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Application Of ISOCS In The Measurement Of Bulk Plutonium Contaminated Waste During Decommissioning

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1- Introduction

A decommissioning project was undertaken where the waste was anticipated to be < 100Bqg^{-1} total Pu.

Usual methods employed for bulk wastes involve neutron counting techniques such as PNCC. These cannot achieve the detection levels required. Sampling requires a high degree of laboratory support which was not possible given the time constraints. Therefore the use of High Resolution Gamma Spectrometry was investigated.

Fig 1 – Typical Boxes at Project Start

Table 1 - Emission rates per gram from Plutonium aged 30y

	Neutron (ns^{-1} / g)		Gamma (γs^{-1} / g)		
	SF	α/n (PuO_2)	^{239}Pu 129keV	^{241}Am 60keV	^{241}Am 722keV
A Grade	60	45	1.35E5	1.23E8	671
O Grade	360	200	8.67E4	3.19E9	1.74E4

The waste consisted of metals (piping / sheet), plastics, rubble etc typical of a facility that handled Pu. Table 1 illustrates that with the appropriate degree of waste packing the use of ^{241}Am 60keV gamma ray is worthy of consideration in spite of its low energy. A key parameter to determine however is the uncertainty.

2 - Equipment

The HRGS system consisted of an ISOCS characterised BEGe2825 HPGE mounted on a cryostat. The MCA was an Inspector2000 which provides digitally stabilised spectra without the need for a spectroscopy amplifier. This had a USB connection to a laptop running Genie2k spectroscopy software and Canberra's ISOCS calibration software. The waste was packaged into drums in a random fashion. The drums were placed on a turntable at 0.5m from the detector.

The system can also be used for other material as well as drums

Fig 2 – Example Measurement Setup

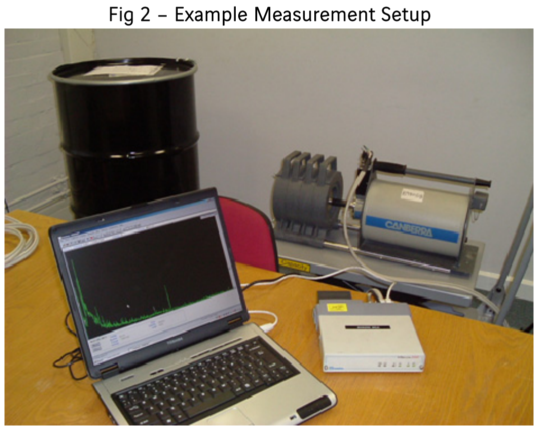
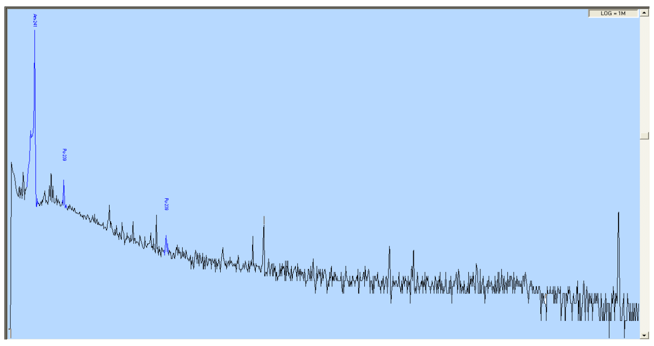


Fig 3 – Measurement of Wrapped Packages

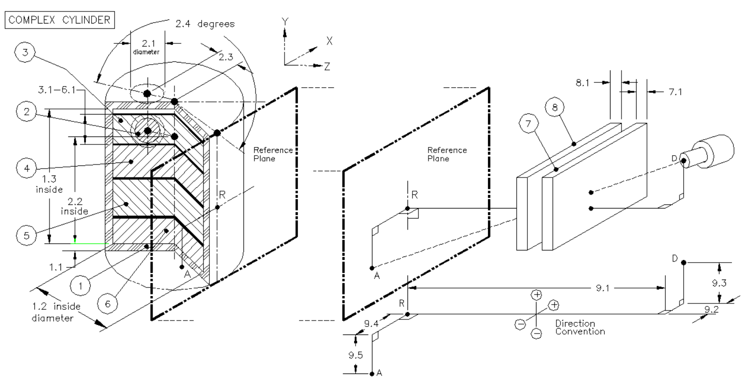
Fig 4 – Drum Spectrum showing ^{241}Am and ^{239}Pu lines



3 - Modelling Methodology

The ISOCS software enables a computer representation of the waste to be generated. This, along with the unique detector characterisation, enables detector efficiencies to be computed at defined energies. The software also enables the input parameters to be varied in a random manner but within defined limits and not only provides a composite efficiency for each energy but an uncertainty that is representative of all the input variations.

