technologies supply suites of systems that, together, operate as complete characterization suites sometimes in conjunction with neutron counting systems fissile material assessments. Typically, the RTR systems are used to check for the presence of any prohibited items inside a drum that may adversely impact onward processing (such as super compaction). RTR images are, additionally, sometimes used to inform gamma spectrometry NDA analysis by allowing an assessment of the fill-height of the waste inside the container. This can avoid important undesirable measurement bias because without knowledge of the fill-height of the waste, it is common practice to simply conceptually homogenize the entire contents, for the purpose of calibration, through the entire volume, which can lead to incorrect modelled bulk density and hence matrix attenuation.

Recent advances in ISOCSTM modelling technologies [3, 4, 5] have made it possible to perform accurate modelling of more complex item geometries than are allowed by the normal "object template" approach of the standard commercially available ISOCSTM, ISOTOPIC and similar software packages. SuperISOCS [5] allows modelling of complex inter-connected objects, repeated structures and voxel lattices, while still using the industry standard ISOCSTM calculation techniques that are widely benchmarked.

Combining information from RTR with the availability of a sophisticated modeling tool forms the basis of this technology development. Mirion and VJT have undertaken a technology development that couples X-ray CT scanning with gamma spectroscopy. The goal is for robust AK, based on visual X-CT data, to be converted to recognition and dimensioning of individual shapes, and for quantified grey-scale data to be converted to material density in each object.

Although Real-Time X-Radiography is often used in qualitative mode, the expertise available, ability to tune the X-ray energy to allow penetration into high densities of waste in various containers, coupled with existing computed tomography (CT) algorithms potentially allows existing X-RTR systems to be softwareenhanced to provide mapping data for the contents of a waste container, as well as clear visible "grey-scale" data allowing object recognition taking advantage of the few-mm spatial resolution that this technology provides.

We have previously demonstrated the benefits of this principle in terms of real tangible improved nuclide activity measurement accuracy for small cylindrical surrogate waste containers, by coupling SuperISOCS analysis based on modelling individual items inferred by inspection of the X-ray scan images [6].

Our team is now extending this work to larger containers (200-liter drums) which is the subject of this paper with results presented in the ensuing sections. Future developments include investigating the possibility of generating a voxelized map that can be automatically converted to a model, and thereby extending the coupling of the X-Ray CT scanning data (voxel density maps) with the standard TGS voxel map analysis software. As we develop this technique, we highlight the accuracy improvements that this makes possible, for 200-liter drums and the operational benefits that are made possible by using the X-Ray CT data instead of a transmission source. This paper describes our analysis and roadmap for development that will allow waste management operators to fully benefit from bringing together these advanced technologies.

RESULTS AND DISCUSSION

Below we review the results of our proof of principle for small (5-gallon) pails, and then we progress with the extension to 200-liter drum measurements.

Proof of principle with a 5-gallon test pail

Initially a 5-gallon pail was loaded with items of known weights, dimensions and composition, and with re-entrant tubes allowing insertion, later, of test sources to permit additional measurements. Figure 1 depicts the can in two stages of loading, so that the different objects are visible. Notably there are two bottles filled with steel shot. Materials and objects were chosen to represent a realistic assembly of mixed waste items encountered by a waste operator. This served as the basis of our scenario modelling, described below. An X-Ray scanner was used at VJT's premises that allows scanning with an appropriate energy commensurate with the chosen container dimensions and weights.



FIGURE 1. 5-gallon test pail.

Our approach is based on the use of SuperISOCS modelling to conduct "what if" scenario analysis, including studying the impact of specified assumed radionuclide distributions inside each test container. We are able to study the impact, within a given physical waste matrix "map" as obtained by the X-Ray scan, of making different assumptions for the nuclide activity distribution based on recognition of the contents themselves. In the SuperISOCS model for the 5-gallon pail we assumed that all of the activity was concentrated within the iron-shot filled bottles. For contrast, we then made a second model to simulate what results would be achieved without any knowledge of the individual items within the pail, but only the overall weight and material composition which is then homogenized throughout the entire pail volume. In addition, for the reference case the activity was assumed to be uniformly distributed throughout the entire pail volume.

Figure 2 compares the reference assumption-based results with the more accurate results from our new X-Ray CT + SuperISOCS procedure. As can be seen, the standard assumption can result in significant underreporting of the true activity, and this is due to self-shielding in the iron-shot-filled bottles as well as their locations. The degree of this under-reporting varies from approximately 25 to 40 % depending on the gamma energy and is indicative of the accuracy improvement achievable by the combination of the SuperISOCS + X-CT.



