Use of a Shielded High Resolution Gamma Spectrometry System to Segregate LLW from Contact Handleable ILW Containing Plutonium – 13046

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ABSTRACT

Dounreay Site Restoration Limited (DSRL) have a number of drums of solid waste that may contain Plutonium Contaminated Material. These are currently categorised as Contact Handleable Intermediate Level Waste (CHILW). A significant fraction of these drums potentially contain waste that is in the Low Level Waste (LLW) category. A CANBERRA Q2 shielded high resolution gamma spectrometry system is being used to quantify the total activity of drums that are potentially in the LLW category in order to segregate those that do contain LLW from CHILW drums and thus to minimise the total volume of waste in the higher category. Am-241 is being used as an indicator of the presence of plutonium in the waste from its strong 59.54keV gamma-ray ; a knowledge of the different waste streams from which the material originates allows a pessimistic waste 'fingerprint' to be used in order to determine an upper limit to the activities of the weak and non-gamma-emitting plutonium and associated radionuclides. This paper describes the main features of the high resolution gamma spectrometry system being used by DSRL to perform the segregation of CHILW and LLW and how it was configured and calibrated using the CANBERRA In-Situ Object Counting System (ISOCS). It also describes how potential LLW drums are selected for assay and how the system uses the existing waste stream fingerprint information to determine a reliable upper limit for the total activity present in each measured drum. Results from the initial on-site commissioning trials and the first measurements of waste drums using the new monitor are presented.

INTRODUCTION

DSRL have a significant number of drums containing potentially Plutonium Contaminated Material (PCM). These are currently categorised as Contact-Handleable Intermediate Level Waste (CHILW). A project is now underway to assess the radionuclide inventory of each drum and, where possible, re-categorise drums into the lower category of Low Level Waste (LLW) and develop the best possible approach for sentencing LLW and CHILW drums with activity data which is as accurate as possible. This will reduce waste consignment costs for DSRL and reduce the pressures due to limited capacity to accommodate future CHILW waste arisings.

The key requirement in terms of robustly segregating LLW from CHILW, is to demonstrate that the total alpha activity in a candidate drum is less than 0.1 $GBq.tonne^{-1}$. This requires an accurate assessment of the total alpha activity concentration, with due consideration of the Total Measurement Uncertainty (TMU) according to nationally accepted best practice guidelines [1]. This assessment may, depending on the way in which the results are being used, consist of making “worst case” assumptions regarding the calibration conditions.

The drums are legacy waste drums approximately 200 litres in capacity with a variety of dimensions and waste contents (soft, metal etc). The waste also spans a variety of densities (0.01 to 1 $g.cm^{-3}$).

A study was conducted to develop an optimised methodology for performing the segregation measurements. The use of both existing assay systems, and new systems, was considered. In developing such a methodology, it was necessary to consider the following factors:

- Reliability of segregation between CHILW and LLW (i.e. accuracy at the LLW / CHILW boundary).
- Accuracy of activity sentencing for the full range of stored CHILW and LLW.
- Minimum Detectable Activity (of total alpha activity) with regard to the LLW upper boundary.
- Measurement cycle time.
- Whether it is possible to use existing systems on site and their availability and accessibility.
- Use of proven technology (tried and tested techniques).

Q2 System

Following a review of the above factors, DSRL chose to purchase a Q2 drum monitor. This incorporates a high sensitivity shielded gamma spectrometry system, providing the best possible sensitivity for Pu measurements. The Q2 system incorporates 4 inches of “low background” steel shielding on all sides and 3 detectors viewing a 200 litre drum in segments.

The standard Q2 system (see Figure 1) has sufficient sensitivity that CHILW / LLW segregation can be performed with reliable policing of the 0.1 $GBq.tonne^{-1}$ total alpha LLW upper threshold, even when "worst case" assumptions have to be made regarding the spatial distribution of the activity within the waste matrix.

Figure 1. Q2 shielded High Resolution Gamma Spectrometry (HRGS) system.

Spectrometry

The difficulty associated with measuring Pu waste by gamma spectrometry is due to the low abundance of many of the plutonium lines. The energies and abundances of the key gamma emissions from Pu-239, which typically represents a major contribution to the total alpha activity, are listed in Table I.

