# Measurement of the U-235 Content of Concreted Waste Drums - 8244

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## ABSTRACT

A challenging assay situation recently arose, whereby the fissile (i.e. total plutonium plus U-235) content of a population of 164 historical waste drums containing concrete needed to be measured, to comply with nuclear safety limits for transport to, and interim storage within, an Engineered Drum Store.

BIL Solutions Ltd has developed a new methodology for measurement of the U-235 content of these "concrete" drums, because the approach normally used by the in-situ Drum Monitors was found to be overly pessimistic. Initial investigations indicated significant quantities of uranium were present in these drums (but negligible plutonium), mixed with Np-237 and / or Ra-226. These initial measurements also indicated that the uranium was likely to be depleted in enrichment. The U-235 content was therefore determined by measuring the U-238 mass via the passive coincident neutron emission, and combining this with the U-235 and U-238 isotopic abundances, obtained by analysis of a gamma spectrum.

Of the uranium isotopic analysis codes available, the FRAM (Fixed energy, Response function Analysis with Multiple efficiencies) software was selected as being most suitable for this application. A wide gamma-ray energy range is used (i.e. 120 keV to 1200 keV) which was considered more likely to yield results when there is significant attenuation. The software is also user configurable, enabling interferences from the other radionuclides present (i.e. Np-237 and Ra-226) to be accounted for. A series of test measurements were performed with well-characterised uranium sources attenuated by concrete shielding, to gain confidence in the performance of FRAM under such conditions. These test measurements indicated that FRAM was able to correctly determine the enrichment of heavily shielded uranium.

The new U-235 measurement methodology was then applied to the population of concrete drums; successfully yielding U-235 results despite the dense waste matrix and significant interference from Np-237 and / or Ra-226. The U-235 mass results obtained were significantly more accurate than those that would have been obtained had the traditional approach (i.e. transmission corrected 186keV count rate) been used, facilitating safe transport and storage of the drums.

## INTRODUCTION

As part of the Plutonium Contaminated Materials (PCM) retrievals work at the UK Low Level Waste Repository (LLWR), a population of 164 overweight (>150kg) PCM waste drums required transport from the LLWR to engineered storage facilities at Sellafield. These drums had previously been segregated from the general population because their weight exceeded 150kg, the limit for the container used to transport the drums. Prior to transport, these drums required assay in a Drum Monitor to determine their fissile content (total plutonium plus U-235) to demonstrate compliance with nuclear safety limits for transport and subsequent storage. A Best Estimate measurement of the activity of any gamma-emitting radionuclides that are present was also required to comply with transport regulations.

Measurement of the plutonium content was relatively straightforward, since a concrete matrix is within the calibration envelope of the passive neutron coincidence measurement performed by the in-situ Drum Monitors. The U-235 measurement, however, presented a significant challenge. The Drum Monitors normally quantify the U-235 mass by HRGS measurement of the 186keV gamma emission, which is corrected for attenuation within the waste matrix by performing transmission source measurements. This approach was found to be highly inaccurate for these drums, leading to gross overestimates of the U-235 content, because of the following factors:

- The drums have significant uranium content, and hence the count rates in the 186keV gamma peak are appreciable.
- Transmission source measurements indicate that the waste within these drums is very dense, and the correction factors to account for the matrix attenuation will be very large (e.g. measured transmission of 0.00014% through drum diameter at 186 keV in a typical drum). Note that because of the high density of the waste the automated measurement will fail and the correction factors will need to be calculated offline.
- The calculation of the U-235 Nuclear Safety Value makes a pessimistic assumption that the uranium is located at the centre of the drum. In the high density waste present in these drums, this assumption can result in overestimations of the U235 mass by one to two orders of magnitude compared to a case where the uranium is present as a uniform distribution throughout the drum.
- The uranium calibration assumes there is significant self-attenuation within the uranium (i.e. a factor of 8.33 compared to an infinitely dilute source). Application of the uranium Differential Peak Absorption (DPA) analysis [1] to justify removal of this factor was found to be unsuccessful because of the high waste density.
- The collimation of the HRGS detectors is fairly open; such that the field of view (i.e. 27cm on drum axis) is significantly larger than the vertical separation of the segments in the segmented scan (i.e. 6.7cm). This segment overlap results in overestimation of the U-235 mass by at least a factor of three.

The above factors will combine together to yield very large overestimates of the U-235 content (which could easily exceed two orders of magnitude) under typical measurement conditions. An alternative methodology for determination of the U-235 content was clearly required to avoid operational problems due to the calculation of overly pessimistic U-235 mass results.

BIL Solutions Ltd was contracted to develop an appropriate method, the initial stage of which involved investigation of the properties of these overweight drums.

## PROPERTIES OF THE OVERWEIGHT DRUMS

An initial investigation of the overweight drums was carried out to determine their origin, physical properties and the radionuclides present. Unfortunately, despite searches in both local and national document archives, no useful historical information was obtained regarding the origin of these drums which had been stored at the LLWR since the 1960s. It was noted that the drums have sequential identification numbers, which could imply that they were generated from the same nuclear plant or process.

### Visual Inspection of Contents

Eighteen drums were physically opened and the contents visually inspected. All eighteen drums comprised a drum (≈ 200 litres in volume) with a slightly smaller diameter drum inside. The inner drums were all filled with a solid concrete matrix, as shown in Fig.1. Although the origin of the drums is unknown, it is likely that the concrete was added to reduce the dose rate from the high gamma activity present. It is not known whether this concrete was mixed in with the waste, poured into an annulus surrounding an inner waste container or just poured on top of the waste. Besides concrete, the materials present in the waste are unknown and must be assumed to be consistent with general PCM waste.

**Fig. 1. Photograph taken during inspection of one of the overweight drums.**

## Drum Weights and Uniformity of Matrix

The full population of 164 drums were weighed and the majority of the drums (i.e. 81%) had masses in the range 190 kg to 270 kg. The heaviest drum weighed 303 kg, but 4 drums were less than the 150 kg limit, with the lowest weighing 125 kg. Assuming the drums are almost full, as indicated from the visual inspection, the density of the waste is in the range 1.0 g/cm$^3$ to 1.5 g/cm$^3$. This density is slightly lower than expected for solid concrete, suggesting some heterogeneity. To further examine the uniformity of the waste matrix, segmented HRGS transmission source measurements of several drums were performed. The segmented scan uses a Eu-152 source, and the drum is segmented into 16 vertical segments. Example segmented scan results for 3 overweight drums are shown in Table I.

