This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-96SR18500 with the U. S. Department of Energy.

Measured HEU Content in 110-Gallon Overpacks and Partly Filled Drums by Far Field γ-PHA Assay

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ABSTRACT

The 321-M Reactor Fuel Fabrication facility was used to fabricate enriched uranium fuel assemblies, lithium-aluminum target tubes, neptunium assemblies, and miscellaneous components for the production reactors. It operated for 25 years. During this time, thousands of uranium-aluminum-alloy (U-Al) fuel tubes were produced. After the facility terminated operations in 1995, all of the easily accessible U-Al was removed from the building, and only residual amounts remained. The Analytical Development Section of SRTC was tasked to conduct the required assays of highly enriched uranium in the deactivation and decommissioning activities of the facility. In this report we demonstrate successful assay of thirteen 55-gallon and 110-gallon drums in the far field transmission corrected point source acquisition configuration. Several of the drums were assayed using the commercial Q^2 instrument in both the direct mode and in the segmented gamma scanner mode adapted by SRTC in addition to the far field method described here. The 110-gallon drums would not fit into the Q^2 instrument, and so assay by the far field technique was necessary. While the far field transmission corrected method used for these drums is a straightforward technique, it was important to obtain comparison measurements between it and the Q^2 techniques to establish its accuracy for the 110-gallon overpack drums.

INTRODUCTION

The 321-M Reactor Fuel Fabrication facility was used to fabricate enriched uranium fuel assemblies, lithium-aluminum target tubes, neptunium assemblies, and miscellaneous components for the production reactors. It operated for 25 years. During this time, thousands of uranium-aluminum-alloy (U-Al) fuel tubes were produced. After the facility terminated operations in 1995, all of the easily accessible U-Al was removed from the building, and only residual amounts remained.

Prior to the start of the dismantling and removal of the HEU, loose items and small items were placed in 55-gallon drums for shipment to low level Solid Waste.[1] A large number of drums contained low to medium density materials, and many contained high density items. The materials in the drums with high density waste included scrap metal, metal hand tools, power tools, motors, pumps, toolboxes, computer parts, gages, etc. The high density drums were assayed for HEU content using the SRTC adapted Q^2 technique that is described in reference 2.

Each low level waste package presented for disposal in the E-Area Vaults must meet the radionuclide limits specified in WSRC 1S WAC, 3.17.[(3)] WSRC 1S, WAC 2.02[(4)] specifies acceptable methods for quantifying waste package activity. Among the acceptable methods are Dose-to-Curie, direct sampling, and direct assay. Examples of acceptable assay equipment include use of the Canberra Q^2 system as specifically cited in WAC 2.02. Using the assay results, together with a known radiological distribution, the generator can quantify waste package isotopic activity and compare against WSRC 1S, WAC 3.17 limits to certify compliance with the radiological criteria.[3]

All the drums containing low to medium density materials were assayed using the 315-M Canberra Q^2 Waste Assay System. This report discusses far field transmission corrected γ-PHA measurements performed on three 110-gallon overpack drums that were too large for assay in the Q^2 instrument. It also includes far field γ-PHA assay of four partly filled 55-gallon drums that were unsuitable for assay in the Q^2 due to the non-uniform fill. The assays were performed in a manner identical to the far field transmission corrected measurements using the permanent scrap can assay station.[5]

The report uses comparison measurements on six high-density drums to verify the measurements on the overpack and partly filled drums. The six high-density drums were measured using the direct Q^2 system, using the adapted Q^2 system, and using the far field measurements of this report. The comparison measurements demonstrate excellent agreement, and confirm the good science of the far field acquisitions.

