*Design and Calibration of the AWCC for
Measuring Uranium Hexafluoride*

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DESIGN AND CALIBRATION OF THE AWCC FOR MEASURING URANIUM HEXAFLUORIDE

by

T. R. Wenz, H. O. Menlove, G. Walton, and J. Baca

ABSTRACT

An Active Well Coincidence Counter (AWCC) has been modified to measure variable enrichment uranium hexafluoride (UF_6) in storage bottles. An active assay technique was used to measure the ^{235}U content because of the small quantity (nominal loading of 2 kg UF_6) and nonuniform distribution of UF_6 in the storage bottles. A new insert was designed for the AWCC composed of graphite containing four americium-lithium sources. Monte Carlo calculations were used to design the insert and to calibrate the detector. Benchmark measurements and calculations were performed using uranium oxide samples. The Monte Carlo generated calibration curves benchmarked to uranium oxide resulted in assay values that agreed within 2 to 3% of destructive assay values. In addition to UF_6 , the detector was also calibrated for HEU ingots, billets, and alloy scrap using the standard Mode 1 end-plug configuration.

INTRODUCTION

The Active Well Coincidence Counter¹ (AWCC) has been modified to measure the uranium content of highly enriched uranium hexafluoride (UF_6) stored in cylindrical bottles. The cavity height in an AWCC with standard end plugs is 20 cm. The UF_6 storage bottles are more than 50 cm long and the active length in the bottles is roughly 40 cm. This measurement required designing new inserts for an existing AWCC to optimize the counting response over the active region of the storage bottles. The detector with the new inserts was calibrated primarily with Monte Carlo calculations. Supplemental calibration measurements were also performed using uranium oxide and uranium metal samples as applicable. The difficulty in obtaining and fabricating UF_6 calibration samples limits the number of experimental measurements that can be made. We have developed techniques to make the AWCC measurements relatively insensitive to the heterogeneous distribution of the UF_6 .

In addition to measuring UF_6 , the same detector will also be used to measure the fissile content of uranium billets, uranium alloy scrap, and high-enriched uranium (HEU) ingots. These measurements will be made in the AWCC using the standard end plugs in the Mode 1 configuration,² which has a cavity height of 22 cm. The general design of the end plugs will be reviewed based on the information obtained from the design of the new insert to see if the end plugs can be

further optimized (see Appendix A). The AWCC was calibrated for the additional material types by using both Monte Carlo calculations and experimental measurements.

MONTE CARLO CALCULATIONS FOR CYLINDER INSERT DESIGN

The AWCC inserts were designed using the Monte Carlo transport code MCNP³ with the ENDF/B-V nuclear data file.⁸ Three principal criteria were used in designing the UF₆ inserts for the AWCC: (1) achieve a uniform detector response over the active region of the UF₆ bottles, (2) increase the overall response of the detector per gram of material measured, and (3) minimize the error associated with the measured real rate.

The UF₆ cylinders have an overall length of approximately 50 cm, excluding the cylinder cover, and an outer diameter of ~10 cm. The bottles have a metal flange with an outer diameter of ~12 cm. This flange diameter is the maximum radial dimension of the bottle. The length of the cylinders prevents the use of an end plug at the top of the cavity, but a graphite end plug is used in the bottom for physical placement of the cylinder. The UF₆ is deposited along the inner surfaces of the cylinders and is assumed to gather predominantly in the lower end of the cylinder rather than the upper end. As a result, the bottom end plug was also designed to increase the measurement efficiency at the lower end.

Graphite and polyethylene were considered for making the insert. Calculations were made for cases in which the cylindrical inserts were composed of graphite, polyethylene, and a mixture of the two materials. The absolute real-count rates for each of the insert designs are shown in Fig. 1. The count rates are plotted as a function of location in the detector and are based on a fixed source strength from four americium-lithium sources. Four americium-lithium sources were used in the design of the insert to give a relative uniform coverage over the UF₆ active region without using a scanning procedure. The "graphite-poly band" design consists of two 8.4-cm-thick rings of polyethylene replacing graphite at the locations of the inner two americium-lithium sources. This is a multisectional insert design with alternating layers of graphite and polyethylene. In the "graphite-poly cylinder" case, the insert was composed of graphite and the americium-lithium sources were placed in polyethylene cylinders approximately 0.5 cm thick. This configuration resulted in the highest response and the lowest statistical error; therefore, it was chosen for the final design.

