Applications Where Snap is BPM for Radioactive Waste Assay - 8033

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ABSTRACT

Historically, the Atomic Weapons Establishment (AWE) at Aldermaston in the United Kingdom (UK), has used a variety of assay techniques to measure the radioactive content of a diverse range of waste packages from decommissioning, operational and legacy sources. The regulator, the Environment Agency in the UK, places conditions and limits on AWE through an authorisation within the Radioactive Substances Act (RSA93). The conditions and limits require Best Practical Means (BPM) measurements to be used to demonstrate compliance with the authorisation. Hence, the assay technique employed needs to achieve a balance between risk of exposure, environmental considerations, technological considerations, health and safety considerations and cost effectiveness, without being grossly disproportionate in terms of money, time or trouble.

Recently published work has concluded that the Spectral Non-destructive Assay Platform (SNAP) assay system is BPM for Depleted Uranium (DU) waste assay at AWE (1) and low level plutonium in soft drummed waste, HEPA filters and soils (2-4). The purpose of this paper is to highlight other applications where SNAP represents BPM for radioactive waste assay. This has been done by intercomparison studies of SNAP with other assay techniques, such as Segmented Gamma Scanner (SGS) and Passive Neutron Coincidence Counter (PNCC). It has been concluded that, for a large range of waste packages encountered at AWE, SNAP is BPM.

INTRODUCTION

This paper starts with a historical review of radioactive waste assay at AWE and then examines some of the more recent BPM studies for specific applications. The SNAP assay system is described and its performance compared with other systems (e.g. SGS, PNCC) for actual and simulated waste packages. Finally, conclusions are drawn highlighting the areas where SNAP represents BPM for radioactive waste assay.

HISTORICAL

Over the years many different techniques have been applied to radioactive waste assay at AWE. However, after the ban on deep sea disposal in 1983, all plutonium contact material was stored on site as Intermediate Level Waste (ILW). This was because there is no ILW repository in the UK and there was a lack of confidence in the available measurement techniques to demonstrate compliance with the Low Level Waste Repository at Drigg (LLWRD) Conditions For Acceptance (CFA). This limits Pu and Am alpha activity to < 0.1 GBq/t (< 100 Bq/g) per consignment. For a typical average mass (60 kg) 200 l waste drum this equates to < 6 MBq or 2mg Pu (table I) and PNCC and SGS detection limits are typically around 100 mg total Pu.

Table I UK Waste Categories

Category	Bq/g Pu alpha	Pu (mg/drum)
UK ILW	Above 4,000	Above 80
UK LLW	100 to 4,000	2 to 80
LLWRD	0.4 to 100	0.008 to 2
UK BRC (Below Regulatory Concern)	Below 0.4	Below 0.008

At the Waste Management 2000 (WM2000) conference (Tucson, Arizona) AWE staff met with Los Alamos National Laboratory (LANL) staff to discuss the application of SNAP to waste assay at AWE.

Subsequently a SNAP system was procured in 2001. From 2002-2003 three papers were published demonstrating the feasibility of meeting the LLWRD CFA for soft drummed waste, HEPA filters and soils (2-4). The strategy was to monitor the 60 keV signature from Am-241 ingrowth in Pu, using the SNAP assay system and calculate the Pu alpha activity from the known isotopic fingerprint for the waste stream.

Initially the technique was limited to low-density and low atomic number (Z) waste streams because of the limited range of the 60 keV photon. However, drum radiographs revealed that many drums containing metallic wastes had relatively light shielding properties (e.g. filter housings). Hence the applicability of the technique was broadened.

By 2007 around 10,000 waste packages (drums, filters, wrapped items) were characterised using the SNAP assay system. It was found that a large proportion (up to 80 % for some legacy stores) met the LLWRD CFA and could be sent for off site disposal. This option being significantly cheaper (£250 per drum) in comparison to indefinite long-term storage at AWE followed by consignment to an ILW repository (estimated at around £10,000 per drum). A front page article published in the June 2007 AWE Today paper (issue 42) highlighted these achievements by recognising that the ILW reclassification work undertaken had already resulted in cost savings in the region of £10 million.