**Table I. Calculated 186 keV Transmission Correction Factors for 3 Overweight Drums**

| Segment Number | Transmission Correction Factor at 186 keV Calculated by Drum Monitor | | |
|---|---|---|---|
| | Drum 1 (250 kg) | Drum 2 (250 kg) | Drum 3 (230 kg) |
| 1 (bottom) | 3.864 | 1.465 | 5.000 |
| 2 | Too dense | Too dense | Too dense |
| 3 | Too dense | Too dense | Too dense |
| 4 | Too dense | Too dense | 7.501 |
| 5 | Too dense | Too dense | Too dense |
| 6 | Too dense | Too dense | Too dense |
| 7 | 6.824 | 4.203 | 7.848 |
| 8 | Too dense | 3.742 | 5.172 |
| 9 | 7.864 | 3.658 | Too dense |
| 10 | 5.798 | 3.198 | Too dense |
| 11 | Too dense | Too dense | Too dense |
| 12 | 1.327 | 6.419 | 1.627 |
| 13 | 1.133 | 1.219 | 1.137 |
| 14 | 1.217 | 1.765 | 1.502 |
| 15 | 1.124 | 1.214 | 1.145 |
| 16 (top) | 1.152 | 1.969 | 1.597 |

The calculated transmission correction factors at 186 keV are shown in Table I: this is the factor by which the count rate in the U-235 186 keV gamma peak is multiplied to correct for matrix attenuation. It can also be seen that in many segments, the high density of the waste matrix meant it was not possible to calculate the transmission fraction at 186 keV (the required gamma peaks from the Eu-152 transmission source were not detected), thus preventing calculation of the U-235 mass using the normal method employed by the Drum Monitors. It is also evident that the matrices are heterogeneous, since the 186 keV transmission varies significantly from segment to segment, indicating variations in waste density. From the data it is also apparent that the top of the inner drum containing the dense matrix occurs at approximately Segment 11 or 12, which is consistent with a ≈ 80 cm fill height. Although only three drums have been included in Table I, data for the wider population of overweight drums is entirely consistent.

## Radionuclide Constituents

Passive gamma spectra were acquired for all of the overweight drums to determine their radionuclide constituents. Fig. 2 summarises the significant radionuclides identified in these spectra.

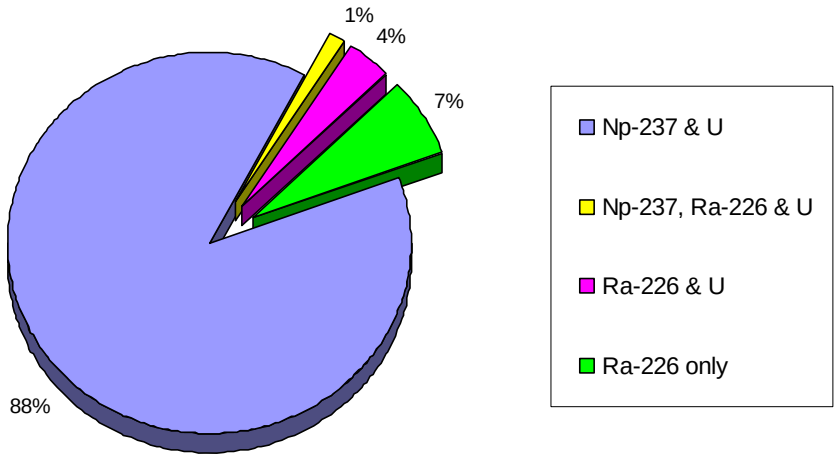


**Fig. 2. Summary of Significant Radionuclide Constituents of Overweight Drums.**

As can be seen from Fig. 2, most of the drums (i.e. 88%) contain Np-237 mixed with uranium. A significant fraction (i.e. 11%) contains significant amounts of Ra-226, either mixed with Np-237 and / or uranium or in isolation. Almost all of the drums also contain small quantities of Cs-137 and Ra-226, and a fraction of the drums (i.e. 21%) show small gamma peaks characteristic of plutonium (i.e. at 208 keV, 375 keV and 414 keV). However, the gamma peaks detected were very small, and the nuclides identified may therefore just be present in the background and not actually within the drum. Another possibility is that the plutonium is present as trace amounts of contamination on the outer drum and is not inside in the inner waste drum. Contamination is considered more likely, since the overweight drums were previously stored in an area with a history of plutonium contamination. Further evidence to support the lack of significant amounts of plutonium in these drums was borne out by the low neutron count rates that were measured for the drums with plutonium gamma peaks evident. These low neutron count rates rule out the possibility that there is a significant amount of plutonium present, and that the gamma emissions are being heavily shielded by the dense concrete matrix.

The presence of Np-237 and Ra-226 might explain why the drums were filled with concrete, since both nuclides have daughter products with high specific gamma activities. The presence of significant amounts of Np-237 is likely to be associated with the production of Pu-238. Np-237 is a key component in the manufacture of Pu-238, which has widespread use as an alpha emitter or heat source. It is not known why Ra-226 is also present in several of the drums, as this would not normally be associated with the same process.

## Uranium Isotopic Analysis

Uranium Isotopic analysis was then performed on the gamma spectra acquired for the overweight drums to ascertain the uranium enrichment. Two different commercially available software packages were evaluated for this purpose; PC/FRAM (Fixed energy, Response function Analysis with Multiple efficiencies) developed by the Los Alamos National Laboratory [2][5], and MGAU (Multi-Group Analysis Uranium) developed by the Lawrence Livermore National Laboratory [3][6]. The FRAM software uses a much wider gamma energy range in the analysis (i.e. 120 keV to 1001 keV) compared to MGAU (i.e. 80 keV to 130 keV), and is therefore expected to perform better in situations where there may be significant attenuation. FRAM also allows more user interaction, and it is possible to adjust the analysis parameter files to cope when there are nuclides present which emit gamma-rays that interfere with those used in the uranium analysis. However, this flexibility means that FRAM requires a more experienced user and the results must be scrutinised to confirm their acceptability. In contrast, MGAU can be used more as a "black box".