EXPERIMENTAL

Because the three 110-gallon overpack drums did not fit into the Q^2 assay chamber, we had to assay them using a far field transmission corrected γ-PHA configuration. We used the same acquisition configuration to assay the four partly filled 55-gallon drums since the Q^2 instrument was not qualified to assay drums that are less than 100% full. Since those acquisitions in July 2001, Casella and Dewberry have performed the necessary development work to qualify the Q^2 for partly filled drums.[6] But in July 2001 each drum was assayed from a distance of 54 inches using the high purity germanium (HPGe) detector that was energy and efficiency calibrated in WSRC-TR-2000-00317.[(7)] We used the 4.41-g HEU source wt2025a for the transmission correction measurements.[8]

The seven drums were assayed in the traditional far field configuration using equation (1).

$$[\text{U-235}] = (2.36\times10^{-5})(\text{cps})(\text{d})^2(\text{Cf}_T), \qquad (1)$$

where the first term is the far field point source calibration constant in units of g-sec/cm^2, and the distance term is in units of cm. The last term is the transmission correction factor that is explained in references 5, 9, and 10. We obtained daily QC checks of the system using the Cs-137 check source SRM4207-59 as prescribed by reference 11. The far field data we acquired are summarized in Table 1. The transmission correction factor (Cf_T) for high-density drum 7989 was calculated in the far field configuration by equation (2).

Cf_T(7989) = SQRT{cps(T0)/[cps(7989_T) – cps(7989)]}.

$$Cf_T(7989) = \sqrt{cps(T0)/\left[cps(7989\_T) - cps(7989)\right]}$$

$$= \sqrt{3.45/\left[\left(533/300\right) - \left(32/300\right)\right]} = 1.44. \qquad (2)$$

The unabsorbed count rate in (2) was determined at a distance of 90.5 inches and was 3.45 cps. Note this unabsorbed spectrum also qualifies as a QC check of the system. Using this data and applying (1), we measured an HEU content of 4.30 g for the source.

The acquisitions were obtained on the 313-M loading dock. A photograph of a typical acquisition is shown in Figure 1. We had a turntable specially built for these acquisitions to accommodate the 110-gallon drums (as well as a standard 55-gallon drum) so that the detector could obtain an isotropic time-averaged view of each drum. A background spectrum is shown in Figure 2, the Cs-137 QC check spectrum is shown in Figure 3, and typical sample γ-PHA spectra are shown in Figures 4 and 5. Figures 2 – 5 appear at the end of this paper.

We also obtained far field transmission corrected acquisitions of the six high-density drums and of the source-only drum listed in Table 2. These data were acquired at a source to detector distance of 54 inches to obtain a comparison between the far field measurements and the two types of Q^2 measurements obtained on these high-density drums. These comparisons were necessary to validate the far field measurements made on the overpack drums and partly filled drums. The direct Q^2 measurements and adapted Q^2 measurements for these drums are reported in references 12 – 14.

Table 1. Far field data acquired for the three overpack drums and four partly-filled high density drums.

Date	Drum Number	t_T = CT (sec) 4.41g U + Drum	C_T = counts 4.41g U+Drum	σ_T = count error 4.41g U + Drum	t_C = CT (sec) Drum	C_C = counts Drum	σ_C Drum
7/11/2002	7989	300	533	27	300	32	10
7/11/2002	7990	300	667	28	300	34	10
7/11/2002	7991	300	524	24	300	14	7
7/11/2002	7992	300	533	26	300	17	7
7/11/2002	8069	300	690	28	300	56	9
7/11/2002	8070	300	729	28	300	-4	11
7/11/2002	8071	300	556	24	300	74	10

S_T (4.41g Uranium Standard transmission @90.5 in) =	3.45 cps

Figure 1. Photograph of the gamma PHA setup.

The measured U-235 contents on these seven drums were determined using equation (1). The transmission correction values were obtained using a weighted average of the transmission correction factors obtained for each of the three drum segments in the adapted Q^2 measurements. That is, the correction factor for each segment was determined by the technique described in reference 2. Then each segment was weighted by the measured activity coming from that segment. The weighted correction factor for each drum was determined by equation (3).

$$\mathbf{CfT_{weighted}} = \sum CF(i)cps(i) \Big/ \sum cps(i), \qquad (3)$$

where the sum is over each segment i = 1,2,3. For drum 2350 this yields

$$CfT_{weighted} = \{(2.77)(4.33) + (7.26)(4.90) + (5.83)(3.72\}/\{4.33 + 4.90 + 3.72\} \quad (3)$$

$$= 5.35.$$

Table 2. Far Field Acquisitions for the six high-density drums counted in November 2001.