BPM STUDIES

A BPM study starts with an objective. For example to segregate LLW from ILW. Then all of the cost considerations (e.g. time, money, personnel exposure, training, maintenance, etc.) are balanced against the performance considerations (e.g. accuracy, detection limit). A simple BPM study with a limited number of criteria, taken from reference 1, is summarised in table II.

Table II DU waste assay options BPM summary

Option	Costs	Accuracy	DL	Suitability
Passive Neutron Counting (PNC)	Low	Medium	Poor	No
Passive Neutron Coincidence Counting (PNCC)	High	Medium	Poor	No
Active Neutron Counting (ANC)	Very High	High	Good	Too expensive
Segmented Gamma Scanner (SGS) (Low Resolution)	Medium	Medium	Good	Inferior accuracy
Segmented Gamma Scanner (SGS) (High Resolution)	High	High	Good	Too costly and complex
Passive Low Resolution Gamma Spectrometry (LRGS)	Low	Medium	Good	Inferior accuracy
Passive High Resolution Gamma Spectrometry (HRGS) (SNAP)	Medium	High	Good	Yes
Gross activity	Low	Medium	Fair	No
Chemical analysis	Medium	Low	Good	No

Reference 1 concludes that SNAP is BPM for DU waste assay because it delivers high accuracy and good detection levels with relatively low costs and time and effort compared to the alternatives.

SNAP

Table III summarises the principal components of the SNAP assay system and figure 1 shows SNAP monitoring a standard waste drum positioned on a rotating turntable.

Table III SNAP Assay System Components

Component	Specification
Detector	High Purity Germanium (HPGe) N-type (45 % relative efficiency)
Collimator	20 mm lead (copper lined) fitted flush with detector end cap
Multichannel analyser	ORTEC digiDART
Computer	Laptop with windows 98
MCA emulator	Maestro 32
Analytical software	SNAP (5)
Drum turntable	10 rpm
Trolley	ORTEC ISOCART

Figure 1 SNAP monitoring a waste drum on a turntable

The SNAP hardware consists of standard High-Resolution Gamma Spectrometry (HRGS) components. It is trolley mounted for portability and operates in purely passive mode (i.e. without a transmission source). Normally the detector is located 'far field' (e.g. at one drum diameter as shown in figure 1). The SNAP analytical software, from Eberline Services, corrects the detector calibration for counting geometry and gamma ray attenuation. It also has routines for differential peak analysis and plutonium and uranium lump corrections so that all gamma signatures from a given isotope provide consistent results. The reader is referred to the SNAP user's manual for a detailed description of SNAP features and analysis procedures (5).

INTERCOMPARISON STUDIES

Waste Drum Standards (NPL)

AWE has participated in a National Physical Laboratory (NPL) 200 l drum comparison exercise. This involved sending a NPL drum standard, spiked with uniformly distributed activity, for assay at nuclear establishments throughout the UK. Whilst at AWE the NPL drum was assayed using SNAP and AQ2 (an assay system with 3 HPGe detectors and 3 transmission sources positioned to correct for photon attenuation through the top middle and bottom of the drum). The results are summarised in table IV and are expressed as a percentage of the true activity declared by NPL (7).

Table IV Percentage of true activity (NPL drum standard)

Isotope	SNAP	AQ2
Am-241	81	790
Cs-137	95	95
Co-60	94	75

The overestimation of Am-241, by AQ2, is attributed to the transmission measurement giving a false identification of the waste package type. The drum standard package was a 200 l drum with a 0.8 mm steel wall, however AQ2 software identified it as a 100 l drum (1.1 mm steel) inside a 200 l drum (1.5 mm steel) with a steel sheath (1.5 mm steel). Hence the shielding overestimate of 3.3 mm would give an activity overestimate of almost a factor of 20 at 60 keV.