Table I. The gamma energies and abundances of three of the most abundant Pu-239 lines [2]

Energy (keV)	Abundance (%)
129.296	0.00631
375.054	0.001554
413.713	0.001466

This means that the Minimum Detectable Activity (MDA) can be very high resulting in the false consignment of a large number of drums as CHILW without any Pu activity actually being measured in those drums. However, Am-241 is normally associated with Pu waste as a daughter product of Pu-241. As this is legacy waste there is a certain amount of in-growth so the minimum expected percentage of Am-241 to total Pu is 2.14%. These "fingerprint" percentages are either 3.13% in one fingerprint or 34.00% in another fingerprint.

The most abundant line associated with Am-241 is the 59.54 keV line with an abundance of 35.9% [2]. Using Am-241 will effectively reduce the MDA on the same type of waste to less than a tenth of that from the Pu-239 gamma lines.

The software associated with the Q2 also allows for nuclide identification and reporting of energy lines across the gamma spectrum so the total activity of the package can be taken into account. This also allows for powerful expert review, diagnostics and spectral analysis of waste drums which have unusual properties.

Quantitative Analysis

In order to quantify the Am-241 activity from the measured net full-energy peak area of the 59.54 keV line, the combined efficiency of the detectors for that energy needs be determined. Other gamma energies in the spectrum will have different efficiencies. With traditional calibration techniques relying upon radioisotope sources, a range of peaks covering the entire energy range of interest need to be observed from a known, multi-energy certified source, in order to provide a calibration across this energy range.

There are a number of methods to conduct an efficiency calibration, the most common is to measure simulated waste drums and to interpolate the results in order to obtain the calibration functions for each individual type of legacy drum. This efficiency calibration requires the use of simulated matrices in appropriate container types with appropriate densities and reference radioisotope sources mimicking the expected distributions of the legacy waste. The problem in this case for the Dounreay application, is the cost of achieving this for all 4 main waste types, 2 container types (with and without liners) and for a range of densities (at least 4 densities are considered to be appropriate between 0 and $1 g.cm^{-3}$) whilst ensuring that the properties of the calibration drums are still representative of the real waste streams. The total number of calibration measurements required would be at least 32 which would be time-consuming and expensive, as would designing and creating the simulated matrices and obtaining certified sources. As a result this can become an expensive option and also leading to delays in the site waste management programme

An alternative is to use computed efficiencies. This can be done using the In Situ Object Calibration Software (ISOCS) [3]. The ISOCS calibration method is a convenient tool for calibrating the detector efficiency as a function of energy for a wide variety of source geometries and activity distributions. The ISOCS method consists of a detector, which has been characterized at the factory in order to determine its intrinsic efficiency and the user input of source and detector geometry data. The ISOCS software uses these to produce the efficiency calibration. The large number of efficiency calibrations required can then be run in a relatively short space of time.

The system is to be used to segregate stored CHILW from LLW at Dounreay. Following an investigation of the development of a radiometric approach for sentencing, it was found that there are likely to be four main descriptions of waste in the stored drums. The drums themselves also vary in their construction with some having different clamp types for the lids, which makes a negligible difference to the radiometric assay and others having a Alcathene liner, which will make a difference to the detector response. As a result of this, eight container types have been created with the four main waste contents and with or without a liner as seen in Table II.

Table II Container types created in NDA2000 and modelled in ISOCS geometry composer

Drum Name	Composition description	ISOCS materials
Soft	50% paper, 50% plastics	50% cellulos, 50% lpolyeth
Soft Metal	50% mild steel, 25% paper, 25% plastics	50% csteel, 25% cellulose, 25% lpolyeth
Rubble Soil	50% concrete, 50% soil	50% concrete, 50% dirt1
Metal	100% mild steel	100% csteel
Liner Soft	50% paper, 50% plastics	50% cellulos, 50% lpolyeth
Liner Soft Metal	50% mild steel, 25% paper, 25% plastics	50% csteel, 25% cellulose, 25% lpolyeth
Liner Rubble Soil	50% concrete, 50% soil	50% concrete, 50% dirt1
Liner Metal	100% mild steel	100% csteel

Figure 2 shows an example of a combined, multi-efficiency, multi-density calibration for soft waste.