Drum	Counts	sigma	Count Time	d	cps
01 Nov BG	-33	28	599.52		-0.055
Nov 01 BG	56	45	1215.92		0.046
BG Sum	-10	61	1815.92		-0.0055
FD2350	196	25	299.72	54	0.654
FD2398	3359	71	385.4	54	8.72
FD2092	1729	50	299.68	54	5.770
CSQC11-15	2816	55	59.68	54	47.18
wt2025	1323	41	159.7	54	8.284
FD2082	1729	50	299.68	54	5.770
FD2382	10958	127	530.42	54	20.659
FD2275	4223	78	303.56	54	13.91

RESULTS and DISCUSSION

The measured values for each of the three 110-gallon overpacks and for the four partially filled 55-gallon drums are listed in the spreadsheet of Table 3. Note that all seven items have measured transmission correction values in the range (1.19 – 1.47) and that all seven have measured HEU contents near our sensitivity limit of 0.1 g. The 110-gallon overpacks 8069 and 8071 have finite contents of 0.15 ± 0.08 g and 0.23 ± 0.10 g. The transmission correction values demonstrate that none of these seven drums is strongly absorbing of the 185 keV γ-ray photon. By itself, that lends very good credibility to the results. The remainder of this section contains a comparison of measured values for the six high-density drums that were assayed by the direct Q^2 method, by the adapted Q^2 method, and by the far field point source technique of this report. We include this comparison and discussion to present a measure of the accuracy of the far field, point source technique when measuring drums. The far field measurements are made using the Excel spreadsheet that we developed for the 321-M scrap cans.[5] This spreadsheet calculates the transmission corrected point source content from a far field acquisition using equation (1).

$$[\text{U-235}] = (2.36\times10^{-5})(\text{cps})(d)^2(Cf_T). \qquad (1)$$

The results for all of the far field measurements on the six high-density drums are listed in Table 4. Also in Table 4 we list results for the 4.41-g HEU standard that was counted in the far field configuration inside the standard Q^2 empty drum. All of these results lend excellent support to the far field measurements. The contents of each drum were calculated with the transmission corrected far field point source equation (4).

$$[\text{U-235}] = (2.36\times10^{-5})(\text{cps})(54\times2.54)^2(Cf_T), \qquad (4)$$

where all of the terms have the same meaning as equation (1), and the acquisition distance is 54 inches.

The Q^2 source spectrum that was acquired with the 4.41-g HEU standard inside of the empty Q^2 standard drum yielded an excellent QC check of the system with a measured value of 4.49±0.23 g. For this spectrum we used the transmission correction factor for the empty drum. We measured this value of 1.22±0.05 in reference 2. The correction factor (column 3) for each of the six drums was taken as a weighted average of the correction factor measured individually for each segment when the drums were assayed with the adapted Q^2 technique. These values are recorded in the Excel spreadsheet NEWSS on the hard drive Hard1 of one author's (RAD) computer and in reference 15.

Table 3. Spreadsheet for T, T-correction, and results of the overpacks and partially filled drums.

Date	Drum Number	t_T = CT (sec) 4.41g U + Drum	C_T = counts 4.41g U+Drum	σ_T = count error 4.41g U + Drum	t_C = CT (sec) Drum	C_C = counts Drum	σ_C Drum	Trans Corr	M_U = ^{235}U (grams) Drum	$2\sigma_U$ = M_U Error Drum
7/11/2002	7989	300	533	27	300	32	10	1.44	0.10	0.112
7/11/2002	7990	300	667	28	300	34	10	1.28	0.09	0.088
7/11/2002	7991	300	524	24	300	14	7	1.42	0.04	0.094
7/11/2002	7992	300	533	26	300	17	7	1.42	0.05	0.099
7/11/2002	8069	300	690	28	300	56	9	1.28	0.15	0.084
7/11/2002	8070	300	729	28	300	-4	11	1.19	-0.01	0.081
7/11/2002	8071	300	556	24	300	74	10	1.47	0.23	0.104