Heterogeneous Waste Drum Standards (AWE)

Heterogeneous drum standards were constructed by filling the bottom third of a 200 l drum with solid wood roundels (high density region), the middle third with empty polythene bottles (low density region) and leaving the top third empty. A source was then positioned either towards the bottom or middle of the drum (just inside the drum wall) and the drum positioned on a turntable for monitoring by SNAP, SGS and AQ2. The results, expressed as a percentage of the true activity, are summarised in table V.

Table V Percentage of true activity (AWE drum standards)

Isotope	Position	SNAP	SGS	AQ2
Am-241	Bottom	96	*	307
Am-241	Middle	139	*	102
Cs-137	Bottom	107	139	123
Cs-137	Middle	138	97	119
U-235	Bottom	86	38	34
U-235	Middle	114	24	21
Pu-239	Bottom	74	103	47
Pu-239	Middle	99	76	74

*Cd filter precludes low level Am-241 measurement with SGS

SNAP results were calculated based on uniform matrix density and could be improved by applying the actual matrix density variations to the modelling. However, they show that SNAP tended to overestimate when the point source was located in a less dense region towards the middle of the drum (i.e. closer to the detector). The SGS overestimated when the source was located in a relatively high-density region of the drum, but underestimated U-235 due to source self-absorption effects which was flagged by abnormal peak area ratios (e.g.143/205 keV of below 2/1). SNAP compensated for this effect by using a lump correction routine. AQ2 also underestimated U-235 and Pu-239, but overestimated Am-241 by a factor of 3 when positioned in a relatively dense region within the drum.

U-235 Waste Drums

Table VI summarises the results obtained from some of the higher activity legacy waste drums (containing U-235 contamination) together with results from simulated waste drums (containing packets of U-235 swarf) and waste drum standards (containing uniformly distributed waste and randomly positioned U-235 sources).

Table VI Results for U-235 waste drums (U-235 g)

Matrix	Activity	SNAP	SGS	AQ2
Legacy waste	Contamination	54.2	41.5	32.6
Legacy waste	Contamination	43.0	38.4	25.8
Legacy waste	Contamination	58.0	47.4	14.3
Legacy waste	Contamination	39.8	34.0	24.7
Legacy waste	Contamination	49.8	39.6	27.8
Legacy waste	Contamination	53.1	40.6	30.1
Simulated waste	Swarf (5 x 2 g)	10.3	3.9	4.0
Uniform waste	Sources (10 + 5 g)	15.9	< 1	< 1

SNAP tends to give higher results for legacy waste drums, compared to SGS and AQ2, because small lump corrections are sometimes required due to aggregates of contamination. These corrections are larger when lumps of U-235, in the form of swarf or sources, are introduced which impacts on the performance of the SGS and AQ2.

Pu-239 Waste Drums

The estimated distribution of Pu activity within drums from all sources (i.e. decommissioning, legacy and operational) is summarised in table VII. However, it is recognised that some legacy waste stores have much higher proportions of LLWRD (up to 80 %).

Table VII Distribution of Pu activity within waste drums

Category	Pu g/drum	% of total drums
LLWRD	< 0.002	40
UK LLW	0.002 to 0.080	20
UK ILW	0.1 to 1	10
UK ILW	1 to 10	25
UK ILW	10 to 100	5
UK ILW	> 100	<< 1

Typical Pu isotopic compositions gave the detection levels for drums summarised in table VIII. SNAP gave good detection levels, with short count times. Hence it can be used to rapidly segregate wastes.

Table VIII Detection levels for Pu

Technique	Measurement	Count time (s)	Pu detection limit (g)
SNAP	Am-241 @ 60 keV	100	0.00005
SNAP	Pu-239 @ 129 keV	100	0.03
SNAP	Pu-239 @ 414 keV	100	0.1
SGS	Pu-239 @ 414 keV	2700	0.1
AQ2	Pu-239 @ 414 keV	1800	0.005
PNCC	Pu-240	1800	0.1

Studies on lower activity drums (up to 0.5 g Pu) showed that reasonably consistent results were obtained from the PNCC and SNAP, using the 60, 129 and 414 keV gamma signatures, given the poor counting statistics with a 100 s count at 129 & 414 keV. Table IX summarises the results in order of increasing Pu content based on the 60 keV signature and the isotopic fingerprint for the waste stream. Around 70 % of the time the 60 keV result was within a factor of 2 of the PNCC result.