A summary of the advantages and disadvantages of these two software packages for uranium isotopic analysis of the overweight drums is provided in Table II.

**Table II. Advantages and Disadvantages of FRAM and MGAU for Analysis of Overweight Drums.**

| | Advantages | Disadvantages |
|---|---|---|
| **FRAM** | Broad energy range (120 keV to 1001 keV) so less affected by attenuation.<br>Very versatile; user can modify parameter files to correct for interferences. | Wide energy range means accuracy of results highly dependent on efficiency fit.<br>Requires a more experienced user. |
| **MGAU** | Narrow energy range so results less dependent on accuracy of efficiency fit and less prone to interference.<br>Simple to use. | Analyses low energy region only (84 keV to 130 keV), which is more easily attenuated.<br>Limited scope for user configuration. |

FRAM was considered the more likely of the two software packages to accurately determine the uranium enrichment of the overweight drums, because of the broad energy range and ability to modify the parameter files to cope with interferences. However, further confidence in the ability of FRAM to perform accurate measurements under the challenging measurement conditions posed by the overweight drums was required before it could be used to calculate the U-235 mass Nuclear Safety Values. Test measurements were conducted to examine the performance when measuring well-characterised uranium sources shielded by different thicknesses of concrete shielding. Three different enrichments tested: natural (i.e. 0.72% U-235), 1.46% U-235 and 2.13% U-235, shielded by 10cm and 20cm thick concrete (density = 2 g/cm$^3$). Low enrichment sources were selected to represent as closely as possible the conditions of the overweight drums. Tests were also performed to simulate the interference from other nuclides present in the overweight drums. Interference from a Ra-226 source, located adjacent to the uranium, was also tested. Unfortunately no Np-237 sources were available. The effect of source-to-detector distance was also examined, since the Drum Monitor will position the uranium 40 cm to 60 cm from the HRGS detector.

The results obtained from the testing of FRAM are summarised in Table III.

**Table III. Results from Test Measurements of Uranium Sources with FRAM.**

| Sources Present (%U-235) | Thickness of Concrete (cm) | Source to Detector Distance (cm) | Count time (s) | FRAM Measured %U-235 (±1σ statistical uncertainty) | Relative Deviation from Reference Value % |
|---|---|---|---|---|---|
| **Effect of Concrete Shielding** | | | | | |
| 915g Natural Uranium Metal (0.72% U-235) | 0 | 10 | 3600 | 0.79 ± 0.08 | 10% |
| | 10 | 10 | 7200 | 0.63 ± 0.29 | -12% |
| 829g $UO_2$ Pellets (1.46% U-235) | 0 | 10 | 1100 | 1.60 ± 0.18 | 10% |
| | 10 | 10 | 4500 | 1.19 ± 0.22 | -19% |
| 944g $UO_2$ Pellets (2.13% U-235) | 0 | 10 | 1800 | 1.87 ± 0.24 | -12% |
| | 10 | 10 | 7200 | 1.66 ± 0.27 | -22% |
| | 20 | 20 | 15300 | 0.10 ± 0.04 | -95% |
| **Effect of Ra-226 Interference** | | | | | |
| 915g Natural Uranium Metal (0.72% U-235) + 0.3 MBq Ra-226 | 0 | 10 | 2100 | 0.75 ± 0.39 | 5% |
| 829g $UO_2$ Pellets (1.46% U-235) + 0.3 MBq Ra-226 | 0 | 10 | 1800 | 1.81 ± 0.20 | 24% |
| 944g $UO_2$ Pellets (2.13% U-235) + 0.3 MBq Ra-226 | 0 | 10 | 1800 | 2.31 ± 0.32 | -10% |
| **Effect of Varying Source-to-Detector Distance** | | | | | |
| 944g $UO_2$ Pellets (2.13% U-235) | 0 | 8 | 1800 | 2.15 ± 0.09 | 1% |
| | 0 | 20 | 1800 | 2.21 ± 0.17 | 4% |
| | 0 | 40 | 3300 | 2.13 ± 0.21 | 0% |
| | 0 | 60 | 3900 | 1.98 ± 0.26 | -7% |
| | 0 | 80 | 4200 | 2.07 ± 0.33 | -3% |

The tests with different thicknesses of concrete shielding indicated that it was possible to measure the uranium enrichment through 10 cm of concrete shielding, although an increase in measurement time was necessary and some systematic bias was evident. The measurement with 20 cm shielding failed despite a long measurement time. Comparison of the efficiency curves fitted by FRAM for these test measurements against those obtained for the overweight drums, indicates that the shielding of the uranium in the overweight drums is generally less than 10 cm thick concrete. This may indicate that the uranium is heterogeneously distributed in the drums, with the signal measured for the uranium near the edge of the drum dominating.

The tests with a Ra-226 source present indicated that the FRAM parameter files could be modified to successfully cope with interference from Ra-226. The guidelines for modification of parameter files in Reference [2] were followed, and the FRAM Developers at Los Alamos National Laboratory (LANL) were contacted, who confirmed that the approaches we had taken for the interferences from both Ra-226 and Np-237 were appropriate. To cope with Ra-226, the interferences to the U-235 186 keV and U-238 258 keV peaks were stripped away (by reference to a nearby Ra-226 daughter peak), and Ra-226 daughter peaks were used for FRAM internal calibrations. Similarly, to cope with Np-237 the interference to the U-235 143keV peak was stripped away, and Np-237 daughter peaks were used for internal calibrations.

The tests at a range of source-to-detector distances showed that there was a small systematic bias at greater source detector distances, and that as expected longer count times were required as the separation increased.