S_T (4.41g Uranium Standard transmission @90.5 in) = 3.45 cps.
Transmission Correction = SQRT(S_T/$Drum_T$),
where:$Drum_T$ = ((C_T/t_T) - (C_C/t_C)).
M_U (grams ^{235}U) = (C_C/t_C)(2.36E-05)$(90x2.54)^2$(SQRT(S_T/$Drum_T$)),
where:Can_T = ((C_T/t_T) - (C_C/t_C)).
$2\sigma_U$ = (M_U)(2.0)SQRT((SQRT$((\sigma_T/C_T))^2+(\sigma_C/C_C))^2$))/$(C_C/t_C))^2$+SQRT$(7/236)^2$+SQRT$(0.112)^2$).

Note in Table 4 the values of HEU content listed in column four are in very good agreement with the adapted Q^2 results of column six. Agreement with the direct Q^2 results of column five are fairly good, but not as exact. Note further that especially for drums FD2398 and FD2092 the agreement between columns four and six is very good. These two drums contain the gearbox (item 1891) and electric motor (item 1890) that we assayed in a holdup configuration with the billet assay station. Results were reported in reference 9 for these two items. In reference 9 we reported contents of (6 – 25 g HEU) in the gearbox and (26 ± 26 g HEU) in motor M28619. Agreement between these three redundant acquisition configurations was one of the important points that we made in the external summary paper presented on the 321-M assay measurements.[10]

Table 4. Results for the six high-density drums counted in November 2001 by far field acquisitions.

Drum	cps (far field)	Transmission Cf	HEU far field	direct Q^2 result	adapted Q^2 result	Drum mass
source	8.285(257)	1.22(5)	4.49(23)			
FD2275	13.91(26)	5.51(11)	34.0(9)	12.4	30(10)	62
FD2398	8.716(184)	4.76(23)	18.4(10)	30.3	17(7)	298
FD2082	5.769(167)	1.97(0)	5.04(15)	5	2.87(156)	152
FD2382	20.569(239)	3.05(5)	28.0(6)	21.7	30(10)	70
FD2350	0.654(83)	5.35(34)	1.55 (24)	0.78	1.17(56)	123
FD2092	5.771(167)	3.43(15)	8.79(46)	16.7	8.68(270)	245

CONCLUSION

With this report we demonstrate successful assay of thirteen 55-gallon and 110-gallon drums in the far field transmission corrected point source acquisition configuration. Several of the drums were assayed using the commercial Q^2 instrument in both the direct mode and in the segmented gamma scanner mode adapted by SRTC in addition to the far field method described here. The 110-gallon drums would not fit into the Q^2 instrument, and so assay by the far field technique was necessary. While the far field transmission corrected method used for these drums is a straightforward technique, it was important to obtain comparison measurements between it and the Q^2 techniques to establish its accuracy for the 110-gallon overpack drums.

The important feature in the comparisons involved obtaining good transmission correction measurements. The results for the comparison measurements between the far field technique and the segmented Q^2 technique are very favorable. The measured contents of the six drums by the far field measure all agree within 2 grams or within 13% (whichever is greater) of the adapted Q^2 measurement. Note these results are over a very large mass range of 62 kg to 298 kg. We believe this demonstrates very good credibility for the far field measurements of all thirteen drums, including the three 110-gallon overpacks and the four partly filled high-density drums.

In previous reported results ADS has requested a Cf-shuffler neutron measurement be performed on several of these drums. This request is based on the extreme high density of some of these drums, which have yielded significant transmission correction factors of up to 50 with measured HEU contents of up to 60 grams. Correction factors of this magnitude for γ-ray measurements strongly suggest the use of neutron activation techniques for SNM assay. Since measured values are available from these γ-PHA measurements, such a comparison would yield especially valuable data.