FIGURE 2: The % under-reporting as a function of energy when using the traditional approach (smearing the entire contents over the pail volume). Both techniques assume continuous rotation of the pail.

Extrapolating benefits to 200-liter drums (and larger containers)

Real-world benefits from such accuracy improvements can be understood if one considers a waste sorting project (for example between the broad categories of a) TRU (TRansUranic waste) / PCM (Plutonium Contaminated Material) / ILW (Intermediary Level Waste) and b) LLW (Low Level Waste). The waste disposition cost differential is large, if a large proportion of the containers has an activity that is uncertain to a level of a few tens of percent so that the true activity could be either side of the TRU / LLW boundary. Since the typical approach would apply a conservative over-reporting of activity in the majority of cases, the cost savings are potentially enormous if one could remove that bias as this new technique does.

If one extrapolates to larger containers (such as 200-liter drums) that are more practical for every-day waste management practices, then the accuracy and cost savings is exponentially increased. For large containers the bias % ranges are closer to the "100%" or "factor of 2" level and thus open up potential savings for a much larger proportion of a waste population than if the waste was treated in small containers.

It is also necessary to consider the impacts of different types of materials. The data in Figure 3 illustrates the dependencies of these accuracy improvements on the type of matrix material with reference to metallic or soft waste, albeit for a rather extreme scenario of measuring Pu through the characteristic 60 keV gamma emission from Am-241 which is subject to large measurement uncertainties. The MDA (Minimum Detectable Activity) is inversely proportional to the detection efficiency modelled, so gives a fair indication of the potential measurement bias if the new advanced techniques are NOT used. For example, a ratio of 10 on the vertical axis denotes a potential under-reporting of the Pu activity by a factor of 10. As can be seen, the benefits of being able to avoid (through judicious deployment of advanced technologies such as those presented in this paper) the requirement to use traditional "worst case" assumptions regarding spatial location of radioactivity, are large.



FIGURE 3: Illustration of variability of detector efficiency with matrix density, material type and source position, for a 200-liter drum loaded with homogeneous waste. The "worst case" corresponds to all the activity being at single point source at the location with lowest possible efficiency (bottom center).

Now the exponential nature of gamma ray attenuation means that uncertainties increase rapidly and exponentially with container dimensions. The benefits for large containers such as ISO shipping containers, are therefore obvious. As a rule of thumb, the "point of inflection" is typically quoted as being 1 m³, and containers / bags of this volume are therefore typically used for bulk soil / concrete measurements.

The X-Ray CT scanning technique can be applied to large containers including up to several cubic meters with careful selection of X-ray generator target voltage. This spectrometry modelling analysis technique can be used to predict the accuracy improvement that can be expected for containers such as B25, Standard Waste Boxes (SWB), and thus used as a project planning tool to allow waste managers to plan their equipment requirements, assess what is a realistic accuracy to expect, and thus better forecast waste storage and shipment requirements and costs.

Demonstration with a 200-liter test drum

A test drum was identified having contents typical of modern decommissioning operations, with several every-day objects including inactive tools and pieces of plant pipework loaded at random inside the drum. Additionally, a set of density calibration rods were mounted around the outside of the drum, allowing the grey-scale X-Ray images to be interpreted to create density maps of the drum contents. The images were analyzed using standard X-Ray viewer software. In Figures 4 and 5, we show scan results for a 200-liter drum, designed specifically to demonstrate capabilities for typical decommissioning waste.



FIGURE 4: RTR scan: On left, an example 2D view of the 200-liter test drum: On right, stitched image showing complete drum contents and 3D view.



FIGURE 5: RTR scan: different views of 200-liter test drum: Zooms showing details of individual items, tools, canisters and cabling.

The X-Ray CT scan of the bottom part of the drum--containing the most dense items and so the focus of the study--has been analyzed and a 3D SuperISOCS model containing the most significant items has been performed.

Figure 6 shows 3D views of the following main items for which a model has been created, namely:

- a lead brick assembly (in a stepped pattern),
- a motor and a pipe,
- a drill (battery pack visible),
- the bottom part of a steel shot bottle.
- a valve (comprising a section typical of a plumbers tap)

For the objects above, the images were inspected to determine the locations and dimensions, and where appropriate item container wall thicknesses (the valve). The same process used in the analysis of the 5-gallon pail was followed with the following key analysis steps to develop representative SuperISOCS models.

