#### Assay of PCM Crates and Drums using the BIL Solutions Ltd. Imaging DISPIM<sup>®</sup> System

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### ABSTRACT

BIL Solutions Ltd Decommissioning In-Situ Plutonium Inventory Monitor (DISPIM<sup>®</sup>) is a versatile neutron and gamma assay system designed to be deployable in a wide range of decommissioning and waste management situations. The system utilizes Totals and Coincidence neutron counting to determine both total Pu mass values and the 3D Pu mass distribution inside the target object. DISPIM<sup>®</sup>'s modular, mobile design allows the system to be used in the assay of a range of objects of varying size and shape in locations where alternative monitoring equipment would not normally be available.

The DISPIM<sup>®</sup> system has recently been successfully used for the in-situ measurement of crated and drummed Plutonium Contaminated Material (PCM) waste items to provide Pu mass values in preparation for their treatment as part of a decommissioning program.

A description of the operation of the DISPIM<sup>®</sup> system is given, including calibration of the system prior to the on-site survey, the measurement and quality control methodology employed during assay of waste items and discussion of the data analysis techniques. The results of "imaging" measurements (i.e. Pu mass distribution results) recorded from the PCM crate measurements are likely to be of particular benefit when planning any future operations on these waste items.

The results of this DISPIM<sup>®</sup> survey represent a complete solution for the customer with minimal requirement for use of their own resources. Technically substantiated Pu mass assay results have been provided which can now be easily adopted for use in criticality safety cases for the future treatment of these waste items. In summary, the DISPIM<sup>®</sup> system has successfully delivered a safe, fast and cost effective measurement solution to this challenging assay task and demonstrated its flexibility as a PCM monitoring system.

## INTRODUCTION

The Decommissioning In-Situ Plutonium Inventory Monitor (DISPIM<sup>®</sup>) is a versatile monitoring tool useful in a range of waste management, decommissioning and security applications. Consisting of both Coincidence and Totals neutron counting combined with High Resolution Gamma Spectroscopy (HRGS) measurement capabilities, the system represents a complete solution, able to meet the unique challenges often associated with Pu assay. Furthermore, the modular design and versatile, highly deployable nature of the system allows for a wide range of object sizes and shapes to be assayed in-situ, in environments where the use of alternative monitoring equipment would be impractical or prohibitively expensive.

A unique feature of DISPIM<sup>®</sup> lies in the "imaging" capability of the system. Using a mathematical model the system can determine plutonium concentration locations based on recorded NCC and TNC count rates from an item. This feature is particularly useful in decommissioning activities, allowing targeted clean-up by decommissioning teams, thereby minimising dose uptake and aiding safety during further decommissioning operations.

This paper describes the use of the system in recent measurements of crated and drummed PCM waste items. The measurement campaign described included planning and on plant measurements through to expert analysis and the presentation of technically substantiated Pu mass results used in the confirmation of historic assay values. The results have subsequently formed the basis of criticality safety cases to allow movement of the items to an engineered long-term storage facility.

# DESCRIPTION OF THE DISPIM® SYSTEM

The measurement system<sup>1</sup> (Fig. 1) consists of neutron counting and HRGS components which can be operated either in conjunction or as standalone systems. The neutron counting component of the system consists of 24 identical neutron-counting modules, each containing 2 He<sup>3</sup> detectors. While each of the 24 modules can be deployed in free space or a purpose built deployment mechanism, standard deployment is horizontally located within a background-shielded assembly, which is in turn located on a pump truck. This deployment allows easy configuration around a variety of differently shaped objects, at a range of heights.

<sup>&</sup>lt;sup>1</sup> DISPIM is a registered trademark of BIL Solutions Ltd

The DISPIM Imaging technology is covered by European Patent EP1012630 and US patent 402968. The Neutron Counting technology is covered by International Patent W0 00/67044 and US patent 6912485



Fig. 1. The DISPIM® Imaging System

The HRGS system comprises a  $LN_2$  cooled germanium detector and associated electronics. The detector is collimated and located on a pump truck similar to those housing the neutron detector assemblies. This allows the detector to be maneuvered easily to record spectra from either an entire measurement item or areas of specific interest such as Pu hot spots identified using the neutron counting system.