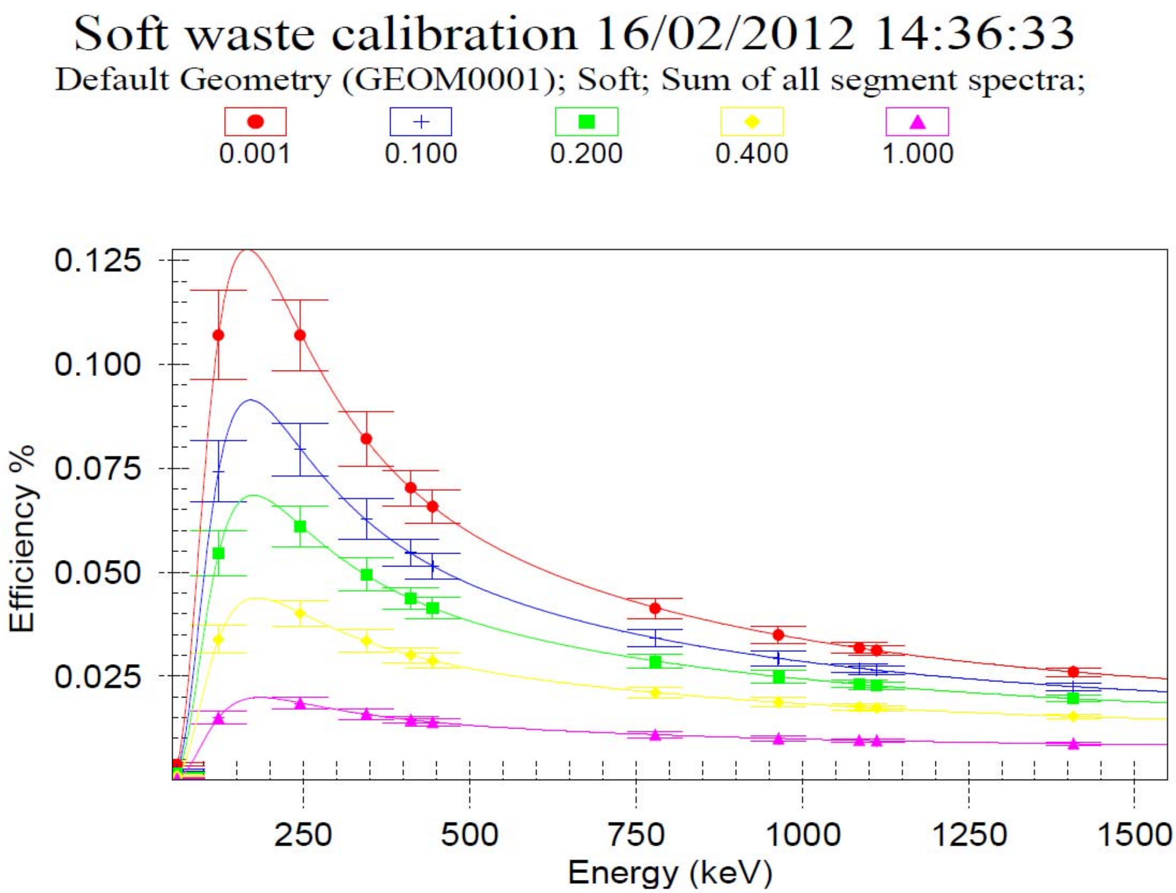

Figure 2. Efficiency calibration for Soft waste, expressed for different matrix densities ($g.cm^3$)

Uncertainties

There are a number of random uncertainties, arising from counting statistics and the uncertainties on the ISOCS calibrations (see error bars on Figure 2) which are easily accounted for and follow a standard Gaussian distribution. These factors are included in the reports produced by the Q2 software.

