Developments in Very Low Level Waste/Exempt Waste Assay at AWE - 12000

T.J.Miller
AWE, Aldermaston,
Reading, Berkshire,
England, RG7 4PR
Tel: 0118 982 7449
Fax: 0118 982 5796
e-mail: Timothy.Miller@awe.co.uk

ABSTRACT

Portable High Resolution Gamma Spectrometry (HRGS) has been developed, for Very Low Level Waste (VLLW) and Exempt Waste (EW) assay at AWE, in order to meet the latest reduced clearance levels of < 1 Bq/g (or Bq/cm2) for uranium (U) contaminated wastes and < 0.15 Bq/g (or Bq/cm2) for plutonium (Pu) wastes.

Studies have focused on a 10 kg bag of low density soft waste monitored either as a rotating cylinder, contained within a shortened plastic drum liner, or as a contained disk monitored on each broad side. Liquid and surface contaminated metal wastes have also been studied.

It was established that monitoring the disk gave the best detection levels, but uncertainties rose more sharply, compared to the cylinder, as detector offset was reduced. Exempt detection levels were readily achieved for all U compositions encountered at AWE and for most Pu compositions (via Am-241 measurement). However, performance will need to be enhanced for those Pu compositions with relatively high Pu/Am-241 activity ratios. Recommendations are made for further developments to enhance the performance of this technique so that exempt clearance can be achieved for all Pu compositions encountered.

INTRODUCTION

Exempt clearance levels, in the UK, have recently reduced from < 11.1 to < 1 Bq/g for U wastes and from < 0.4 to < 0.15 Bq/g for Pu wastes (1 Bq/g Pu-241; 0.1 Bq/g Pu alpha emitters). Table 1 summarises the latest activity ranges, for each waste category, together with the associated disposal routes and costs.

It can be seen that VLLW categorization avoids relatively costly LLW disposal charges and utilization of limited space at the UK national LLW repository at Drigg. The lower EW categorization gives a relatively smaller cost saving compared to VLLW.

Hence, the principal objective of this work was to develop portable HRGS for the best detection levels and lowest measurement uncertainties in low density soft wastes generated at AWE. Studies have focused on a typical 10 kg bag of waste that can be conveniently contained in a reproducible counting geometry by placement inside a shortened 200 l plastic waste drum liner. This was monitored as a rotating cylinder and as a disk counted on each broad side in order to determine the counting geometry with the best combination of low detection levels and uncertainties. Liquid and metal wastes have also been studied.

Table 1 Activity ranges, disposal routes and costs for the main radioactive contaminants and waste categories encountered at AWE

Category	Activity range (Bq g^{-1} or cm^{-2})	Disposal route	Disposal cost (£ per 200l drum)
ILW	> 4,000 Pu, U > 12,000 beta	Indefinite storage at AWE until a national ILW repository is available	40,000
LLW	< 4,000 U < 12,000 beta	LLW repository at Drigg	250
LLWD	< 100 Pu	LLW repository at Drigg	250
VLLW	< 4 Pu, U	As authorised	50
EW	< 1 U < 0.15 Pu	As AWE policy dictates	20

MEASUREMENTS ON WASTE CYLINDER

The plastic drum liner was cut to 50 cm in height and had an internal diameter of 55 cm and external diameter of 56.4 cm. Soft waste was represented by 11 kg of paper rolls, with a fill height of 38 cm, giving a typical soft waste bulk density of 0.12 g/cc. The cylinder was placed on a rotating turntable and a general purpose, flush collimated, HRGS detector (HPGe, N-type, crystal: 6.14 cm diameter x 8 cm thick) located at 10, 20, and 25 cm offsets from the centre/middle of the shielded drum liner (figure 1). Figure 2 shows a schematic of the counting geometry and the extreme activity locations where the detector would over or under respond compared to a uniform distribution of activity throughout the waste matrix.

A traceable Am-241 source (259 kBq encapsulated in thin plastic) was placed inside the waste material and the detector response (cps/Bq) measured at four axial locations (0, 7.9, 15.7 and 23.6 cm off the central cylinder axis) and at four heights (6.3, 18.8, 31.3 and 43.8 cm up from the cylinder base) in addition to the extreme locations shown in figure 2. The purpose of measuring the extreme locations was to quantify the maximum and minimum response, in a single bag, for the AWE waste management group. However, it is recognized that the

ultimate disposition will be a group of bags. Therefore, while each bag might have an extreme, the group value will be the average of all the bags.