A mobile electronics and processing cabinet houses both the system power supplies and data processing electronics and PC's. Operator interface is controlled from the cubicle.

# **OPERATION OF THE DISPIM<sup>®</sup> SYSTEM**

## **Crated Waste Items**

## Deployment

The measurement of crated waste items follows a specific quality controlled procedure developed to ensure technically substantiated, reliable results are provided from a survey. Up to six DISPIM<sup>®</sup> assemblies are deployed around the waste item, to gain the best possible coverage of the item. The item dimensions and position of the assemblies relative to the crate are then input into the DISPIM<sup>®</sup> software. The re-configurable nature of the system and the fact that the geometry of waste items is different from measurement to measurement means that the neutron detection efficiency (and hence calibration) of the system also changes from measurement to measurement. To account for this, the system is supplied with a 3-dimensional map of detection

efficiency points around each detector module. Along with the operator-supplied information regarding the deployment geometry for the waste item, this allows the system to automatically reconstruct a calibration map for each measurement configuration. DISPIM<sup>®</sup> uses this calibration map to accurately predict the expected response of each detector module for any distribution of neutron sources within that particular waste item.

### **Quality Control**

Following specification of the item and measurement geometry, a standardisation routine is completed using a Cf-252 source placed at fixed locations within each detector assembly to confirm that the system has been correctly deployed and that each neutron module is operating correctly. The standardisation takes the form of a PASS/FAIL test of each module to determine that neutron count rates are within pre-determined limits.

The correct operation of the DISPIM<sup>®</sup> system is further tested during the calibration check phase of an item measurement. However, the main purpose of this phase is to test the item for neutron interfering material. This check ensures that the neutron interfering properties of the item are not outside the calibration of the system and in addition allows a suitable matrix correction to be applied to the final Pu mass results should this be appropriate. For crated waste items, the check involves measurements of a Cf-252 source (the same source used in standardisation measurements already described) located on each face of the crated waste item. The source positions are input in the DISPIM<sup>®</sup> software and the system automatically calculates the expected count rate which would be observed at each neutron module assuming the source was located in free space. The actual measurements are then compared with the calculated values and against predefined limits. The results of the calibration check are used to confirm that the neutron interfering properties of the item are acceptable within the system calibration. In addition to this, the results are also used to determine whether an "imaging" or "non-imaging" DISPIM<sup>®</sup> measurement can be performed.

#### Assay

Following a successful calibration check of the system, the total neutron response of each module and the coincidence response of each assembly and the system are measured, usually over a period lasting several hours or ideally overnight. At the completion of the measurement the system reports a "non-imaging" Pu-240 equivalent mass result for the waste item based on the calculated "worst case" efficiency within the measurement envelope. In addition, an "imaging" measurement uses a mathematical model to select plutonium concentration locations and predicts all module count rates. The agreement obtained between the predicted and actual measured count rates of each module is then evaluated using a chi-square test. Single sources and multiple sources are iteratively located in the model by a non-linear least squares approach until the chi-square test reaches a minimum value. Once the minimisation process has completed, the model parameters indicate the relative activity of each plutonium concentration and its position on 3 dimensional co-ordinate axes. In this instance, Pu mass values at each Pu hot spot and for the whole item are determined using the calculated measurement efficiency at the

predicted Pu location. In most instances, the "imaging" algorithms can provide a significantly more accurate assay result.

Once the Pu content of an item has been determined, HRGS measurements of either the whole item or specific Pu hot spots located in the "imaging" result can be made and used to determine the isotopic composition of the material. This value (or alternatively default isotopic values if isotopic composition of the item is already known or cannot be measured) can then be combined with the Pu-240 equivalent mass results determined from neutron measurements to provide a total Pu mass result for both the whole system and individual Pu hot spots.

