VLLW/Exempt Waste Assay at AWE Using Cronos - 12001

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ABSTRACT

Previous studies have indicated that waste bag photon monitors, such as the CANBERRA Cronos-11, have the potential to assay photon emitting uranium (U) and plutonium (Pu) contaminated wastes at the new UK VLLW/exempt waste thresholds of 1 Bq/g U and 0.15 Bq/g Pu. However, the technique has limitations associated with the operation of its plastic scintillation photon detectors. These include the absence of isotopic information and a variable detector background that is modified by the chemical and physical composition of the waste as well as the presence of any naturally occurring radioactive material (NORM). Hence, it is vital to operate Cronos in a low/non-fluctuating background environment and segregate wastes according to isotopic fingerprint, NORM concentration and waste composition.

This paper presents results from subsequent Cronos measurements on a range of uncontaminated waste materials, contaminated wastes from Pu and U facilities and a National Physical Laboratory (NPL) waste package standard. The results were consistent with other assay techniques and show that Cronos can be used for exempt waste assay provided that it is operated in a low/non-fluctuating background area and the waste is well segregated before monitoring.

INTRODUCTION

The Cronos bag monitor has a 350 l counting chamber (63.5 x 63.5 x 87 cm) surrounded on all six sides by plastic scintillation photon detectors (5 cm thick) and lead shielding (5 cm thick) (figure 1). All photons, from 50 to 2,000 keV, interacting with the detectors are counted. Previous work has confirmed supplier's isotopic detection levels and mapped the detector response to photon sources located throughout the counting chamber [1]. It was concluded that Cronos had the potential to assay wastes with consistent isotopic fingerprints, at the new exempt thresholds of < 1 Bq/g U and < 0.15 Bq/g Pu (1 Bq/g Pu-241; 0.1 Bq/g Pu alpha), when sited in areas with low/non-fluctuating photon background. Careful segregation of wastes was required because the background counts were significantly modified by waste characteristics, such as: NORM content, density, composition and distribution in the counting chamber.

This paper examines the results from subsequent Cronos measurements on a range of uncontaminated materials in order to establish the ‘blank’ mean and standard deviation (σ) for the count rates (cps) from the principal waste material types encountered. Detection limits were re-calculated using the measured σ values.

Contaminated wastes, from Pu and U facilities (and NPL waste standards), were also measured with Cronos and the results checked against high resolution gamma spectrometry (HRGS) for consistency.

Figure 1 Cronos-11 bag monitor

MEASUREMENTS ON UNCONTAMINATED MATERIALS

Only low bulk density, low atomic number (Z) materials were examined because of the difficulties in Cronos calibration for high density, high Z objects [1]. Approximately 35 kg of each material type was spread evenly throughout the 350 l counting chamber to give a bulk density of 0.1 g/cc. Repeat 600 s counts were done to generate ten measurements for each material. Table 1 summarises the mean and σ values achieved for the net counts (i.e. gross counts minus counts for the continuously updated Cronos background).

Table I. Mean net cps from 35 kg samples of uncontaminated materials

Material	Net cps	σ	Net cps + 2σ
Empty chamber	1.8	9.7	21.2
PVC	3.4	6.1	15.6
Tyvek	25.0	3.8	32.6
Paper	41.1	9.0	59.1
Marigold gloves	61.5	16.3	94.1
Overshoes	66.1	5.9	77.9
Supertex coveralls	68.0	15.1	98.2
Orange coveralls	138.4	6.9	152.2

The results in table 1 highlight the importance of segregating waste into material types having similar net cps. Large variations were also observed for different batches of the same material type. For example a different batch of 35 kg paper gave around 133 cps [1]. Detection levels (2σ) were calculated using equation 1 and the previously reported counting efficiencies [1] and are summarized in table 2.

$$DL = 2\sigma \div (E \times G) \quad \text{(Eq. 1)}$$

where:
DL = detection limit (Bq/g)
σ = standard deviation for blank (10 cps)
E = counting efficiency (0.00606 cps/Bq for Pu; 0.00635 cps/Bq for HEU and 0.042 cps/Bq for DU)
G = sample mass (35 kg)

Table II. Detection levels (Bq/g)

Composition	DL (Bq/g)
Pu	0.09
HEU	0.09
DU	0.014

MEASUREMENTS ON WASTE FROM A PLUTONIUM FACILITY

Table 3 summarises the results for eight bags of waste produced by a Pu facility. The sample activity was calculated by dividing the net cps by the Pu counting efficiency and the sample mass. The blank activity (i.e. blank cps/E x G) was then subtracted to give the Pu activity measured above the apparent Pu activity indicated for the blank.