Each detector response factor (other than those for the extremes marked in figure 2) was weighted, according to the volume element represented by the source position, in order to derive the detector response for uniform distribution of Am-241. This was plotted against detector offset, together with the maximum and minimum detector response, in figure 3. The data points were fitted using a polynomial (poly) trend-line. As the offset was reduced the detector response increased, but the range between over-response and under-response also increased. Spectral Nondestructive Assay Platform (SNAP) software was used to generate comparative uniform response factors that were similar to those measured (table 2). The performance of SNAP has been verified against a range of National Physical Laboratory (NPL) waste package standards (1).

Figure 1 Counting geometry for cylinder

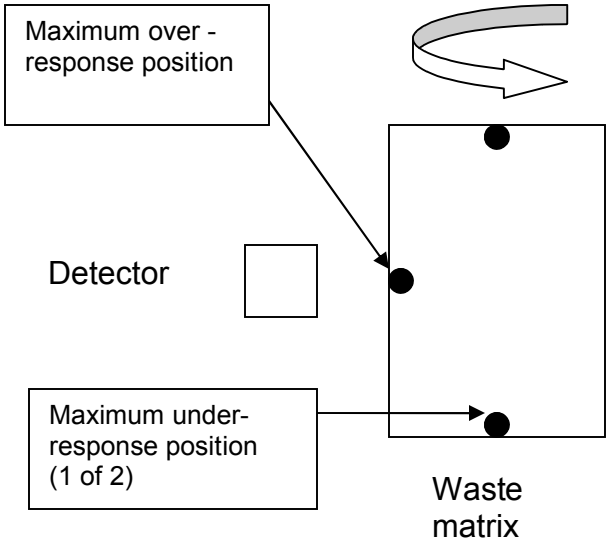

Figure 2 Counting geometry with extreme source locations

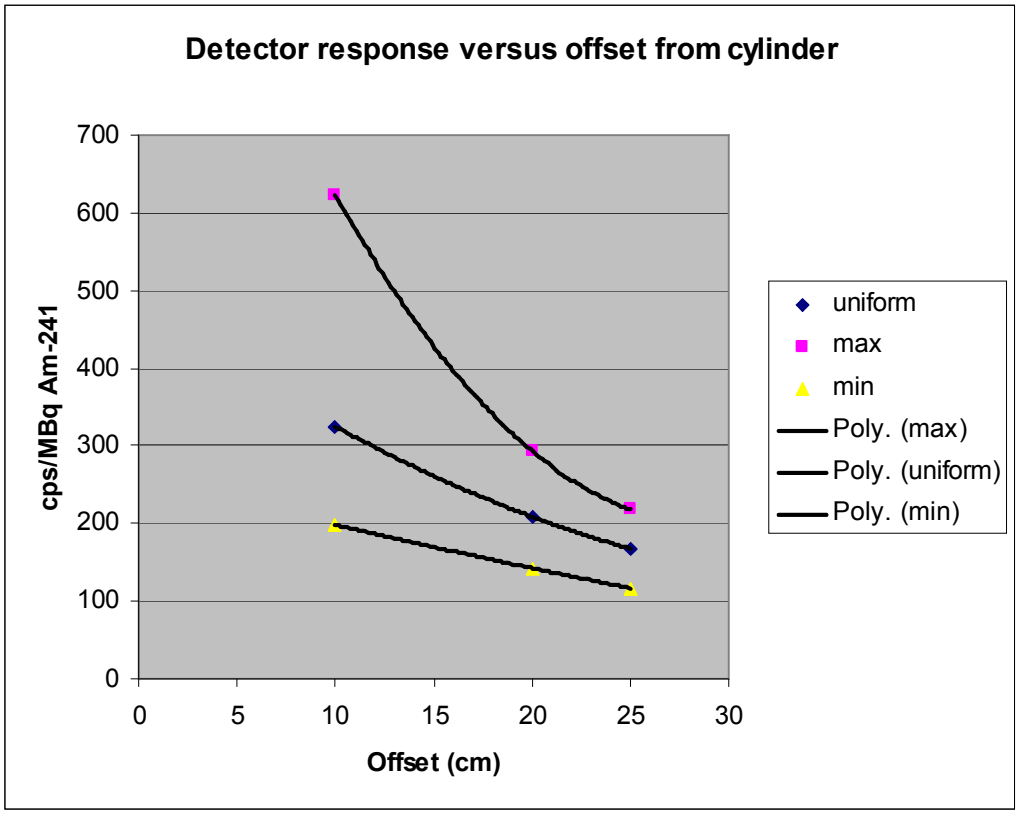

Figure 3 Detector response factors for cylinder

Table 2 Comparative uniform response factors (cps/MBq) for cylinder

Detector offset (cm)	Nuclide	keV	Measured	SNAP
10	Am-241	60	324.7	283.5
20	Am-241	60	208.7	194.3
25	Am-241	60	167.5	161
10	Th-234	93	-	43.8
20	Th-234	93	-	30
25	Th-234	93	-	24.8
10	U-235	186	-	371.9
20	U-235	186	-	253.8
25	U-235	186	-	210.3

MEASUREMENTS FOR WASTE DISK

The Am-241 source was measured at 5 cm intervals along the detector axis and at 5 cm intervals at 7.9, 15.7, 23.6 and 27.5 cm off axis. All response factors were adjusted for attenuation by 0.1 g/cc density soft waste by using tables of mass attenuation coefficients and path-lengths from the source positions to the detector.

The uniform response factor, for a 10 kg disk of waste measuring 55 cm in diameter by 40 cm depth, was then calculated by weighting each source location according to the volume element that it represented for detector locations at 10, 20 and 25 cm from the centre of the broadside of the disk. The extreme source locations (e.g. 27.5 cm radius) were not used to derive the uniform response factor.

For 2 sided counting (i.e. inverting the bag halfway through the count), the maximum response is in the center of the cylinder and was calculated as the average of the maximum and minimum on-axis response factors. The minimum response was achieved for a source location at the mid edge of the disk (27.5 cm off axis).

Figure 4 shows that all response factors increased as detector offset reduced, but the maximum and minimum response differed even more sharply from the uniform response than noted for the cylinder (figure 2).

Table 3 summarises the close agreement between measured response factors for the uniform disk geometry with those calculated using SNAP software.