This system has been configured to measure the ^{241}Am activity and to use that to derive the activity of other nuclides associated with the plutonium and related isotopes present in CHILW. The Total Measurement Uncertainty (TMU) in this case comprises:

- random uncertainties (described above), assumed to follow a Gaussian distribution and
- systematic uncertainties from the lack of knowledge of the activity or matrix distribution – which are assumed to be the main contributors to the systematic uncertainties

The uncertainties associated with the activity distribution as a function of detector response may not follow a Gaussian pattern and are skewed due to effects of attenuation and inverse square law [4]. Therefore the calculation is split into an increased and reduced response. As the worst case scenario is of more interest, the minimum response was calculated using ISOCS Uncertainty Estimator (IUE). IUE was used to calculate the efficiency of the combined three detectors with a single source containing all of the activity in the drum located at random radial and vertical position. The position which produces the lowest efficiency in comparison to the Volume Weight Average (VWA) is then used as R_{min} for that density. This IUE calculation is then repeated for each density. The R_{min} is then plotted versus density and a quadratic fitted and the whole process is repeated for each matrix and with and without liners.

The upper limit values across the density range are calculated using the following equation which is based on the NDA2000 algorithms:

$$R_{min} = R_{min1} + R_{min2}.D + R_{min3}.D^2 \qquad \text{(Eq. 1)}$$

Where:

R_{min} is the minimum response of the system to the same activity, expressed as a ratio to the volume weighted average response

D is the average density of the waste in the drum.

The quantities R_{mini} (where i=1,2,3) are the empirically determined coefficients for the offset, the slope, and the quadrature terms of the equation. IUE has been used to determine these values.

The systematic uncertainty is then calculated (equation 2) as an upper bounding limit. As the distribution is not Gaussian, no standard deviations are given.

$$Syst.Uncert. = \left(\frac{activity}{R_{min}}\right) - activity \qquad \text{(Eq. 2)}$$

From this analysis, a maximum density (*MaxD)* was determined corresponding to the value required to ensure the system software does not try to give results for containers where the density is above the values at which the upper limit can be calculated for.

The reason for the limit MaxD is to inhibit the assay of the drums above certain densities where the uncertainties can no longer be calculated accurately. The quadratic R_{min} equation is an approximation which becomes incorrect for high density drums especially those containing metals. Figure 3 and Figure 4 show the comparison as the percentage of metal in the waste is increased from 0% to 50%. In these examples MaxD is set at 1 g.cm^{-3} for soft waste and 0.4 g.cm^{-3} for soft and metallic waste.

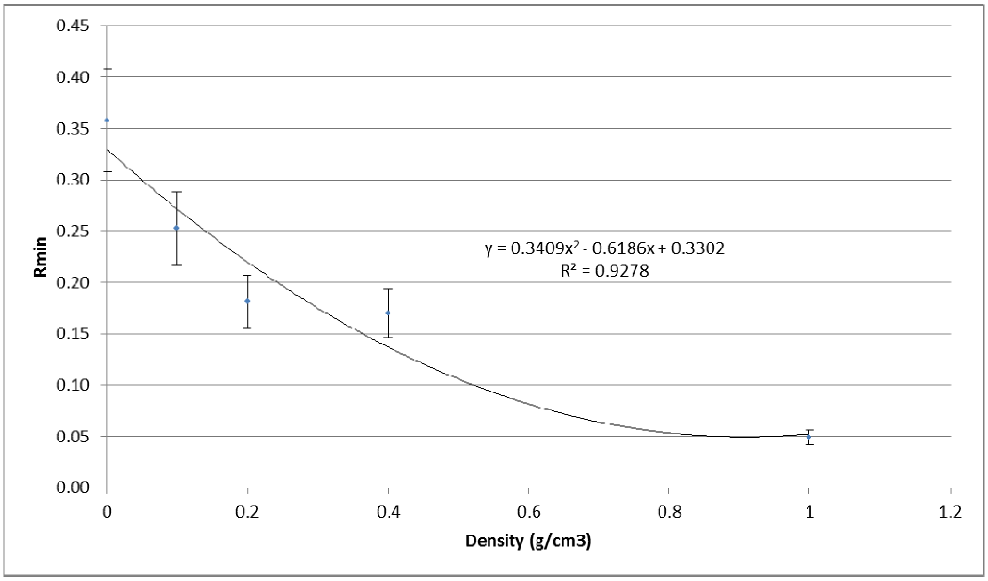

Figure 3. Upper limit minimum response as a function density for soft waste drums. The quadratic fit is accurate up to 1 g/cm^3