- Assessment of key materials
- Assessment of object weight based on practical considerations,
- X-CT scan-inferred wall thickness estimates where possible and assumed knowledge for example of internal motor assembly components (for the motor and the drill)
- Assessment of likely contamination profile for each item based on physical nature of each object.

The assessment from the X-ray images provides additional useful information since for example the lead blocks in a real-world scenario would only be contaminated on the external surfaces, whereas the motors are more likely to comprise internal contamination if the motor and drill had operated inside a plant decommissioning area comprising of airborne or disturbed (from drilling operations) contaminated materials, leading to intake through the motor vents. This is information that can feed into the activity distribution in the SuperISOCS model. This highlights the importance of careful assessment of the operational history and knowledge of the properties of the items loaded into a waste drum, in order for robust assessments to be made as to the measurement accuracies and errors that may be encountered.



FIGURE 6: 3D scan view of the main items in the 200-liter drum. From top left and rotating clockwise the following objects are depicted: (a) Lead block assembly (b) Motor and pipe (c) Drill (battery pack visible at bottom right) (d) steel shot filled bottle (e) Valve / tap.

SuperISOCS models were then created to produce as close a match to the true contents as possible as depicted in Figure 7. After making practical assessments of the mass of each item, the remaining known mass of the drum contents was smeared homogeneously over the contents of the entire drum.

After making the above considerations, we took the assumption that the items would be contaminated equally in terms of Bq/g, as might be a typical case as a result of pre-screening of items based on handheld dose probe measurements prior to loading into the drums. This gives a realistic potential basis for distributing the activity across these 6 items inside the drum. In practical scenarios, a similar analysis would need to be undertaken, dependent on the physical nature of the individual items and their "operational history" as discussed above.

Figure 7 compares side-by-side cross sections of the X-ray CT scans and of the SuperISOCS model for the 200-liter test drum.



FIGURE 7: Cross sections of the 200-liter test drum from the SuperISOCS model and the X-ray CT scan of the bottom part, respectively horizontal (lower views) and vertical (upper views).

Using this model and assumptions on the source positions, several models were built and compared to the reference model of a standard homogeneous approximation. Similar to the 5-gallon pail studies, results for each case were averaged from four different detector positions around the drum as an approximation to the drum being rotated continuously. Figure 8 shows the results for the different cases.

CASE01-HOM is the model with a homogeneous distribution of the matrix and source and serves as the reference for the other cases. The other cases represent various assumptions of activity distribution. CASE 03_ALL assumes each item (as well as the matrix) has the same mass activity (Bq/g, based on the mass of the item); note that this can be very different from assuming equal relative concentrations for each item. CASE06-MATRIX assumes only the matrix has activity while the other defined objects are solely attenuating materials. CASE07-LEAD assumes the activity is only in the lead block and only on external surfaces. CASE08-MOTOR assumes the activity is only in the motor and here the contamination is distributed in air internal to the motor, on the motor components, and on external surfaces as well. CASE09-PIPE is similar with the activity inside the pipe. CASE12-SHOT is the steel shot where the activity is distributed homogeneously throughout the steel.



FIGURE 8: Accuracy improvement analysis of SuperISOCS + X-CT deployed for the 200-liter test drum. The % under-reporting or over-reporting is shown, as a function of energy, had a traditional approach been used instead (smearing the entire contents over the pail volume). Both techniques assume continuous rotation of the drum.

Figure 8 shows that, depending on the gamma energy of the nuclide of interest, incorporating the object information available from the X-Ray CT scan can improve under-reporting of approx. 50-100%. For the lead brick and the steel shot the under-reporting bias is 40% or higher at all energies (and therefore all nuclides). While these results were from models treating each item as the only source of activity, it is indicative of the bias that could be unnecessarily assigned across the board to a Total Measurement Uncertainty (TMU) budget when no internal information from the drum is considered. Although additional cases and a further assessment of the complete drum contents will add to the analysis of the impact of using X-Ray CT images for improving drum results, the primary objective is demonstrated in these results.

FUTURE WORK

In this section we explore how the X-Ray CT and ISOCSTM/SuperISOCS tools can be coupled as technology building blocks, allowing advanced flexible analysis. As the next step this would mean automating much (if not all) of the manual steps used in the current work and ultimately X-Ray CT scanning could potentially be coupled with a system as complex as TGS and integrated to allow seamless characterization and using existing hardware suites and software upgrades wherever possible.

The basic principles of the X-Ray CT scanning technique are illustrated in Figures 9 and 10. In Figure 11 we show an example of a SuperISOCS model, of a set of sample bottles that was placed inside a 200-liter test drum used for laboratory intercomparison purposes to illustrate the equivalency of the techniques.