Figure 2 is a diagram of the AWCC with the final insert design in place. The insert is made of graphite and accommodates four americium-lithium sources encapsulated in polyethylene cylinders. The graphite increases the measured response over the polyethylene case because of fewer neutron absorption events in the moderator. The polyethylene cylinders around the sources lower the neutron energy spectrum to further increase the fission rate in the sample. As indicated in Fig. 2, the polyethylene thickness of the lower right americium-lithium source has been reduced to lessen the efficiency of this source to further flatten the response profile. The sample cavity is lined with cadmium (0.5-mm-thick) to eliminate thermal neutrons from the interrogation.

A graphite reflector is located at the bottom of the detector to vertically position the active region of the cylinder and to increase the counting efficiency at the lower end of the cylinder. An aluminum plate with a polyethylene liner rests on top of the graphite plug to center the cylinder radially in the counter. A removable cadmium liner is on the inner surface of the graphite insert. A flange at the top of the liner (shown in Fig. 2) aids in moving the liner.

Fig. 1. Comparison of the real-count rates for various UF_6 cylinder insert designs from Monte Carlo calculations.

The Monte Carlo calculations were used to determine the vertical positions of the four americium-lithium sources that would provide a uniform axial response to the UF_6 . The axial detector response was calculated for a cylindrical UF_6 sample 5 cm tall, with a 9-cm diameter, containing 650 g of UF_6 (410 g ^{235}U). These profiles are shown in Fig. 3 for both reals and totals responses with (Fig. 3a) and without (Fig. 3b) the cadmium liner. The design objective was to obtain a fast-neutron response profile (for the case with the cadmium liners) that was within $\pm 10\%$ of the average and to maximize the counting efficiency at the bottom of the cylinder where much of the UF_6 is expected to condense. Figure 4 compares the reals response with the materials-testing-reactor (MTR) fuel element insert² and the newly designed insert. The bar at the top of this figure represents the location of the UF_6 in the cylinders.

PRECALIBRATION CALCULATIONS AND MEASUREMENTS

In addition to calibrating the AWCC for UF_6 with the new insert, we also calibrated it for three additional material categories: HEU billets, HEU aluminum alloy scrap, and HEU ingots. Figure 5 is a diagram of the AWCC with the standard end plugs in the Mode 1 configuration; this configuration will be used for the three additional material categories. In this configuration, a 2.5-cm polyethylene disk was removed from the bottom end plug to increase the sample cavity height by the same amount.

The detector was calibrated by performing Monte Carlo runs simulating each of the material categories to calculate the coincidence rates so that calibration curves could be generated. Next, measurements were made with the AWCC in the two detector configurations with samples that were similar to the measurement categories. Cans of uranium oxide were used as representative

Fig. 2. Cross sectional drawing of the AWCC with the graphite insert for the UF_6 cylinder.

samples for UF_6 , billets, and aluminum alloy scrap. Uranium metal disks were used as representative samples for the two types of ingots. The measured count rates were then compared with the rates calculated using the Monte Carlo code to model the measurement. The calculated rates tended to be biased high by roughly 33%, so the calibration curves were renormalized to account for this effect. The measurement data are presented later in this report. The figures and tables presented in this section contain the Monte-Carlo-generated calibration curves renormalized to the experimental data for uranium oxide and metal as appropriate.