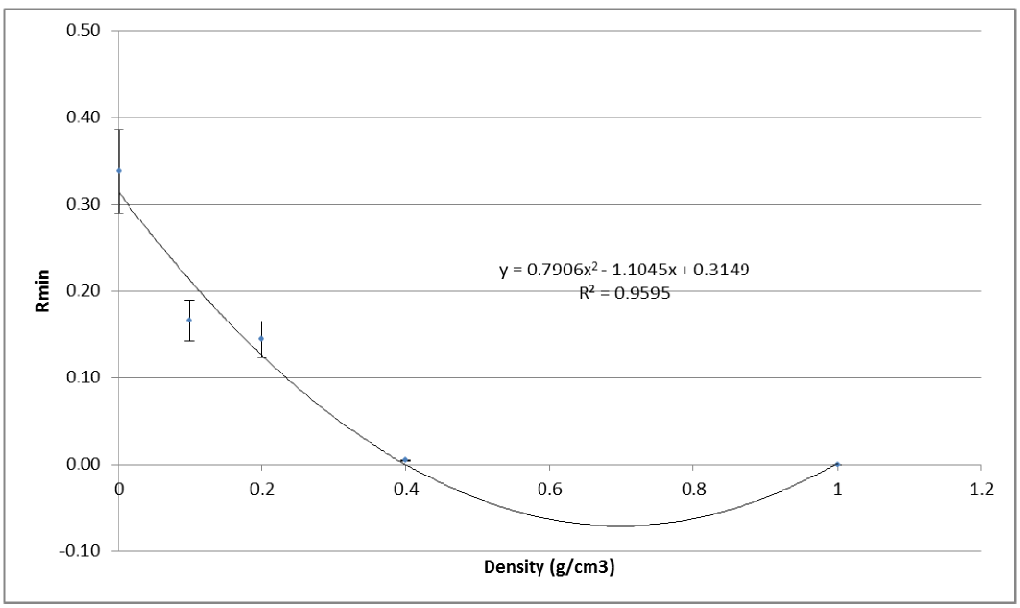

Figure 4. Upper limit minimum response as a function density for drums containing soft and metallic waste. The quadratic fit is accurate up to 0.4 g/cm^3

RESULTS OF THE COMMISSIONING TRIALS AT DOUNREAY

Commissioning trials, completed at Dounreay in November 2012, we carried out on a subset of the total number of drums which will be put through the Q2. The drums selected for the trials had been chosen because, from a review of their contents, were considered likely to be in the LLW category.

Table III summarises the results from those drum items which were identified as LLW by the Q2. They all contain soft waste and are of low density. Some have Am-241 identified in the drums and in others only MDAs are reported (indicated by the “yes” in the table).

Table III. Drums reported as containing less than 0.1 $GBq.tonne^{-1}$ alpha.

Density ($g.cm^{-3}$)	Waste contents	Gross Weight (kg)	Am-241 MDA?	Upper limit activity $GBq.t^{-1}$	Below 0.1 $GBq.t^{-1}$?
0.062	Liner Soft	48.50	Yes	0.05	pass
0.073	Liner Soft	50.60	Yes	0.04	pass
0.078	Liner Soft	51.70	No	0.08	pass
0.081	Liner Soft	52.30	No	0.01	pass
0.092	Liner Soft	54.40	Yes	0.04	pass
0.096	Liner Soft	55.30	Yes	0.04	pass
0.098	Liner Soft	55.50	Yes	0.04	pass
0.099	Liner Soft	55.80	Yes	0.04	pass
0.115	Liner Soft	59.10	No	0.03	pass
0.127	Liner Soft	61.50	Yes	0.04	pass
0.132	Liner Soft	62.50	Yes	0.05	pass
0.132	Liner Soft	62.40	Yes	0.05	pass
0.153	Liner Soft	66.60	Yes	0.06	pass
0.155	Liner Soft	66.90	Yes	0.05	pass
0.17	Liner Soft	70.00	No	0.09	pass
0.175	Soft	55.70	Yes	0.06	pass
0.199	Liner Soft	75.80	Yes	0.07	pass
0.405	Liner Soft	116.90	No	0.06	pass
0.606	Liner Soft	157.10	No	0.07	pass