Fig 5 – ISOCS Complex Drum Template



The modelling template allowed variations in container size, wall thickness, and material. In addition variations in waste material mixtures, bulk density and activity distribution were applied. The limitations on the input parameters are, in most cases, driven by the limitations on what can be placed into the container. For example, drum weight is measured on a drum by drum basis and therefore places limits on the variations in bulk density.

4 - Modelling Results

Table 2 shows the efficiency and calibration uncertainty for a drum which is filled to capacity and one which is half filled and hence able to achieve a higher bulk density. For this project the worse case uncertainty was applied as a base uncertainty for all the drums measured.

Table 2 – Efficiencies and Uncertainties

Nuclide	Energy (keV)	Filled drum at 0.3gcm^{-3}			Half filled Drum at 1.91 gcm^{-3}		
		Efficiency (cps Bq^{-1})	Massimetric Efficiency (cps / Bqg^{-1})	Uncertainty (%)	Efficiency (cps Bq^{-1})	Massimetric Efficiency (cps / Bqg^{-1})	Uncertainty (%)
^{241}Am	59.54	1.49 E-5	0.94	40	3.22 E-6	0.64	52
^{239}Pu	129.296	7.35 E-5	4.62	14	1.62 E-5	3.25	24
^{235}U	143.767	7.52 E-5	4.73	13	1.69 E-5	3.39	22
^{235}U	185.72	7.19 E-5	4.52	13	1.69 E-5	3.39	19
^{235}U	205.316	6.74 E-5	4.22	11	1.61 E-5	3.23	18
^{239}Pu	375.05	4.00 E-5	2.52	11	1.08 E-5	2.15	16
^{239}Pu	413.71	3.66 E-5	2.30	11	9.99 E-6	2.00	15

5 - Minimum Detectable Activities (MDA)

The above calibration methodology was applied to a 15 minute background measurement and the following MDA's were achieved using the standard Currie method.

Table 3 – Individual Nuclide MDA's

Nuclide	LLW Drum			
	Full @ 0.3gcm^{-3}		Half Full @ 1.91gcm^{-3}	
	Bqg^{-1}	Bq	Bqg^{-1}	Bq
^{235}U	0.05	3.1 E3	0.07	1.3 E4
^{239}Pu	476	2.99 E7	677	1.4 E8
^{241}Am	0.30	1.91 E4	0.44	8.8 E4

As can be seen the MDA's for ^{239}Pu preclude using these to achieve the required sensitivity of 100Bqg^{-1} total Pu (including ^{241}Am). However if the nuclide composition of the waste is understood i.e. it has been fingerprinted then it maybe possible to achieve this detection limit by combining it with the ^{241}Am results. The project allowed for a programme of nuclide fingerprinting. When this was applied the following MDA's for the mix were produced:

Table 4 - Total MDA in Bqg^{-1}

'Reference' Nuclide	Half Full LLW Drum @ 1.91 gcm^{-3}			
	Nuclide	Total	Total Alpha	Inferred ^{241}Pu
^{239}Pu	677	1504	954	556
^{241}Am	0.44	7.33	4.4	2.7

When determining if the waste is less than the required threshold (100Bqg^{-1}) the sum of all the alpha emitting nuclides within the fingerprint has to be determined. To this sum is added the upper 95% confidence interval i.e. 1.645σ where σ is the combination of counting statistics and the calibration uncertainty defined above.

6 - Conclusions

By understanding the waste in terms of its variations in material matrices, activity distributions, bulk densities and measurement geometry it is possible to determine a realistic uncertainty that can be applied to the determination of the 95% confidence level activity within plutonium contaminated material.