Using these test measurements, criteria were developed to determine whether the FRAM results were acceptable. Spectra were saved at approximately 100s intervals throughout each measurement, analysed with FRAM and examined in detail. The %U-235 results were found to significantly deviate from the expected value at early count times, but stabilise as the count time increased and certain conditions relating to the quality of the gamma spectrum improved. Examination of these intermediate results made it possible to identify criteria that could be used to ascertain whether the FRAM results were accurate. To give an example, the uncertainty on the U-238 258 keV peak was found to be a useful indicator of the accuracy of the FRAM results. When this peak was detected with a reasonable statistical uncertainty (i.e. <30%), there was a strong likelihood that the FRAM results would be accurate. This indicator was particularly useful when measuring the overweight drums, where it was not possible to determine the measurement time in advance. Examination of the uncertainty on the 258 keV peak allowed the operator to stop the measurement at the appropriate time, thus avoiding counting the drum for too long or not long enough.

In summary, these tests gave confidence that FRAM could be used to obtain accurate uranium isotopic analysis results for the overweight drums, providing:

(i) An appropriate parameter set is used in the analysis to account for interferences from Np-237 and Ra-226.

(ii) The results are examined and confirmed to meet the following criteria:
- Fitted efficiency curve has the expected shape.
- The fits on the individual analysis peaks are good.
- Statistical uncertainty on key peaks (e.g. 186keV, 258keV, 1001keV) are acceptably low.
- Isotopic analysis results are consistent with MGA (if also successful) and with range expected for depleted uranium (i.e. >0.2% U-235).

(iii) A systematic uncertainty is applied to the results to account for the observed bias and ensure that the nuclear safety U-235 mass values would not be significantly underestimated. This was conservatively determined to be 22.7% at 1 sigma confidence.

The FRAM and MGAU uranium isotopic results for a sample of 25 of the overweight drums are shown in Fig. 3.

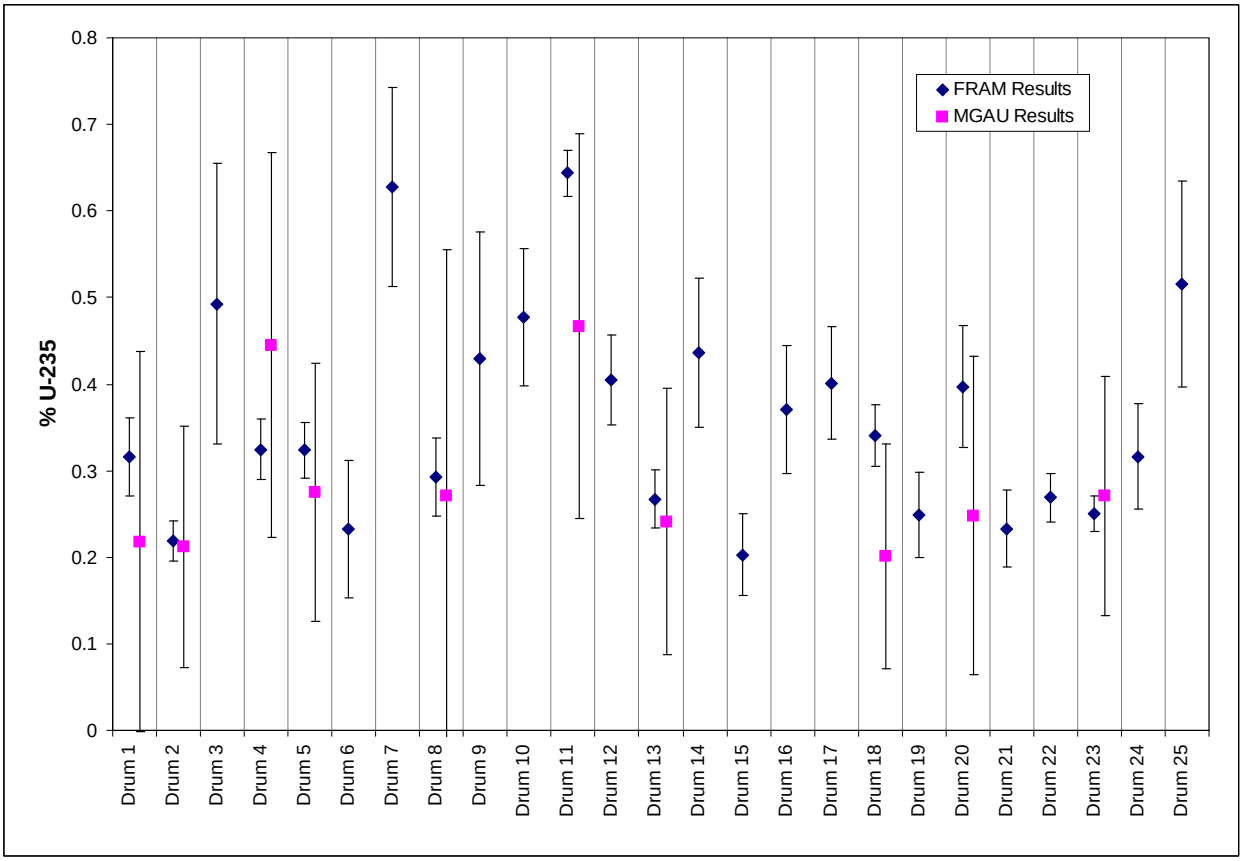


**Fig. 3. %U-235 Results from FRAM [5] and MGAU [6] for a Sample of Overweight Drums (error bars are the 1 sigma statistical uncertainties reported by the isotopic analysis software).**

It is evident from Fig. 3 that, as expected, FRAM is more successful when determining the isotopic composition of these overweight drums. FRAM was able to produce isotopic analysis results for all 25 drums in the sample, but MGA was only successful for 10 of the drums. Furthermore, it can be seen that although the MGAU results are consistent with the FRAM results in all cases, there is a very large statistical uncertainty on the MGAU results. This is not surprising given the low gamma energy range used in the MGAU analysis. This region of the spectrum has a large Compton background due to the significant matrix attenuation (and hence scattering) and the presence of higher energy gamma emitters such as Np-237 and Ra-226.