Table IV shows the drums where the Upper limit result from the Q2 was above the 0.1 $GBq.tonne^{-1}$threshold. Note that for a number of drums, the upper limit cannot be calculated as the drum weight was above the limit applied when calculating R_{min}.

All of these drums are of a variety of weights and waste contents but the common factor is that Am-241 has been reported in all of them.

Table IV. Drums reported as containing greater than 0.1 GBq.tonne^{-1} alpha.

Density (g.cm^{-3})	Waste contents	Gross Weight (kg)	Am241 MDA?	Upper limit activity GBq.t^{-1}	Below 0.1 GBq.t^{-1}?
0.063	Liner Metal	48.70	No	0.33	fail
0.07	Liner Soft Metal	50.00	No	1.13	fail
0.079	Liner Soft	51.80	No	0.35	fail
0.081	Liner Soft	52.20	No	0.36	fail
0.081	Liner Soft	52.20	No	0.13	fail
0.087	Liner Soft	53.50	No	0.17	fail
0.089	Liner Soft Metal	53.80	No	0.11	fail
0.095	Liner Soft	55.00	No	1.95	fail
0.096	Liner Soft	55.30	No	0.11	fail
0.102	Liner Soft	56.50	No	0.14	fail
0.109	Liner Soft	57.80	No	0.48	fail
0.109	Liner Metal	57.80	No	2.57	fail
0.11	Liner Soft Metal	58.10	No	1.46	fail
0.117	Liner Metal	59.30	No	0.16	fail
0.123	Liner Soft	60.70	No	0.19	fail
0.126	Liner Soft Metal	61.30	No	0.34	fail
0.128	Liner Soft Metal	61.70	No	0.29	fail
0.148	Liner Soft Metal	65.70	No	1.38	fail
0.156	Liner Soft	67.20	No	0.13	fail
0.158	Soft	52.20	No	0.21	fail
0.176	Liner Soft	71.20	No	0.49	fail
0.195	Soft Metal	59.80	No	0.45	fail
0.228	Liner Soft Metal	81.60	No	2.54	fail
0.233	Liner Rubble Soil	82.60	No	0.62	fail
0.233	Liner Metal	82.60	No	2.59	fail
0.253	Soft Metal	72.20	No	13.17	fail
0.3	Soft Metal	82.10	No	1.49	fail
0.301	Soft Metal	82.30	No	1.49	fail
0.305	Metal	83.10	No	limit	fail
0.313	Soft Metal	84.80	No	1.80	fail
0.387	Liner Soft	113.30	No	0.13	fail
0.408	Liner Soft Metal	117.60	No	limit	fail
0.455	Metal	114.60	No	limit	fail
0.456	Metal	114.80	No	limit	fail
0.488	Liner Rubble Soil	133.60	No	5.00	fail
0.559	Liner Soft	147.80	No	3.72	fail

Density ($g.cm^{-3}$)	Waste contents	Gross Weight (kg)	Am241 MDA?	Upper limit activity $GBq.t^{-1}$	Below 0.1 $GBq.t^{-1}$?
0.604	Liner Soft Metal	156.80	No	limit	fail
0.605	Liner Metal	157.00	No	limit	fail
0.653	Liner Soft	166.70	No	8.88	fail
0.658	Metal	157.20	No	limit	fail
0.66	Metal	157.60	No	limit	fail
0.661	Liner Soft	168.30	No	9.03	fail
0.72	Liner Soft	180.00	No	0.30	fail

DISCUSSION

It can be seen from Table III and Table IV that the majority of those drums measured in the trial have been sentenced as CHILW (69%). Although this may not desirable from the customer perspective, it is not surprising since the particular subset of drums used in the trials is generally heavier (greater density) than the average of the legacy waste inventory and measurement of Am-241 in metal wastes poses some difficulties as the attenuation coefficient is higher at 59.5 keV than for example plastics, as Table V shows.