Highly Enriched UF_6

We chose an active assay technique using the AWCC to assay the quantity of ^{235}U in UF_6 . This form of uranium has the added feature of possessing an internal neutron source. The neutrons are produced from an $(\alpha\text{-n})$ reaction in which α particles formed from the decay of ^{234}U interact with fluorine to produce neutrons. Appendix B discusses the option of passive counting and the self-interrogation⁵ of the UF_6 along with the reasons passive counting was not used here,

Fig. 3. Axial response profiles for the AWCC with the graphite insert for the UF₆ cylinder for (a) fast and (b) thermal modes from Monte Carlo calculations.

Fig. 4. Comparison of the axial response profiles for the AWCC with the old MTR insert and the new UF₆ insert for fast mode operation from Monte Carlo calculations.

primarily because of large statistical errors and the nonuniform sample distribution in the storage bottles.

Two types of bottles are used to store the UF₆. The geometry differences between the two bottle designs are minimal; as a result, the calibration curves generated for UF₆ are applicable to either bottle. Both bottle designs contain internal fins where the UF₆ resides; for the purpose of

Fig. 5. Cross sectional drawing of the AWCC with the standard end plugs in the Mode 1 configuration.

Monte Carlo calculations, the fins and UF_6 were treated as a homogeneous mixture. The calibration curve was obtained by adjusting the fill height of the UF_6 to obtain the various mass loadings assuming a meniscus depth of approximately 5 cm. The calibration data were calculated for both intermediate and high enrichments. Figure 6 shows the calibration curves for both enrichments with cubic curve fits and the corresponding coefficients. Note that the lower enrichment data have a higher response per gram ^{235}U than the high enrichment. This effect appears to be the result of a corresponding reduction in neutron self-shielding.

The Monte Carlo generated calibration curves for the high and intermediate uranium enrichment differ from each other by ~10-15% depending on the ^{235}U mass value. The reason for the difference is the increase in neutron self-shielding for the higher enrichments. For implementation, the UF_6 enrichment can cover the range from ~20% to 90%. To better match the calibration curves

Fig. 6. Precalibration curves for UF_6 in the AWCC with the graphite insert at enrichments of 30% and 80%.

to the enrichment being measured, we have used a linear extrapolation of the two calibration functions shown in Fig. 6 to match intermediate enrichment values. We assumed that the measured response at a new enrichment is a linear extrapolation between the calculated responses at the two enrichments. This approach works well as long as the extrapolation is small, but it should not be used for large steps in calibration functions because the neutron self-shielding effect is inherently nonlinear. Using this extrapolation approach, we generated additional calibration functions at enrichments of 20% and 50% to supplement the MCNP curves at 30% and 80%. The additional calibration coefficients are given in Appendix D.

With the active assay technique, the calculated response in Fig. 6 has two components. One is from induced fission caused by the internal (α -n) neutrons and the other component is from the induced fission caused by the americium-lithium neutron sources. Table I lists the percentage each component contributes to the total based on Monte Carlo calculations. The final calibration includes both the active and passive (self-interrogation) components and no attempt is made to separate these two components.

The renormalization of the real-time data calculated from Monte Carlo to the benchmark uranium oxide measurements takes the following form:

$$R_{UF_6}^{pre-cal} = R_{UF_6}^{mcnp} \cdot \left[\frac{R_{UO_2}^{meas}}{R_{UO_2}^{mcnp}} \right]$$

HEU Ingots

Calibration curves for two types of uranium metal ingots are presented in this section. In these calculations, the AWCC is in the Mode 1 configuration and both ingot types are placed on top of a 5-cm-tall can. The first type is a right circular cylinder whose height is adjusted to achieve the various mass loadings. The base position is always the 5-cm-tall can. A cubic equation with a zero intercept was fitted through both sets of enrichment data and the resulting coefficients are included in Fig. 7. The intermediate-enrichment data do not extend beyond 7 kg because the cylinder height would exceed the sample cavity height.

The high-enrichment calibration curve is substantially above the intermediate-enrichment curve for the high ^{235}U mass range shown in Fig. 7. This is because the multiplication effect is larger than the neutron self-shielding effect for the HEU metal ingots with mass values above 3 kg. For the ^{235}U mass range below 1 kg, the neutron self-shielding effect dominates and for ^{235}U mass values between 1-3 kg, the two effects almost cancel each other.