The results in Fig. 3 indicate that the uranium present in these drums is depleted in enrichment (i.e. %U-235 less than 0.72%). The FRAM results obtained for the full population of overweight drums were also consistent with those presented for the sample of 25 drums: the measured enrichments ranging from 0.2% U-235 to 0.9% U-235, with 93% of the measured results being below 0.72% U-235. Note that 77% of the results were between 0.2% and 0.5% U-235. Although the FRAM results for a few drums (i.e. 7%) exceeded natural enrichment, they were consistent with natural or depleted uranium when the uncertainty on the %U-235 results was taken into consideration. Further qualitative evidence for the low enrichment of the uranium can be obtained from the gamma spectra, because the gamma peaks associated with higher enriched uranium (e.g. the 120.9keV peak from U-234) were absent.

There was also no evidence of uranium with different enrichments being present in the same drum. A significant proportion of the drums were measured more than once, with the drum either being rotated continuously or held static throughout the measurement. The results from the measurements of the same drum in different orientations were always in good agreement, indicating that it is highly unlikely that different enrichments are present in the drum. The scatter seen in the FRAM results in Fig. 3 may be explained by inaccuracy in the efficiency curve fitted by FRAM as part of the analysis, which was caused by the significant attenuation and resultant uncertainty in the measured gamma peak areas. It is also possible that the uranium enrichment does vary in each drum.

## METHODOLOGY

Measurement of the total fissile content (total plutonium plus U-235) was required to demonstrate compliance with nuclear safety limits for transport and subsequent storage of these drums. Quantification of the activity of any gamma-emitting radionuclides present was also required to comply with transport regulations. BIL Solutions Ltd therefore devised the methodology summarised in this section to calculate the required results.

### U-235 Nuclear Safety Mass

The alternative methodology proposed by BIL Solutions Ltd to calculate the U-235 mass makes use of the passive neutron emission from U-238 combined with a HRGS measurement of the uranium isotopic composition, to determine the U-235 mass.

The passive neutron emission rate from U-238 is relatively small (i.e. the U-238 spontaneous fission neutron emission rate is $1.36 \times 10^{-2}$ n.s$^{-1}$.g$^{-1}$, compared to $1.02 \times 10^{3}$ n.s$^{-1}$.g$^{-1}$ for Pu-240 [4]); however, as reported above, the initial investigative measurements performed on the overweight drums have shown that the passive neutron signals from U-238 are measurable above background. This is because:

- measurements indicate that the uranium is depleted and that there are kilogram quantities present,
- inspection of gamma spectra indicates that the drums do not contain significant quantities of plutonium.

The U-238 mass was determined from the measured passive neutron “Reals” (i.e. coincident) count rate from the Passive Neutron Coincidence Counter (PNCC) measurement performed by the Drum Monitor. Note that the passive neutron “Totals” count rate could also be used for the measurement, which would offer an improved detection limit because of the higher detection efficiency. However, the preliminary measurements indicated that the Reals count rate was measurable above background, and use of the Reals count rate was preferred because it would produce a more accurate U-238 mass. The Totals count rate was found to fluctuate significantly due to drum movements within the area the Drum Monitor is located, whereas the Reals count rate was relatively immune to such variations.

Background measurements are performed periodically by the Drum Monitor (at four hourly intervals), and the lower of the two backgrounds bounding each overweight drum PNCC measurement are used to background correct the Reals count rate.

The Drum Monitor PNCC systems were calibrated using plutonium sources, and the measured Reals count rate is related to a Pu-240 equivalent mass using a calibration curve. In order to utilise this PNCC measurement to determine the U-238 mass, cross-calibration measurements were carried out using similar neutron counting equipment and well-characterised uranium and plutonium standards, to determine a U-238 neutron calibration for the Drum Monitors. This was required to account for the differences in the nuclear decay properties and neutron energy spectra of Pu-240 and U-238. A gram Pu-240 equivalent to gram U-238 conversion factor of $68134 \pm 2542$ was obtained from the experimental measurements, and was applied as an adjustment to the Reals calibration curves used by the Drum Monitors.

The Drum Monitor PNCC measurement includes an Add-A-Source (AAS) measurement using a Cf-252 neutron source to correct for moderation/absorption of the passive neutron signal within the waste matrix. The AAS measurement was calibrated using a wide range of matrix materials, and a concrete matrix is within the current calibration envelope. The matrix correction factor and associated uncertainty determined from the AAS measurement were therefore used in the calculation of the U-238 mass to correct for reductions in detection efficiency caused by neutron interactions with the matrix materials. Note that since the AAS correction factor determined with a Cf-252 source can be applied to Pu-240, it is also applicable to U-238. This is because there are only small differences in the neutron energy spectra from spontaneous fission of U-238 and Pu-240.

In order to calculate the U-235 mass from the U-238 mass, information on the relative abundances of U-235 and U-238 was required. This was primarily obtained using the FRAM software package, although MGA was used to check for consistency with FRAM when successful. Both codes require different format spectra (FRAM = 8k @ 0.125 keV/channel: MGAU = 4k @ 0.075 keV/channel), but the gain can be adjusted offline and the same spectrum analysed with both codes. As discussed earlier, the FRAM results were examined to confirm that they met certain criteria to confirm their acceptability. Similar criteria were also defined for MGAU results, as follows: any minor warning messages reported must not invalidate the results; the reported total peak fit error must be less than 10%; the peak fits must be good; and the uranium isotopic analysis results be reasonable (i.e. consistent with the expected range for depleted uranium).

If both FRAM and MGA were unable to determine the isotopic composition of the uranium, a pessimistic default value of natural uranium was assumed. This was supported by further analysis of the gamma spectrum to confirm enriched uranium was not present.

The U-235 Mass calculation is summarised in Table IV.