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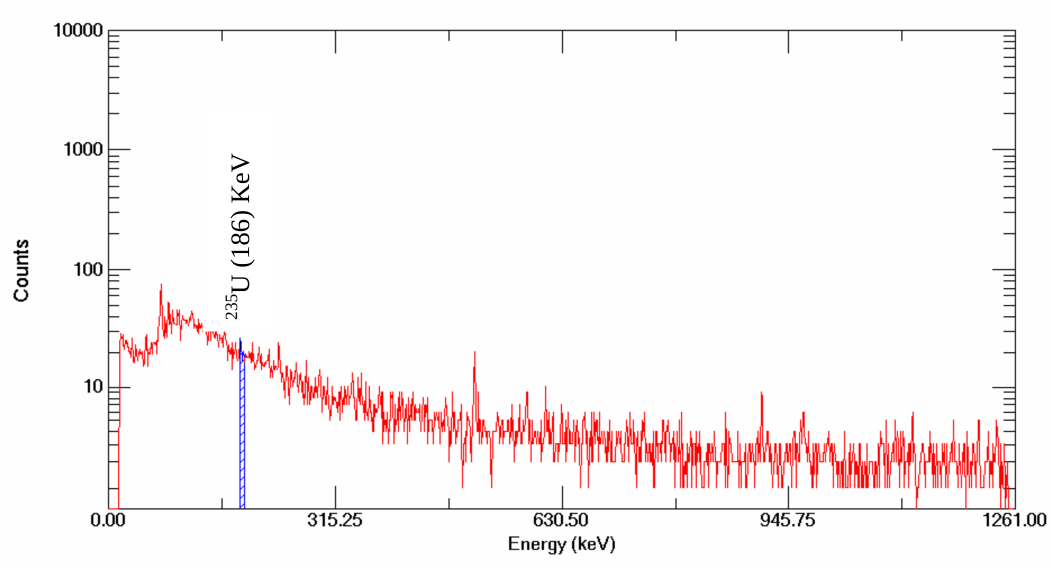

Figure 2. Background Spectrum.

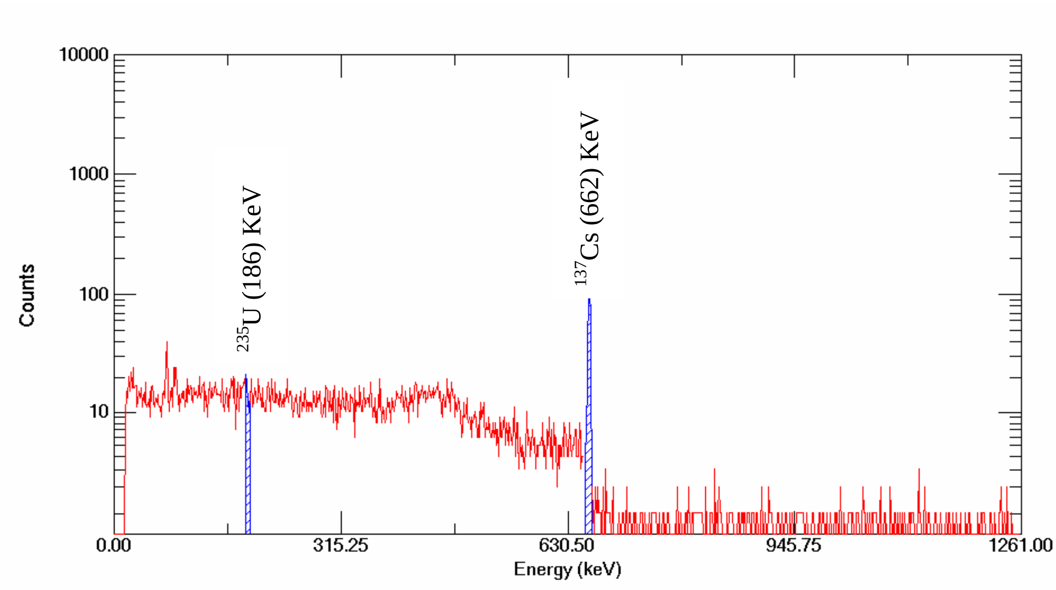

Figure 3. QC Check Spectrum.