Table V. Mass attenuation coefficients of Iron and Polythene at 60 keV [5].

Material	μ/ρ ($cm^2.g^{-1}$)
Iron	1.205E+00
Polyethylene	1.970E-01

It is very difficult to sentence waste containing metals based on the Am-241 signal and this effect was observed in the preliminary calculations (see Figure 5).

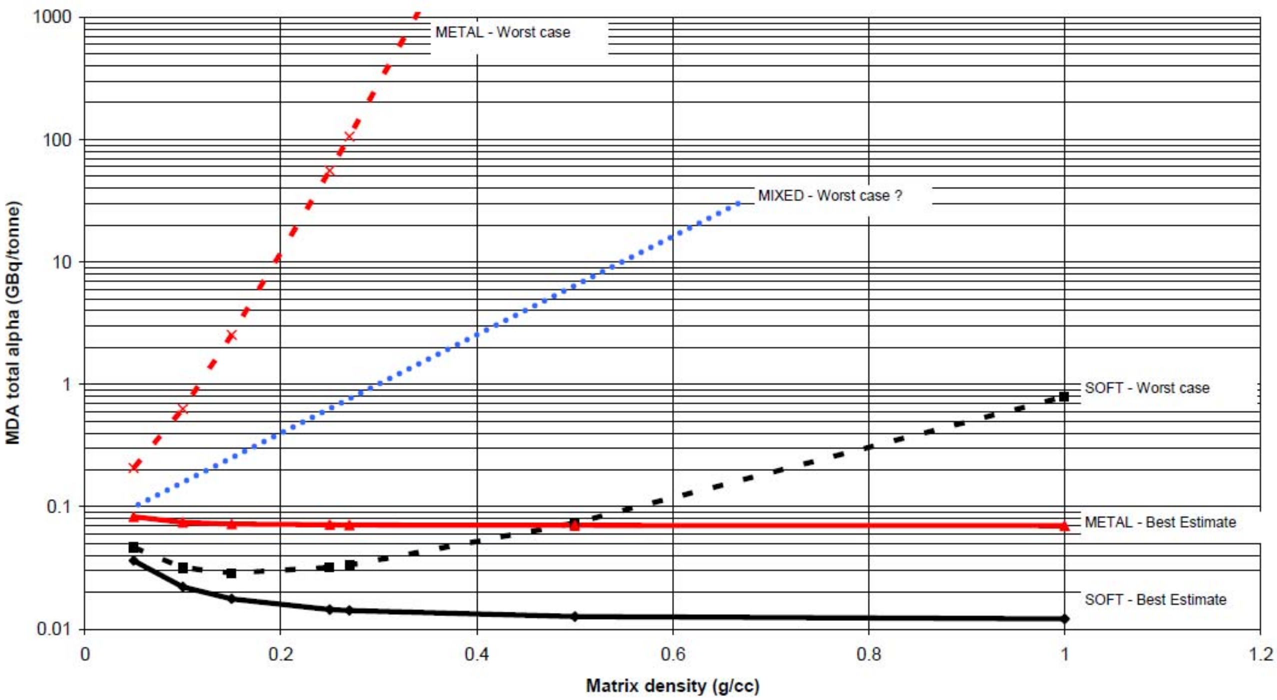

Figure 5. Predicted performance of a single ISOCS detector with no shielding.

CONCLUSION

It can be seen from these results and discussions that the Q2 system performs as expected. They confirm that this measurement technique has the potential to provide large cost savings for DSRL due to providing the ability to downgrade a significant number of drums from CHILW to LLW, with the associated reduction in storage and disposal costs for DSRL. Further work to analyse the potential activity distribution at the worst case (under conditions in which there is a measureable

Am-241 gamma signal) may allow for improved accuracy and therefore may lead to more drums being sentenced as LLW and thus providing additional cost savings.

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