**Table IV. Summary of U-235 Mass Calculation**

| **Calculate U-238 Mass** |
|---|
| • Correct Reals count rate for background.<br>• Multiply by U-238 calibration factor and Reals matrix correction factor to determine the U-238 mass.<br>• Calculate associated statistical and systematic uncertainties using standard error propagation formulae. |
| **Calculate Uranium Mass** |
| • Divide U-238 mass by the U-238 abundance (from FRAM, MGAU or Defaults) to determine the total uranium mass.<br>• Calculate associated statistical and systematic uncertainties using standard error propagation formulae. |
| **Calculate U-235 Mass Nuclear Safety Value** |
| • Multiply uranium mass by the U-235 abundance (from FRAM, MGAU or Defaults) to determine the U-235 mass.<br>• Calculate associated statistical, systematic and Total Measurement Uncertainties (TMU) using standard error propagation formulae.<br>• Add the TMU to the U-235 mass at the three sigma confidence level to calculate the U-235 mass Nuclear Safety Value. |

## Total Plutonium Nuclear Safety Mass

Examination of the gamma spectra for the overweight drums indicates that they do not contain significant amounts of plutonium. 34 of the 164 (i.e. 21%) of the drums showed some evidence of plutonium being present, with very small gamma peaks at 208keV, 375keV and 414keV being detected. However, in all cases the neutron count rates were very low (and consistent with background), indicating that there could not be significant masses of plutonium present. It is suspected that the plutonium detected in the gamma measurements is present as trace amounts of contamination on the drums. These drums contain depleted uranium, and the measured passive neutron emission is consistent with there being kilogram quantities of U-238 present. The PNCC measurement performed by the Drum Monitor is calibrated in terms of grams Pu-240 equivalent, so during the automatic assay the measured neutron signal will be attributed to plutonium even if it is not present. The neutron emission rate from U-238 is much lower than for Pu-240, so the neutron emission from kilogram quantities of U-238 will only be reported as a few grams of total plutonium. Note that in the absence of measurable plutonium gamma-rays, a default plutonium isotopic composition was applied (i.e. 98% Pu-239 and 2% Pu-240) to convert the Pu-240 equivalent mass from the PNCC measurement to a total plutonium mass.

As the plutonium masses reported for the overweight drums were relatively low compared to the nuclear safety limits for drum transport and storage, the Total Plutonium Mass reported by the Drum Monitor will be assigned to each drum. Clearly this approach is pessimistic, since the same passive neutron signal is being used to calculate both the U-235 mass and the Total Plutonium Mass, and there is strong evidence to suggest there are negligible amounts of plutonium present in these drums. Using the same neutron count rates to determine the U-235 mass results in a bigger contribution to the total fissile content for these drums, since the calculated U-235 masses are higher than the Total Plutonium masses. However, it is considered appropriate to be conservative as the presence of a few grams of plutonium cannot be discounted due to the high density of the waste.

## Best Estimate Radionuclide Activities

The activities of any gamma-emitting radionuclides present in the waste drum must also be calculated to comply with transport regulations. The radionuclide activity calculations for general PCM waste drums were performed in a similar way to the original U-235 measurement, with characteristic gamma-rays for each nuclide of interest being measured and corrected for attenuation within the waste matrix using the Eu-152 transmission source measurement. The main difference was that Best Estimate nuclide activity results are calculated, with the calibration assuming a uniform (or Volume-Weighted Average (VWA)) distribution within each vertical segment of the HRGS measurement. However, due to the high density of the waste, the overweight drums failed the transmission correction measurement in several segments of the drum, and an alternative means of calculating the radionuclide inventory was necessary.

The Drum Monitor currently utilises the Canberra Genie-2000 library-based peak search algorithm to identify the radionuclides present in the waste drum. The nuclide library used by Genie-2000 is currently not set up to measure Np-237 and Ra-226, both of which are present in the majority of the overweight drums. Rather than modify the Drum Monitor so that the nuclide activity calculations are performed online, the calculations of the radionuclide inventory were performed offline using the active and passive gamma data acquired during the Drum Monitor measurement. The gamma acquisition count times were 100 s passive and 200 s active.

The passive gamma spectra for each segment of the segmented scan were analysed using Genie-2000 to identify the nuclides present. The net peak area count rates for the gamma peaks of interest were extracted as part of this analysis. The active gamma spectra from the transmission source measurement of each corresponding segment were visually inspected to determine the most appropriate method for the calculation of the matrix attenuation correction factor. The method used depended on whether the appropriate gamma peaks from the transmission source were detected together with the characteristics of the nuclides being measured.

The following three methods were identified to determine the nuclide activities. These have been listed in order of preference, with the current transmission source correction algorithm being applied first:

1) Use transmission source measurement.
2) Apply Differential Peak Attenuation (DPA) Analysis.
3) Use an average matrix density estimated from high energy gamma-ray transmission.

Once the corrections for matrix attenuation have been determined using one of these three methods, the calculations are performed using the measured net peak area count rates to determine the Best Estimate activity of each identified nuclide. The results obtained for each segment were then summed over the 16 segments to obtain the total drum activity.

A summary of each method is provided below:

**1) Use transmission source measurement.**

The correction for matrix attenuation used currently in the determination of the Best Estimate nuclide activities uses the transmission source measurement data. The correction factor is determined by linear interpolation in log-log space between the bracketing transmission source energies. If, for example, Cs-137 is detected, which has a principal gamma-ray emission at 662 keV, the measured transmission fractions at 344 keV and 778 keV (the closest two peaks from the Eu-152 source) will be used to calculate the transmission fraction at 662 keV. If the gamma-ray from the nuclide of interest does not have two bracketing transmission peaks (e.g. due to attenuation), the transmission is calculated from the nearest peak using assumed mass attenuation coefficients. The calculated transmission is then checked against a lower default limit of 5%, and if lower than this it is set equal to 5%.

For the overweight dense drums, the transmission was set to the default worst case transmission fraction of 5% for most of the nuclides measured, because of the significant attenuation of the transmission source. The transmission fraction could be significantly worse than 5%, particularly if the gamma-ray from the nuclide of interest is low in energy. An alternative methodology was therefore used to estimate the transmission fraction when the transmission had been set to the default value of 5%. If the transmission peaks either side of the nuclide energy were not detected or the transmission fraction had been set to the default of 5%, the higher energy gamma peaks from the transmission source, if detected, were used to extrapolate the transmission at the required gamma energy. This extrapolation required knowledge of the mass attenuation coefficients, and therefore a concrete matrix was assumed when performing the extrapolation.