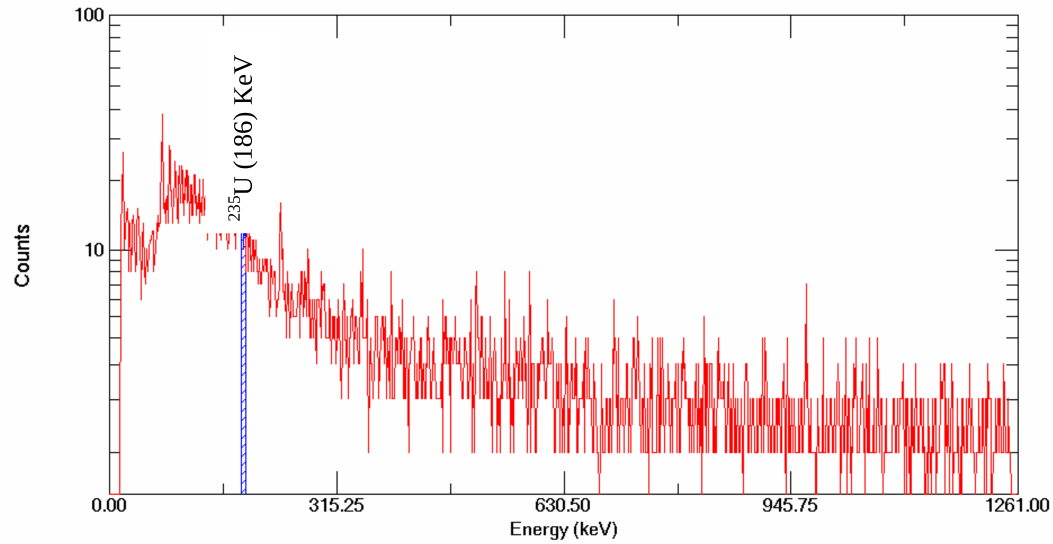

Figure 4. Spectrum 7989.

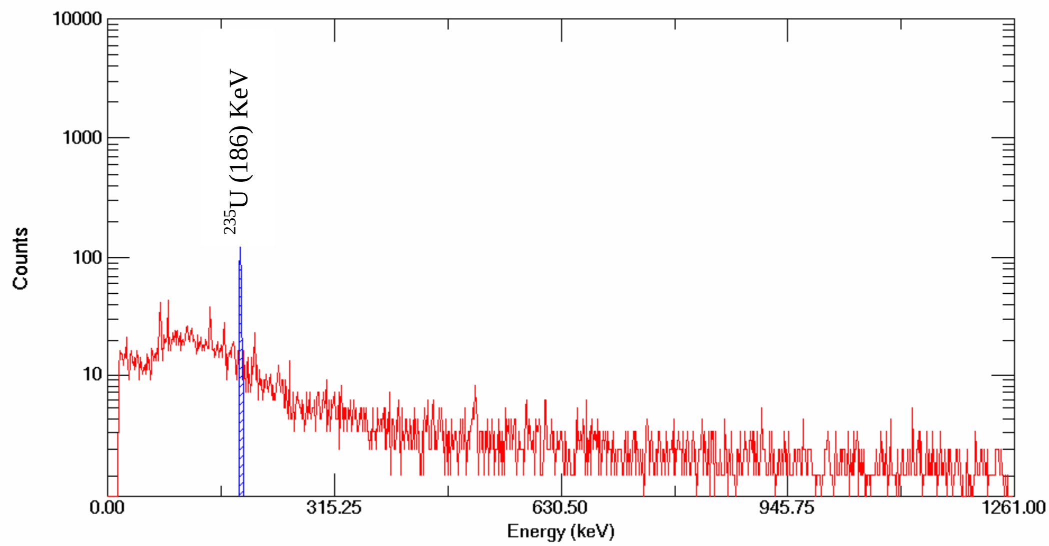

Figure 5. Spectrum 7989-T.