FIGURE 9: RTR CT scan: Basic scanning principles.



FIGURE 10: RTR CT scan: Image Reconstruction – Basics.

With some assumptions, the X-Ray CT reconstruction results could provide a density mapping, for a reference set of materials, robust enough to reproduce the equivalent attenuation of the scanned voxels. The use of voxels lends naturally to integration with the TGS algorithm where the density map is used in the TGS activity reconstruction algorithm in a manner directly analogous to those provided by the transmission source steps of the TGS process. As discussed previously this could prove an interesting alternative to the standard transmission source TGS method.



FIGURE 11: Example SuperISOCS model of a 200-liter drum filled with sample bottles, using repeated structures, equivalent concept and transferrable to a simple voxel map.

Simulating the 5-gallon pail density voxel map

As a proof of principle, a map of the averaged partial densities over each voxel has been computed for the set of materials used in the original SuperISOCS model of the 5-gallon pail as shown in Figure 12 (see also figure 1). This can then be used to create a voxelized SuperISOCS model of the entire pail contents. As an example, Figure 12 shows a simplified version of the manually created detailed model of the pail, but one that could easily be automatically generated from X-Ray CT results and that can be readily used in an automated process with a gamma passive system, either with a simple turntable and straightforward full drum measurement or, for even better accuracy, in combination with a TGS system mechanics and algorithm but without the need for a transmission source. This method uses the density calibration rods that are built into the test drum for the current proof of principle tests. Standard image analysis techniques, as already deployed in routine image analysis, have been used to interpret the X-Ray image grey-scale density to correlated with voxel densities.



FIGURE 12: Proof of principle: creation of a simplified 3D density map using the 5-gallon pail test data

As a demonstration of the improvement in the case of a standard gamma analysis, this voxelized pail model can then be used to generate with SuperISOCS an efficiency curve directly analogous to the one obtained using the detailed SuperISOCS model presented previously. Thus, one can get the most out of the accuracy improvements from using the X-Ray RTR results without the need for an expert to accurately model the drum contents.

CONCLUSIONS

Our results have shown that X-Ray CT scans, coupled with SuperISOCS analysis of standard portable High Resolution Gamma Spectrometry data for a typical 200-liter test drum, can yield substantial benefits in terms of accuracy improvement. For cases where activity was localized in a single object, accurately modelling the items in the drum resulted in a 40% bias reduction for typical fission/activation products that are most commonly relevant for a decommissioning scenario. For low energies typical of nuclear material assay, a 50-100% bias reduction in the activity assessment is seen. This latter case is often assumed as the 'worst case' in a TMU assessment and folded into the assigned uncertainty regardless of the contents in the drum. Our approach would produce a more realistic "worst-case" scenario of activity distribution than an implausible case that has vanishingly low probability of occurrence in a real waste drum.

Our initial work has been focused on use of the X-Ray scanners in image recognition mode, inspecting the image and allowing the dimensions and locations of specific items (e.g., bottles, pieces of scrap wood or metal) within the container to be determined. Later work will allow definition of a CT scan grey-scale voxel grid to be overlaid on a standardized SuperISOCS voxel lattice grid with assumed uniform matrix material composition and density within each voxel, and this allows a far more generalized approach. Both the measurement modalities and the modeling capabilities exist today, and the vision of a complete turn-key characterization suite is within reach.

For measurements with portable ISOCSTM -based gamma spectrometry equipment, the X-Ray CT data can be used both in an object-recognition manner, to allow SuperISOCS modelling of discrete items, or in a voxel density map manner, based on simple software scripting, again allowing automation and high throughput in a real-world facility environment. Similarly, our approach for the TGS software integration is entirely within the scope of a mathematical analysis of the standard existing X-Ray CT and TGS software algorithms, with the goal to allow X-Ray CT voxel density data to be overlaid onto the required voxel grid element lattice shape and grid spacing.

Mirion is developing these advanced accuracy-improvement techniques to exploit the powerful features of ISOCSTM including new innovations that allow more complete and detailed models to be prepared. This is being done to provide a tool-kit strategy that allows these advanced procedures to be made available for a wide range of applications and measurement geometries including not only simple far-field geometries, but also SGS and TGS solutions as alternatives to using transmission sources. Furthermore, as such solutions are ever-more required for the emerging global challenges including robotic deployment of equipment and "sort and segregate" applications (for example sorting tables, conveyors, or loose waste in in accessible areas), these techniques can also be envisioned with other imaging technologies.

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