Table IX Pu assay results for low activity drums

SNAP Pu-239 (mg) (via 60 keV)	SNAP Pu-239 (mg) (via 129 keV)	SNAP Pu-239 (mg) (via 414 keV)	PNCC Pu (mg)
1.2	< 30	< 100	< 100
2.8	< 30	< 100	< 100
3.1	< 30	< 100	< 100
5.1	< 30	< 100	< 100
7.6	< 30	< 100	< 100
12.8	< 30	< 100	< 100
17.8	< 30	< 100	< 100
25.7	< 30	< 100	< 100
31.2	< 30	< 100	< 100
98.6	136	155	< 100
101.8	86	163	< 100
124.7	134	297	227
176.5	153	269	95
200.3	57	132	103
216.5	185	242	134
234.2	226	185	157
235.6	139	207	93
250.8	270	388	196
286.2	222	318	179
323.4	206	231	94
332.6	217	299	187
499.0	461	584	322
556.6	341	553	237

At higher Pu activities, up to 10 g, agreement between SNAP and PNCC was better due to better counting statistics at 414 keV. Some typical results, in order of increasing Pu activity, are summarised in table X. Comparisons with SGS and AQ2 were not available.

Table X Pu results for legacy drums up to 10 g Pu

SNAP Pu-239 (g) via 414 keV	PNCC Pu (g)
2.2	2.1
3.4	3.2
4.0	4.2
4.8	5.0
6.1	6.2
8.1	8.0
10.4	9.7

Table XI summarises the results from some of the higher activity legacy drums. SNAP results were generally in reasonable agreement with the SGS, but the AQ2 results were much more variable. For the higher activity legacy drums the PNCC often gave much higher results. In some cases the high PNCC results can be accounted for by $\propto$, n interactions. For example, the observation of intense 1275 keV gamma rays from Na-22 was indicative of the fluorine $\propto$,n reaction. In other cases it was found that the PNCC results had assumed an incorrect value for the Pu-240 concentration.

Table XI Results for Pu-239 legacy drums (Pu-239 g)

SNAP	SGS	AQ2	PNCC
52.3	46.6	84	127
96.7	65.0	120	168
92.1	51.0	149	155
35.6	27.0	61.2	215
60.7	43.9	110.3	231
50.5	52.2	92.1	289
76.8	68.3	54.1	112
40.5	34.7	40.7	309
77.8	69.9	63.9	106

DISCUSSION

Recently published work has concluded that SNAP represents BPM for DU waste assay and low level plutonium in soft drummed waste, HEPA filters and soils (1-4). The purpose of this paper is to highlight other areas where SNAP represents BPM.

Intercomparison studies, on waste packages encountered routinely at AWE, have shown that SNAP usually gives similar results to other more costly techniques such as SGS and PNCC. Differences between reported results were rare and on inspection could be rationalised.

The SGS is considered to be the gamma-ray technique benchmark for accuracy and SNAP consistently gives similar results, with the additional benefit of being able to recognise (by abnormal peak area ratios) and correct for abnormal shielding effects, such as lumps of uranium and plutonium or lead shielding encapsulating activity. In addition, SNAP is able to give better detection levels and shorter count times because it counts the whole of the drum, rather than individual segments. Furthermore, SNAP is medium cost and has the versatility to perform in-situ assay of a wide range of waste package types, such as: pipes, oil containers, and wrapped packages of various shapes and sizes.

Passive neutron techniques have advantages for the more heavily shielded waste streams. However, NPL Measurement Good Practice Guide No. 34 (6) indicates that underestimation by gamma-ray techniques, even in the most severely attenuating sample matrix, is unlikely to exceed a factor of 2 or 3 if the activity is present as dispersed contamination that is typical for radioactive waste.

Active neutron techniques are costly and can severely underestimate when lumps or aggregates of fissile material are present (6).