All data recorded from the neutron standardisation, calibration and measurement of an item, along with HRGS spectra and isotopic analysis results are automatically archived by the system. Therefore, additional testing, checking and analysis of the measurement data can be performed offline where required.

#### **Drummed Waste Items**

The measurement of PCM drums using the DISPIM<sup>®</sup> system has been performed using a calibration specific to the needs of the customer, who required technically substantiated Pu mass values in verification of historic Pu mass figures for use in criticality safety cases. 18 drums, varying in size between 200-400L required assay using the DISPIM<sup>®</sup> system to determine a nuclear safety Pu mass result and in addition confirm no uranium was present in the drums. The normal DISPIM<sup>®</sup> calibration could be used in the measurement of drummed waste items, however, this was considered to be inappropriate to drum measurements for the following reasons. Firstly, drummed waste items have a significantly smaller measurement volume than that associated with crated items, therefore an "imaging" measurement was not required since the assay errors due to geometry are smaller. Secondly, it was expected that waste drums would be likely to contain significant quantities of matrix material. Therefore, an appropriate matrix correction had to be applied which would allow criticality safety arguments to be satisfied.

Drum measurements (including calibration measurements) were performed using a custom-built measurement platform. For each measurement, the drum is located at the centre of the platform and four DISPIM<sup>®</sup> assemblies reproducibly located at each side around the drum (Fig. 2). Acquisition times varied from drum to drum, however, despite the high background environment reasonable statistical precision could be obtained for most drums from acquisition times of approximately 1 hour. NCC/Pu-240 equivalent calibration factors were determined prior to commencing the survey on plant, as described in the following section.



Fig. 2. Configuration of DISPIM® Imaging System drum measurements

In addition to determination of Pu-240 equivalent calibration factors, "add-a-source" measurements were performed using the DISPIM<sup>®</sup> Cf-252 calibration source attached to the side of the drum. These measurements were performed at two drum heights (approximately the drum half and quarter heights) and from two sides of the drum. The "add-a-source" measurements were repeated during the measurements of waste drums and provided an indication of the level of neutron interfering matrix material contained within the drum.

Following neutron measurements, HRGS measurements were performed on the whole drum from two separate sides. The isotopic composition of the Pu was determined using MGA (Reference [1]) and used in conjunction with Pu-240 equivalent mass results to determine total Pu mass values from each drum.

The measurement of drummed waste items again followed a quality controlled procedure ensuring technically substantiated, reliable results were provided from the measurement campaign.

## RESULTS

## **Crated Waste Items**

Two crated items of PCM waste were measured using the internal DISPIM<sup>®</sup> calibration as described previously and the following results were recorded for each crate:

## Crate 1

Calibration check measurements performed on the crate (as described previously) indicated that no significant neutron interfering matrix material was contained within and that the item was suitable for an "imaging" measurement. NCC and TNC count rates were recorded in an overnight measurement and the imaging algorithms run to determine Pu location. The following 3D representation of the item and DISPIM<sup>®</sup> deployment is shown in Fig. 3. The Pu location determined by the system is also shown.



Fig. 3. DISPIM<sup>®</sup> "Imaging" result from measurement of Crate 1. Two Pu hotspots are indicated by the model

Two Pu hot spots have been identified by the system. Both hot spots are located close to the bottom edge of the item. In this instance the Pu locations determined by the system are located close to the worst case efficiency position within the measurement envelope. Therefore, similar Pu-240 equivalent mass values have been reported in both the "imaging" and "non-imaging" results.

HRGS measurements performed on the crate have identified the isotopic composition of the material within the crate to be consistent with commercial grade Magnox plutonium. This information has allowed a total Pu mass to be calculated for the crate as well as for each Pu hotspot identified by the system.