The second ingot shape consists of a small-diameter right cylinder with a flat base at one end. The calibration curve was obtained by adjusting the length and diameter of the ingot to obtain the various mass loadings. The calibration data were calculated at intermediate and high enrichments. Figure 8 shows the calibration data for this sample with cubic curve fits and the corresponding coefficients. For fissile masses below 800 g, the intermediate-enrichment data behaves like the high-enrichment data; as a result, only a single calibration curve is required. We assumed that the high-mass samples correspond to the high-enrichment material, consequently a 0 to 800-g mass range is sufficient for the intermediate-enrichment material.

TABLE I. Passive and Active Contributions to the Reals Rate from Active Assay of UF_6					
30% enrichment			80% enrichment		
^{235}U (g)	passive ^a (%)	active ^b (%)	^{235}U (g)	passive ^a (%)	active ^b (%)
362.3	3.3	96.7	408.4	6.4	93.6
561.8	4.2	95.8	749.3	9.9	90.1
762.0	5.2	94.8	966.1	11.6	88.4
962.1	6.0	94.0	1132.8	13.0	87.0
1163.3	6.8	93.2	1292.0	13.5	86.5
1362.5	6.9	93.1	1469.9	14.6	85.4
1571.2	7.0	93.0	1676.0	15.9	84.1
			1882.0	17.2	82.8
			2116.2	18.4	81.6

^aThe passive reals rate comes from the $^{234}\text{U}(\alpha, n)$ induced fissions in ^{235}U .

^bThe active reals rate comes from the americium-lithium (α, n) and $^{234}\text{U}(\alpha, n)$ induced fissions in ^{235}U .

Fig. 7. Precalibration curves for cylindrical uranium metal ingots in the AWCC (Mode 1) at enrichments of 46% and 90%.

Fig. 8. Precalibration curve for uranium metal finger ingots in the AWCC (Mode 1) at enrichments of 46% and 90%.

The renormalization of the real-time data calculated from Monte Carlo to the benchmark uranium metal measurements takes the following form:

$$R_{\text{ingot}}^{\text{pre-cal}} = R_{\text{ingot}}^{\text{mcnp}} \cdot \left[\frac{R_{\text{u}}^{\text{meas}}}{R_{\text{u}}^{\text{mcnp}}} \right], \text{ where } R_{\text{u}}^{\text{meas}} = R_{\text{uo}_2}^{\text{meas}} \cdot \left[\frac{R_{\text{u}}^{\text{meas}}}{R_{\text{uo}_2}^{\text{meas}}} \right]_{\text{LANL}}$$

HEU Billets

The billets are composed of a uranium-aluminum alloy in the shape of rectangular parallelepipeds. The calibration data were generated for intermediate and high enrichments. The various mass loadings were obtained by adjusting the ratio of uranium to aluminum. Figure 9 shows the resulting calibration curve for a 20-cm by 8-cm by 2.5-cm billet and Fig. 10 is the data for a 25-cm by 8-cm by 2.5-cm billet. For fissile masses below 800 g, the intermediate-enrichment data behave like the high-enrichment data; as a result, only a single calibration curve is required. In the case of the latter (longer) billet, the geometry of the detector has to be extended to accommodate the added height. The extended Mode 1 geometry used for the longer billets is set up by placing wood spacers on top of the nickel ring so that the top end plug sets higher (~ 3 cm) than in the normal Mode 1 configuration. Both detector configurations use the same aluminum sample insert, which places the billet at a 25° angle for better source-sample coupling; a can to raise the sample is not used because of the tall sample height.

Fig. 9. Precalibration curve for 20-cm-long uranium billets in the AWCC (Mode 1) at enrichments of 46% and 90%.

Fig. 10. Precalibration curve for 25-cm-long uranium billets in the AWCC (Ext. Mode 1) at enrichments of 46% and 90%.