If the attenuation is very high then even the highest energy gamma peaks from the transmission source may not be detected during the measurement. The extrapolation from high energies down to the energy of interest required that several of the higher energy peaks from the transmission source were detected, and the accuracy and validity of the extrapolation approach will reduce if fewer peaks are available or if the count rates in these peaks are low. The transmission correction algorithm was therefore not applicable in some cases due to the high density of the waste matrix. If this occured, then one of the alternative methods described below was implemented.

**2) Apply Differential Peak Attenuation (DPA) Analysis.**

If the detected nuclide emits more than one gamma-ray at different energies, then it may be possible to apply DPA analysis to determine and correct for the attenuation of the gamma-ray signal. The principal is straightforward. If the nuclide being measured emits two or more gamma-rays at different energies, then the ratio of the measured gamma peaks ought to be the same as the branching ratios of the gamma-rays, after correction for differences in the intrinsic detection efficiency of the HRGS detector. However, if there is any attenuation of the gamma-ray signal, the difference in attenuation with gamma energy will bias the measured gamma peak ratios. The measured difference in the gamma peak ratios can therefore be used to calculate the attenuation of the gamma-ray signal. Clearly this technique works best for nuclides that emit several intense gamma-rays across a wide energy range, with a good example being Eu-152. However, the technique can be applied to nuclides such as U-235, U-238, Ra-226 and Np-237, which emit multiple gamma-rays sufficiently spaced in energy and are known to be present in the overweight drums.

This technique is a well-proven method of determining matrix attenuation effects, which will be accurate if the activity of the nuclide is high and the true activity distribution is close to a uniform distribution. If, however, the activity is not uniformly distributed, the DPA analysis will be biased towards the activity that is closer to the detector. This effect will be worse at higher matrix densities and at lower gamma energies. This approach was therefore considered valid for the calculation of the Best Estimate nuclide activities, because an assumption of uniformly distributed activity is acceptable. Clearly this method would not be appropriate for a Nuclear Safety Value calculation, unless conclusive evidence can be obtained to support the assumption of a uniform distribution of activity.

The DPA analysis may prove to be more accurate than the transmission source correction (Method 1 above), since it is a more direct measurement of the attenuation using gamma-rays emitted by the nuclide. However, the transmission source correction was used in preference, if possible, since the DPA analysis is a significant deviation from the normal operation of the instrument.

**3) Use an average matrix density estimated from high energy gamma-ray transmission.**

In cases when the high waste density prevents use of the transmission source measurement and the DPA analysis is not possible, the alternative approach was to estimate the average waste matrix density and use this to determine the attenuation. The estimation of the matrix density in each segment was based on the drum weight and the fill height, which was determined by inspection of the segmented scan gamma transmission data.

Examination of the example U-235 186 keV transmission correction factor data in Table I indicates that the drums are filled up to Segment 11 or 12 and that there is some variation in the density in the segments beneath. Although the automated measurement was unable to calculate the transmission in several segments, inspection of the active gamma spectra indicates that some of the highest energy gamma peaks from the transmission source were detected. These higher energy peaks were therefore used to determine the relative density of the segments and also to check that the calculated density of the segment was correct. The drum weight was attributed to each of the segments according to the relative transmission fractions of the higher energy gamma-rays, to estimate the density in each segment. The measured transmission fractions were then used to confirm whether the estimated density is correct, assuming a concrete matrix material.

Once the density of each segment had been estimated, the attenuation corrections for a uniform distribution of activity were calculated for each of the detected nuclides.

## TOTAL FISSILE MASS RESULTS

Fissile Mass (total plutonium plus U-235) results and radionuclide activity were successfully calculated for all 164 overweight drums using the methodology defined in the previous section. Fig. 4 shows the fissile mass results for a sample of 25 drums (note these are the same drums considered in Fig. 3). The intermediate and final results for 5 of these drums are also summarised in Table V.