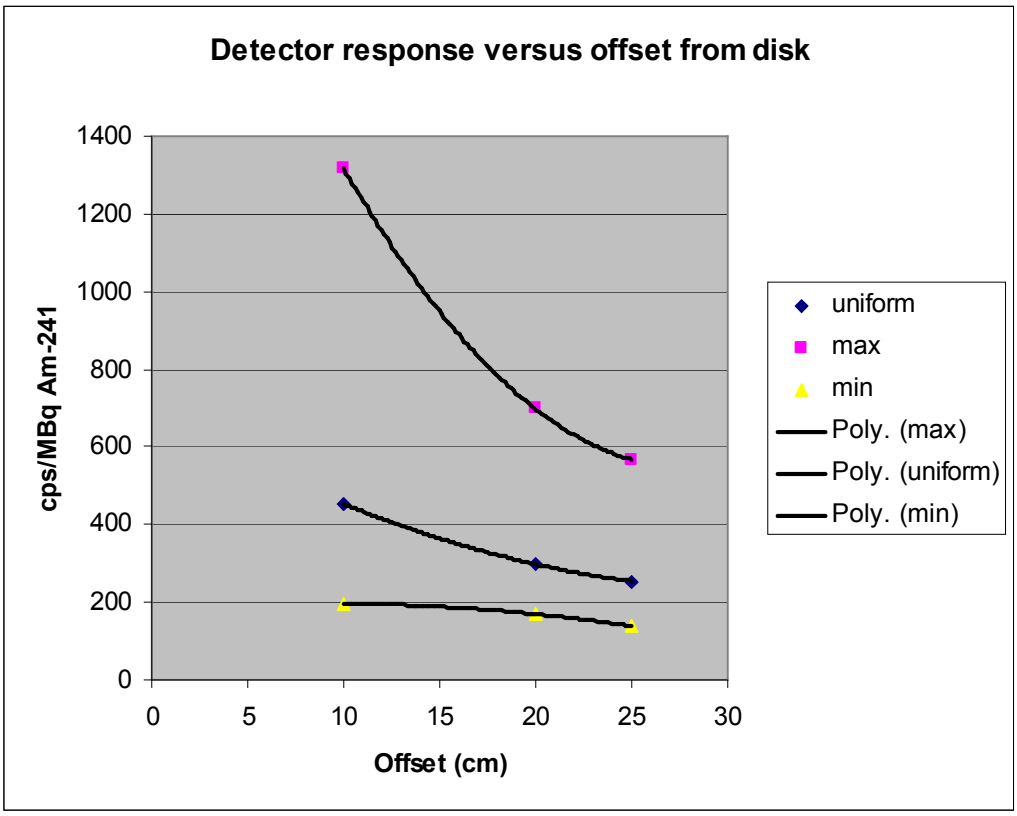

Figure 4 Detector response factors for disk

Table 3 Comparative uniform response factors (cps/MBq) for disk

Detector offset (cm)	Nuclide	keV	Measured	SNAP
10	Am-241	60	453	446
20	Am-241	60	297.9	308.4
25	Am-241	60	253.9	255.1
10	Th-234	93	-	67.3
20	Th-234	93	-	46.5
25	Th-234	93	-	38.5
10	U-235	186	-	550.3
20	U-235	186	-	378.9
25	U-235	186	-	313.4

MEASUREMENTS FOR BOTTLED LIQUID

The National Physical Laboratory (NPL) supplied a certified 1 litre standard solution of 103.6 Bq Am-241, in dilute nitric acid, contained within a 1 litre plastic bottle (17 cm high by 8.6 cm diameter). This was counted with the HPGe detector positioned at 10 cm from the centre/middle of the bottle in order to determine the uniform response factor for comparison with SNAP. Maximum and minimum response factors were measured by placing the Am-241 point source at the bottom/centre and center/edge of a rotating blank bottle containing water only (see figure 2 schematic). Table 4 summarises the results.

Table 4 Comparative response factors (cps/MBq) for bottled liquid

Response factor	Nuclide	keV	Measured	SNAP
Uniform	Am-241	60	1703.5	1836.5
Maximum	Am-241	60	2743.9	-
Minimum	Am-241	60	975.6	-
Uniform	Th-234	93	-	289.1
Maximum	Th-234	93	-	-
Minimum	Th-234	93	-	-
Uniform	U-235	186	-	2480.7
Maximum	U-235	186	-	-
Minimum	U-235	186	-	-

MEASUREMENTS FOR METAL PLATE

The Am-241 point source was measured at 30 cm along the detector axis and at several distances off axis. The uniform response factor, for a 60 x 60 cm surface, was calculated by weighting each response factor according to the relative area represented by the source. Maximum response was at 30 cm along the detector axis and minimum response at 42.6 cm off axis (i.e. at the plate corner). Table 5 summarises the results.