Further to DISPIM<sup>®</sup> measurements of this item, additional measurements have been performed using transmission X-ray exposure and BIL Solutions RadScan<sup>®</sup> : 800 4pi gamma imaging system. The Pu locations identified by DISPIM<sup>®</sup> have therefore been tied in with the measurement results from X-ray exposure, RadScan<sup>®</sup> : 800 and drawings of the crated item and are being used to aid in the construction of a criticality safety case.

#### Crate 2

Calibration check measurements performed on Crate 2 indicated a more significant interfering matrix than that indicated for Crate 1. Therefore, a routine "imaging" result was not immediately possible for this crate. However, as before, NCC and TNC count rates were recorded in an overnight measurement and a Pu-240 equivalent "non-imaging" result quoted (i.e. based on the "worst case" efficiency within the crate). HRGS measurements were then performed, which were consistent with commercial grade Magnox plutonium and total mass values determined.

Offline analysis of the results recorded by the system allowed a number of conclusions to be drawn regarding Crate 2. Firstly, examination of the recorded TNC rates from calibration check measurements indicated the presence of significantly more neutron interfering material than had been observed in Crate 1, where very little interference was observed. Additionally, the observed distribution of counts indicated that the interfering matrix was located at the centre of the crate. These observations suggest determination of an "imaging" DISPIM<sup>®</sup> result could be biased. However, an offline "imaging" analysis was performed and used to provide additional information on the crated item.

Initial attempts to produce an imaging result from the recorded neutron measurement data failed and resulted in large values of  $\chi^2$ . Since information recorded on the side of the crate suggested the possible presence of plutonium fluoride, the effect of altering the ( $\alpha$ ,n) ratio used by the DISPIM<sup>®</sup> imaging algorithm was tested. The ( $\alpha$ ,n) ratio for plutonium fluoride is significantly larger than that of plutonium oxide and can therefore create a significant disparity between recorded TNC and NCC rates, both of which are used in calculation of an imaging result.

Increasing the  $(\alpha,n)$  ratio allowed the best possible fit to the data and an "imaging" result to be calculated. The results are displayed in the same manner as for Crate 1 and indicated the presence of only one Pu hotspot (although additional sources may be likely, which are shielded by the crate's matrix). The Pu mass value determined from the "imaging" result is significantly lower than that from the "non-imaging" analysis since the Pu is located in a position with significantly higher detection efficiency than the "worst case" position. However, given the possible bias in the "imaging" result was assigned to the crate. Again, the results from this measurement are currently being used in construction of a criticality safety case to allow movement of the item to a waste treatment facility.

#### **Drum Calibration Measurements**

200L and 400L calibration drums were prepared with a "worst case" matrix contents of PVC packed at 0.3gcm<sup>-3</sup>. This density is the maximum that can be achieved by manually packing small (~1m<sup>2</sup>) sheets of PVC into the drum and is considered a realistic "worst case" neutron interfering matrix. Each drum (both 200L and 400L) was located on the measurement platform and four DISPIM<sup>®</sup> assemblies deployed around the drum (Fig. 2). Using the Cf-252 calibration source, a series of measurements were performed at a range of positions throughout the volume of each calibration drum. From these measurements it was determined that the optimum

measurement geometry of the system was obtained with the assemblies in their lowest positions, where the lowest module in each assembly is located below the measurement platform. Additionally, these measurements indicated that the position of lowest detection efficiency in both the 200L and 400L drum was in the bottom centre of the drum. At the position of lowest sensitivity in this geometry, detection efficiencies of approximately 3.5% and 2.5% were obtained.

Following determination of the position of lowest sensitivity, 5 further measurements were performed of increasing Pu-240 equivalent masses (using the Cf-252 source and Pu fissile standards) at the position of lowest sensitivity in both the 200L and 400L drum. Using these results Nuclear Safety and Accountancy mass NCC/Pu-240 equivalent calibration factors were then calculated for both the 200L and 400L drums plus PVC matrix material. In addition, equivalent calibration factors were also calculated for drums without the "worst case" PVC matrix material.