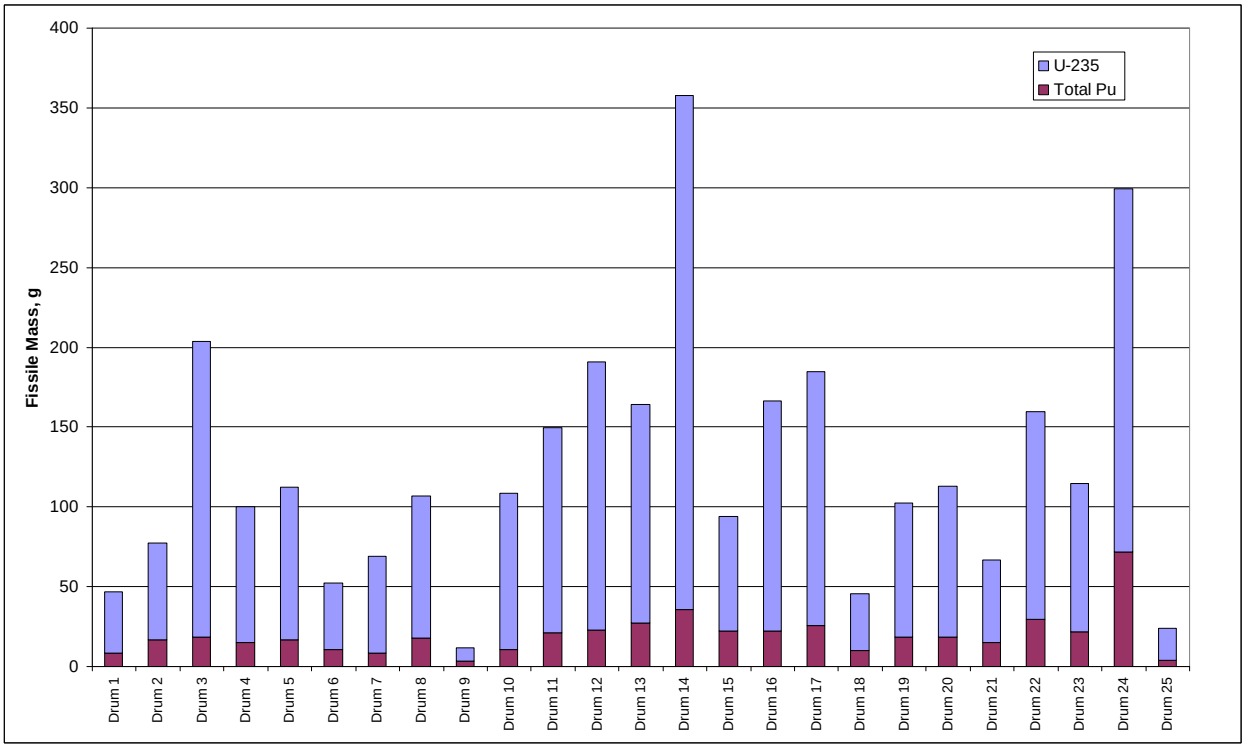


**Fig. 4. Total Nuclear Safety Fissile Mass Results (i.e. Total Plutonium + U-235) for a Sample of Overweight Drums.**

**Table V. Intermediate and Total Fissile Mass Results for 5 Drums.**

| Drum Number | Net Reals Neutron Count Rate ($cs^{-1}$) | U-238 Mass (g) | %U-235 | U-235 Mass (g) | Nuclear Safety Values | | |
|---|---|---|---|---|---|---|---|
| | | | | | Plutonium Mass (g) | U-235 Mass (g) | Total Fissile Mass (g) |
| 3 | 3.81 ± 0.10[a] | 16500.25 ± 750.26[a] 2072.91[b] | 0.49 ± 0.16[a] | 81.74 ± 27.16[a] 21.14[b] | 18.36 | 184.99 | 203.35 |
| 6 | 1.60 ± 0.08[a] | 7241.70 ± 456.48[a] 1487.85[b] | 0.28 ± 0.05[a] | 20.18 ± 3.51[a] 6.16[b] | 10.75 | 41.46 | 52.21 |
| 9 | 0.13 ± 0.06[a] | 612.05 ± 263.61[a] 180.94[b] | 0.43 ± 0.15[a] | 2.64 ± 1.45[a] 0.98[b] | 3.54 | 7.90 | 11.45 |
| 11 | 1.33 ± 0.11[a] | 6902.23 ± 618.74[a] 3951.40[b] | 0.64 ± 0.03[a] | 44.71 ± 4.43[a] 27.52[b] | 21.42 | 128.32 | 149.74 |
| 14 | 9.36 ± 0.19[a] | 37937.89 ± 1608.65[a] 2624.74[b] | 0.44 ± 0.09[a] | 166.49 ± 33.61[a] 39.37[b] | 35.79 | 321.78 | 357.57 |

[a] 1σ Statistical Error

[b] 1σ Systematic Error

The results presented in Fig. 4 and Table V for a sample of the overweight drums are typical of the overall population. The calculated U-235 Nuclear Safety Value masses for all 164 drums ranged from 2.1 g to 846.3 g, with 87% of the results being less than 200 g. FRAM was successful for 65% of the overweight drums, MGAU for 24% and a default uranium isotopic composition was applied to 30%.

It is worth noting that two of the drums considered in Fig. 4, i.e. Drum 9 and Drum 25, showed some evidence for plutonium being present because small gamma peaks characteristic of plutonium were detected. However, the measured neutron count rates for these two drums were the lowest of those in the sample, indicating that there could not be a significant mass of plutonium present. The same was true for the other drums in which small plutonium gamma peaks were detected, giving further credence to the theory that the plutonium was present in the background or as trace amounts of contamination.

Although not reported here, radionuclide activity results were also successfully calculated for all 164 overweight drums using the methodology outlined in the previous section. The uranium activities calculated from the HRGS measurement can therefore be compared against those obtained using the neutron measurement and uranium isotopic analysis. The activities obtained from the gamma measurements were generally significantly lower than those obtained from the neutron measurements. This is because of the reduced accuracy of the HRGS measurements in the high density waste. If the transmission correction approach was used to correct for matrix attenuation, the accuracy will be highly dependent on the assumed uranium distribution. Similarly, if the DPA method was used, the correction will be biased towards any uranium located close to the edge of the drum, and will not adequately correct for uranium closer to the centre of the drum. This may result in an underestimation of the matrix attenuation correction factor and hence uranium activity, depending on the actual distribution of uranium.

## CONCLUSIONS

A new method has been developed by BIL Solutions Ltd to determine the U-235 content of a population of 164 overweight dense drums containing uranium, and significant amounts of Np-237 and Ra-226. The methodology is based upon measurement of the passive neutron emission from U-238, combined with a measurement of the uranium enrichment to determine the U-235 mass. This methodology was subject to independent peer review by our regulators and was given their approval for use. Fissile mass Nuclear Safety Value results were then successfully calculated for all 164 overweight drums, to demonstrate compliance with safety limits for transport and storage. It is worth noting that now all the drums have been measured and confirmed to contain depleted or natural uranium, the criticality hazard is negligible.

The new method to determine the U-235 mass significantly more accurate than the previous approach, which was based on measurement of the 186 keV gamma-ray with transmission correction. This original method required assumptions to be made regarding the distribution of the uranium within the drum, which severely limits the accuracy due to the very high waste density. The new methodology requires no assumptions regarding the distribution of uranium, but does assume homogeneity of uranium enrichment. Inaccurate results could therefore be obtained if uranium with different enrichments were heterogeneously distributed within the same drum. However, this assumption was considered acceptable since no evidence was found for mixtures of uranium enrichments, despite numerous drums being measured more than once in different orientations without rotation. The new method also assumes that the neutrons detected in the PNCC measurement originate from U-238. If this was not true and some plutonium was present, the U-235 mass would be greatly overestimated because of the much higher neutron emission rates from plutonium. However, although some drums did show small gamma peaks characteristic of plutonium, the neutron count rates were low in all cases, indicating that negligible amounts of plutonium were present in these drums.

Best Estimate radionuclide activity results were also successfully calculated for the population of overweight dense drums. Comparison of the uranium activity results obtained from these HRGS measurements against those obtained from the PNCC and uranium enrichment measurements did show significant differences, reflecting the poorer accuracy of the HRGS based measurements in such dense waste matrices, even when the DPA method was used to correct for matrix attenuation.

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