Table 5 Comparative response factors (cps/MBq) for metal plate

Response factor	Nuclide	keV	Measured	SNAP
Uniform	Am-241	60	473.5	459.2
Maximum	Am-241	60	691.5	-
Minimum	Am-241	60	195.1	-
Uniform	Th-234	93	-	66.9
Maximum	Th-234	93	-	-
Minimum	Th-234	93	-	-
Uniform	U-235	186	-	519.2
Maximum	U-235	186	-	-
Minimum	U-235	186	-	-

DETECTION LEVEL CALCULATIONS

Repeat measurements indicated that the background standard deviation (σ), for various locations and count times, was similar to the square root of the background counts (√B). Using lead bricks for partial shielding and a collimated detector, as pictured in figure 1, gave background count rates of around 0.2 cps for each photon region of interest (ROI) in the gamma spectrum. The detection levels (DL) at 2σ in table 6 were calculated, using equation 1, with B = 200 background counts, T = 1000s count time, F = measured or SNAP calculated response factors from tables 2-5 and figures 2-3 at 10 cm and typical isotopic multipliers (M) (i.e. Am-241 x 10 = Pu; U-235 x 40 = HEU; Th-234 x 1.55 = DU). For solid samples G = 10 kg for the cylinder or disk of soft waste; 1 kg for the 1 l liquid bottle, and 3600 cm^2 for the plate.

$$DL = (2\sqrt{B}/TFG)M \qquad \text{Equation 1}$$

The 2σ detection level was chosen to provide an acceptable balance between false positives and negatives. More conventional decision levels (2.33σ) and detection levels (4.65σ) may also be calculated.

Maximum and minimum detection levels, for uranium compositions, may be inferred from the maximum and minimum measurements at 60 keV and the relative detector response at 93 and 186 keV. However, the higher photon energies will produce less variation in detection levels than measured at 60 keV.