Table III. Results for bags from a Pu facility

Material	kg	Net cps	Sample (Bq/g)	Blank (Bq/g)	Pu (Bq/g)
PVC	29.9	-8.6	-0.05	0.02	-0.07 +/- 0.02
PVC	30.2	-5.4	-0.03	0.02	-0.05 +/- 0.01
PVC	33.7	25.4	0.12	0.02	0.10 +/- 0.03
PVC	28	10.2	0.06	0.02	0.04 +/- 0.01
PVC	35.3	9.8	0.05	0.02	0.03 +/- 0.01
Orange coveralls	17.2	152.5	1.46	0.65	0.81 +/- 0.2
Tyvek	13.1	1944	24.49	0.12	24.370 +/- 6.1
Cotton gloves	30.8	35.1	0.19	0.11	0.08 +/- 0.02

The tyvek bag indicated a total activity of 24.37 x 13.1 = 319.2 kBq Pu which was equivalent to 31.92 kBq Am-241 since Cronos was calibrated using an Am-241 source and a Pu/Am-241 activity ratio of 10/1 because Pu makes no significant contribution to the photon count rate [1].

The bag was placed in a standard 200 l waste drum and assayed using an HRGS technique that had been verified as accurate using a number of certified NPL waste drum standards [2]. This indicated 38.8 kBq Am-241 which was 22 % higher than Cronos, but within the +/- 25 % uncertainty established for both techniques [1-2].

MEASUREMENTS ON WASTE FROM A ENRICHED URANIUM FACILITY

Thirty waste bags, from a enriched uranium facility, were assayed using Cronos. The waste was mixed material (PVC, paper, coveralls, gloves) and so it was not possible to subtract a blank activity for the apparent HEU Bq/g from the waste material itself. Hence the net cps was divided by the HEU counting efficiency and the sample mass to give a total HEU activity which included the apparent HEU activity from the waste material in addition to any HEU present. However, 70 % of the bags still indicated < 1 Bq/g HEU (table 4). Detection levels were around 0.1 Bq/g for the heavier bags and 1 Bq/g for the lighter bags.

The two most active bags were checked using HRGS and this gave consistent results (table 5).

Table IV. Results for bags from HEU facility

kg	Net cps	Bq/g
5.9	482	12.9 +/- 3.2
7.1	8.0	0.2 +/- 0.1
9.4	30.5	0.5 +/- 0.1
33.7	52.2	0.2 +/- 0.1
4.4	6.1	0.2 +/- 0.1
7.3	15.1	0.3 +/- 0.1
3.8	-9.3	-0.4 +/- 0.1
3.4	996.1	46.1 +/- 11.5
10.1	14.2	0.2 +/- 0.1
6.8	12.0	0.3 +/- 0.1
7.8	30.4	0.6 +/- 0.2
3.1	22.2	1.1 +/- 0.3
2.7	-6.6	-0.4 +/- 0.1
15.6	-0.7	0.0 +/- 0.1
6.0	110.1	2.9 +/- 0.7
4.1	10.4	0.4 +/- 0.1
11.0	132.7	1.9 +/- 0.5
5.9	12.5	0.3 +/- 0.1
6.1	22.8	0.6 +/- 0.2
7.5	8.4	0.2 +/- 0.1
10.3	7.3	0.1 +/- 0.1
11.7	49.7	0.7 +/- 0.2
4.2	20.8	0.8 +/- 0.2
2.2	13.6	1.0 +/- 0.3
10.3	41.9	0.6 +/- 0.2
5.4	61.0	1.8 +/- 0.5
5.5	16.8	0.5 +/- 0.1
4.4	21.5	0.8 +/- 0.2
3.9	150.9	6.1 +/- 1.5
6.0	59.1	1.6 +/- 0.4

Table V. Inter-comparison between Cronos and HRGS (mg U-235)

Bag	Cronos	HRGS	Cronos/HRGS
1	23.9 +/- 6.0	19 +/- 4.8	1.26
8	49.4 +/- 12.4	41.8 +/- 10.5	1.18