There are, however, two areas of application where SNAP could be usefully supplemented by other techniques. The first is meeting the LLWRD CFA for more attenuating waste streams. The AQ2 could be used, with an extended count time, to measure down to < 2 mg Pu-239 directly by utilising the more penetrating 129 and 414 keV gamma signatures, rather than Am-241 at 60 keV.

The second area is using the PNCC for the more heavily shielded waste streams having high Pu content. However, these drums account for only a very small proportion of the waste at AWE.

SNAP has been able to reclassify over 100 ILW drums per day, using a 100 s count time and forklift trucks to move drums to and from a turntable. Automated versions of AQ2 or SGS and PNCC can only manage around 15 per day because of the much longer counting times normally employed. Tables XII and XIII summarise the above discussion within the BPM studies for EU and Pu waste streams.

Table XII EU waste assay options BPM summary

Option	Costs	Accuracy	DL	Suitability
Total Neutron Counting (TNC)	Low	Medium	Poor	No
Active Neutron Counting (ANC)	Very High	High	Good	Too expensive, underestimates lumps or aggregates of U-235
Segmented Gamma Scanner (SGS) (Low Resolution)	Medium	Medium	Good	Inferior accuracy, no possibility of correcting for abnormal shielding effects such as lumps of U-235
Segmented Gamma Scanner (SGS) (High Resolution) (AQ2)	High	High	Good	Too costly and complex and time consuming. No lump correction capability
Passive Low Resolution Gamma Spectrometry (LRGS)	Low	Medium	Good	Inferior accuracy. No possibility of correcting for abnormal shielding effects
Passive High Resolution Gamma Spectrometry (HRGS) (SNAP)	Medium	High	Good	Yes
Gross activity	Low	Medium	Fair	No
Chemical analysis	Medium	Low	Good	No

Table XIII PU waste assay options BPM summary

Option	Costs	Accuracy	DL	Suitability
Passive Neutron Coincidence Counting (PNCC)	High	Medium	Poor	Applicable to limited numbers of highly shielded high activity drums
Active Neutron Counting (ANC)	Very High	High	Good	Too expensive, underestimates lumps or aggregates of Pu
Segmented Gamma Scanner (SGS) (Low Resolution)	Medium	Medium	Good	Inferior accuracy, no possibility of correcting for abnormal shielding effects such as lumps of Pu
Segmented Gamma Scanner (SGS) (High Resolution) (AQ2)	High	High	Good	Too costly and complex and time consuming. Lump correction capability not effective. Suitable for limited numbers of more shielded low activity drums (AQ2)
Passive Low Resolution Gamma Spectrometry (LRGS)	Low	Medium	Good	Inferior accuracy. No possibility of correcting for abnormal shielding effects
Passive High Resolution Gamma Spectrometry (HRGS) (SNAP)	Medium	High	Good	Yes

CONCLUSIONS

Recent publications have identified specific applications where SNAP represents BPM for radioactive waste assay. Subsequent studies have confirmed that SNAP generally represents BPM for most normally encountered waste packages at AWE. However, SNAP should be supplemented by PNCC for the more heavily shielded, higher level Pu containing waste streams and by AQ2, for the more heavily shielded waste at the LLWRD CFA level.

REFERENCES

(1) T.J.Miller, Depleted Uranium Waste Assay at AWE, Waste Management Symposium, Tucson, Arizona, 2007.
(2) T.J.Miller, Assay of Low Level Plutonium in Soft Drummed Waste, INMM Meeting, Orlando Florida, 2002.
(3) T.J.Miller, Plutonium assay in HEPA Filters at the 100 Bq/g Threshold, INMM Meeting, Phoenix, Arizona, 2003.
(4) T.J.Miller, Plutonium Assay in Soil at the BRC Threshold. Waste Management Symposium, Tucson, Arizona, 2003.
(5) SNAP Users Manual, Eberline Services Inc., February 2001.
(6) NPL Measurement Good Practice Guide, Radiometric Non-Destructive Assay, No. 34, January 2003.
(7) NPL Waste drum intercomparison study workshop, London, UK, September 21, 2007.