Table 6 Detection levels (Bq/g for bulk waste or Bq/cm^{-2} for surface)

Waste item	Material	Uniform	Minimum	Maximum
Cylinder	Pu	0.087	0.045	0.14
Cylinder	DU	0.10	-	-
Cylinder	HEU	0.30	-	-
Disk	Pu	0.062	0.021	0.14
Disk	DU	0.065	-	-
Disk	HEU	0.21	-	-
Bottle	Pu	0.17	0.10	0.29
Bottle	DU	0.15	-	-
Bottle	HEU	0.46	-	-
Plate	Pu	0.17	0.11	0.40
Plate	DU	0.18	-	-
Plate	HEU	0.61	-	-

DISCUSSION

The 1 Bq/g or 1 Bq/cm^2 clearance level was readily achieved for U compositions, even with a 'worst case' activity location within the waste. However, the 0.15 Bq/g clearance level for Pu compositions was only just achieved in this situation.

Furthermore, Pu compositions with higher Pu/Am-241 activity ratios could not be confidently placed in the exempt category.

The performance of the technique could be enhanced by using a larger diameter HPGe crystal since Am-241 response factors are proportional to the detector frontal surface area. For example an 85 mm diameter probe should reduce detection levels by a factor of 1.4, compared to the 61 mm probe, because the surface area and background are roughly doubled.

Compressing the disk and/or increasing the waste mass, is also estimated to give around factor of 2 reduction in detection levels, based on uniform activity distribution. For example simply increasing the mass from 10 kg to 20 kg gave a 25 % reduction in the efficiency for a uniform distribution for a doubling in mass (density). However, this increase in density also increases the self-attenuation which makes the over- and under-response bigger for the situation of non-uniform distributions.

Spectral summing could give a factor of 10 reduction if the spectra from 100 bags were added together (2). However, care is required. Simply counting the bag 100x longer gives a factor of 10 DL reduction, but now it hasn't been shown that each individual bag is compliant. If that is not a requirement then the maximum and minimum for each bag need not be used. Instead some factor of uncertainty should be used for each bag.

A further factor of 3 reduction could be achieved by monitoring within a shielded room, such as that used for in-vivo monitoring (IVM), where backgrounds are a factor of 10 lower. Another strategy would be to use the IVM monitor shown in figure 5.

Figure 5 IVM monitor counting a calibration phantom

The AWE IVM monitor has four germanium detectors (7 cm diameter by 2.5 cm thick) and could be operated within a shielded room. The detection levels at 2σ in table 7 were calculated using equation 1 and the approximate values of 0.05 cps for the combined background of 4 detectors and response factors five times that of the smaller single detector.

Table 7 Detection levels (Bq/g for bulk waste or Bq/cm2 for surface)

Waste item	Material	Uniform	Minimum	Maximum
Cylinder	Pu	0.0087	0.0045	0.014
Cylinder	DU	0.010	-	-
Cylinder	HEU	0.030	-	-
Disk	Pu	0.0062	0.0021	0.014
Disk	DU	0.0065	-	-
Disk	HEU	0.021	-	-
Bottle	Pu	0.017	0.010	0.029
Bottle	DU	0.015	-	-
Bottle	HEU	0.046	-	-
Plate	Pu	0.017	0.011	0.040
Plate	DU	0.018	-	-
Plate	HEU	0.061	-	-

CONCLUSIONS

The techniques tested were capable of meeting the recently reduced clearance levels of < 1Bq/g or < 1 Bq/cm2 for uranium wastes and < 0.15 Bq/g or < 0.15 Bq/cm2 for some Pu wastes. However, further development is required to achieve clearance for Pu wastes having high Pu/Am-241 activity ratios using a single germanium detector. This is because a typical Pu/Am alpha activity ratio is 10/1, but there are rare examples of much higher activity ratios. An alternative solution would be to use the IVM 4 detector system in a shielded room

RECOMMENDATIONS

The performance of the technique could be enhanced by:

- employing larger diameter HPGe detectors, having better efficiency,
- reducing the thickness and increasing the density of the waste disk (for example doubling the disk mass gave only a 25 % reduction in uniform response),
- counting longer
- lowering the background
- spectral summing,
- monitoring within a fully shielded room with IVM detectors (i.e. both lower the background and use higher efficiency detectors).

REFERENCES

1. T.J.Miller, WM2011, paper 11004.
2. R.Lucero, WM2011, paper 11249.