MEASUREMENTS ON THE NPL STANDARD

Calibration has been achieved using the volume weighted technique described in reference 1. The reader is referred to this document for a detailed account of the results. Verification has been done using a certified NPL volume source that fills the Cronos counting chamber. This consists of a corrugated carton (62x62x84 cm) containing a series of nine filter papers (each 58x58 cm), each spiked uniformly with a standard solution of Am-241 and each separately laminated. The filter papers were interspersed within the carton between a series of ten polythene inactive low density inserts (each 60x60x8 cm). The total Am-241 activity was 2857 +/- 12 Bq (reference time 01/01/10) in 29 kg (around 0.1 Bq/g and 0.1 g/cc).The mean cps/Bq Am-241 counting efficiency for the source (6.14 %) was similar to that achieved using volume weighted Am-241 point source measurements in soft waste (6.06 %) [1]. A blank NPL source, with no added Am-241, gave no net counts above background.

DISCUSSION

The best detection levels were achieved with synthetic materials, like PVC, with low net cps above the Cronos background and low net cps variation (σ). Natural materials gave much higher net cps and this was attributed to the presence of NORM such as K-40, Ac-228, and Bi-214 etc. However, the high ambient levels of NORM in the facility precluded verification of this theory by using HRGS to measure NORM levels in the blank materials. Also the Cronos counting efficiency for NORM is high, due to the high yield/high energy photon emissions from NORM. Hence low levels of NORM (undetectable by HRGS) would have a large effect on the Cronos background.

The highest blank results were noted for orange cotton coveralls where 4 net cps/kg equates to an apparent Pu or HEU activity of (4/0.006 x 1000) = 0.67 Bq/g compared to 0.016 Bq/g for PVC. This highlights the importance of segregating waste materials into those having similar net cps.

The waste must also be present as low bulk density, low Z composition due to the difficulties in calibrating Cronos for high density, high Z objects [1].

The background σ, achieved for 600 s counting (3.8-16.3 cps), was much greater than the square root of the background ($\sqrt{B}$ = ($\sqrt{1800x600}$)/600 = 1.7 cps). Also, increasing count time from 60s to 600s gave little reduction in σ compared to the $\sqrt{10}$ = 3.2 x reduction in $\sqrt{B}$ cps. This highlights the importance of locating Cronos in an area of low and non-fluctuating photon background. High energy photons from NORM (e.g. K-40 in building materials) and cosmic radiation can penetrate even two inches of the Cronos lead shielding and generate lower energy photons that increase the background σ. Locating Cronos within a shielded room, as used for in-vivo monitoring equipment, would be a costly, but potentially very beneficial option.

Isotopic fingerprint must be consistent for the waste being assayed because of the different counting efficiencies achieved with different nuclear materials. However, the most commonly encountered Pu and HEU compositions have similar counting efficiencies (around 0.006 cps/Bq).

For Pu wastes the only significant photon emitter is Am-241. Hence Pu calibration is achieved using Am-241 sources and the Pu/Am-241 activity ratio. Provided that this ratio is relatively low (i.e. < 10) exempt waste clearance is feasible. However, some Pu compositions, with low Am-241 content, may only be measured at the VLLW threshold (4 Bq/g) and not the exempt threshold (0.15 Bq/g).

For U wastes the 1 Bq/g exempt threshold is easier to achieve, particularly for DU. However, it is important to use the full 35 kg counting chamber capacity, for low bulk density waste, in order to get the best Bq/g detection levels.

CONCLUSIONS

Cronos has the performance to assay waste at the new clearance thresholds, of 1 Bq/g for U contaminated wastes and 0.15 Bq/g for Pu contaminated wastes, provided that:

- Cronos is located in a area with a low/non-fluctuating photon background.
- Wastes are well segregated according to similar isotopic fingerprint, similar and low photon attenuating properties and similar and low NORM content.
- The counting chamber is filled with around 35 kg of evenly distributed waste.
- Pu/Am-241 activity ratios do not exceed 10.
- The database of background counts for uncontaminated materials covers all material types and batches encountered.

REFERENCES

1. T. Miller, Waste Assay at the Free Release Threshold Using a Box Monitor, *Proceedings of Waste Management Symposia 36th Annual Meeting*, paper 10007, Phoenix, Arizona, USA, March 7-11, 2010.

2. T. Miller, Application of NPL Radioactive Waste Package Standards at AWE, *Proceedings of Waste Management Symposia 37th Annual Meeting*, paper 11004, Phoenix, Arizona, USA, February 27-March 3, 2011.