To verify the appropriateness of the assumed matrix correction, a series of "add-a-source" calibration measurements were performed on the drums. These measurements involved locating the DISPIM<sup>®</sup> Cf-252 calibration source on the surface of each drum and examining the neutron count rates in the DISPIM<sup>®</sup> assembly opposite the source (i.e. through the waste matrix). These measurements were performed on both empty drums and those containing the PVC matrix material. The results recorded from the calibration source measurements are shown in Fig. 4 alongside those recorded from waste drum measurements for comparison.

Recording calibration measurements in this manner served a number of purposes. Firstly, the matrix material of waste drums could be tested to ensure the neutron interfering properties of the waste matrix were not significantly greater than that assumed in the worst case calibration. This test was particularly useful in demonstrating that for drums of intermediate size between the 200L and 400L calibration drums, the 200L calibration drum was suitably pessimistic and could be applied to the determination of Pu mass safely. Additionally, if measurement results could be shown to be comparable to those recorded with no matrix material present, applying a less pessimistic calibration factor could be justified as will be demonstrated in the following section.

#### Waste Drum Measurements

Assay of each of the 18 waste drums was made using the DISPIM<sup>®</sup> system and Nuclear Safety and Accountancy Pu mass values were calculated for each drum. The "add-a-source" transmission measurements from each drum were compared with those recorded during determination of the NCC/Pu-240 equivalent calibration factors and used to determine the most appropriate calibration factor. A selection of the "add-a-source" transmission results are shown in Fig. 4



Fig. 4. Selection of "add-a-source" transmission data from calibration and waste drum measurements (All measurements shown were performed at the drum quarter height. Modules 1-4 represent each pair of neutron detectors in a DISPIM<sup>®</sup> assembly, 4 being in the lowest position)

"Add-a-source" calibration measurements performed on the empty drum and 200L and 400L PVC filled calibration drums are shown for comparison. All of the drums measured (excluding the empty calibration drum) had quarter height transmission values either equivalent to or more commonly significantly lower than those measured at the half height. In addition, of the 18 drums measured, the transmission measured in all was significantly greater than the ~10-20% measured from the 400L calibration drum filled with PVC matrix material (e.g. Drums 2, 4 and 18 in Fig. 4). Therefore, the 400L drum (including PVC matrix) calibration factor was considered to be overly pessimistic in determining mass values for these drums. The 200L calibration factor (including PVC matrix) was therefore used for all except 3 drums (e.g. Drums 7 and 17 in Fig. 4). For each of these three drums the transmission measurements reported count rates significantly greater than those from the 200L calibration drum with PVC matrix. Indeed, transmission values indicated little attenuation of the signal and were generally consistent with the calibration measurements made on an empty drum. For each of these three drums an empty drum calibration was considered the most appropriate and was therefore used to determine Pu content of the drums. Using the appropriate calibration factors, Nuclear Safety and Accountancy Pu-240 equivalent mass values were determined for each of the drums.

Two HRGS measurements were performed for each of the drums, as described. The results recorded ranged from approximately 5 to 16% Pu-240 equivalent. Three drums failed to produce an MGA result due to low Pu content and suitably pessimistic default values were therefore applied. Combining the results of HRGS measurements with Pu-240 equivalent masses then allowed the calculation of total Pu mass values.

All 18 of the waste drum measurements were successfully completed over a period of 6 working days on plant. The results provided have now allowed preparation of a safety case for continued storage of the drums and future transport to a waste treatment facility.

## CONCLUSIONS

The DISPIM<sup>®</sup> system has recently been successfully used for the in-situ measurement of crated and drummed PCM waste items to provide Pu mass values in preparation for treating the items as part of a decommissioning program. The range of items measured of varying size and shape demonstrates the versatility of the system to a wide range of challenges commonly found in waste management and decommissioning environments. In addition, the measurement examples highlight the systems use in a customised measurement solution tailored to meet a specific measurement challenge and its ability to provide technically substantiated results suitable for use in criticality safety assessments.

#### REFERENCES

1. MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances, Volume 1 Methods and Algorithms, R Gunnink, April 1990