The renormalization of the reals-rate data, calculated with Monte Carlo methods, to the benchmark uranium oxide measurements takes the following form:

$$R_{\text{billet}}^{\text{pre-cal}} = R_{\text{billet}}^{\text{mcnp}} \cdot \left[\frac{R_{\text{uo2}}^{\text{meas}}}{R_{\text{uo2}}^{\text{mcnp}}} \right].$$

HEU Aluminum Alloy Scrap

The aluminum alloy scrap consists of uranium and aluminum pieces pressed into the shape of a right circular cylinder. For the Monte Carlo calculations, the mixture of material is assumed to be homogeneous. The cylinders are placed on top of a 5-cm can in the detector. Only a single calibration curve is needed for various enrichments and U/Al ratios because of the dilution of the fissile material by the aluminum. Figure 11 shows the resulting calibration curve for this material with a cubic curve fit and corresponding coefficients.

The renormalization of the reals-rate data, calculated with Monte Carlo methods, to the benchmark uranium oxide measurements takes the following form:

$$R_{\text{scrap}}^{\text{pre-cal}} = R_{\text{scrap}}^{\text{mcnp}} \cdot \left[\frac{R_{\text{uo2}}^{\text{meas}}}{R_{\text{uo2}}^{\text{mcnp}}} \right].$$

Fig. 11. Precalibration curve for cylindrical uranium-aluminum alloy scrap in the AWCC (Mode 1) for variable enrichments.

Table II summarizes the calibration information for each material type presented in this section.

FINAL CALIBRATION CALCULATIONS

Final calibration parameters will be generated using the data in Table II after facility standards, authenticated by the IAEA, have been measured. In effect, this procedure simply renormalizes the precalibration coefficients in Table II to the specific source and detector that will be used in the actual measurements. For the case of a cubic precalibration curve of the form

$$R = a + bm + cm^2 + dm^3 ,$$

a calibration normalization factor k can be defined as follows:

$$k = \frac{R_{\text{precalibration curve-standard}}}{R_{\text{measured-standard}}} .$$

The numerator is the real rate calculated from the appropriate precalibration curve using the known ^{235}U mass of the standard, and the denominator is the real rate measured in the AWCC with the proper inserts and sources installed. The real rates from the subsequent measurement of

TABLE II. Summary of Precalibration Data for Each Material Type. ^{a,c}															
Material	Detector Config.	Enrich. (%)	Calibration Equation	Calibration Constants ^b				Variances ^b				Covariances ^b			
				a	b	c	d	V _a	V _b	V _c	V _d	a:b	b:c	b:d	c:d
Cylinder Ingots	Mode 1	90	$R = a + bm + cm^2 + dm^3$	0	9.41EE-2	6.46EE-6	2.02 EE-10	-	7.02EE-5	3.78 EE-12	1.15 EE-20	-	-1.58EE-8	8.25 EE-13	-2.06 EE-16
Cylinder Ingots	Mode 1	46	$R = a + bm + cm^2 + dm^3$	0	1.13EE-1	-6.15EE-6	8.68 EE-10	-	2.07EE-5	3.65 EE-12	3.64 EE-20	-	-8.42EE-9	7.98 EE-13	-3.60 EE-16
Finger Ingots	Mode 1	all	$R = a + bm + cm^2 + dm^3$	0	1.17EE-1	-5.91EE-5	2.01EE-8	-	1.59EE-5	5.59 EE-11	1.13 EE-17	-	-2.91EE-8	1.25 EE-11	-2.48 EE-14
Al-Alloy Scrap	Mode 1	all	$R = a + bm + cm^2 + dm^3$	0	1.79EE-1	-6.33EE-5	1.42EE-8	-	2.37EE-5	5.41 EE-11	7.00 EE-18	-	-3.47EE-8	1.18 EE-11	-1.92 EE-14
Billets - 20 cm	Mode 1	all	$R = am/(1 + bm)$	1.73EE-1	5.45EE-4	-	-	1.88EE-5	1.94 EE-9	-	-	1.87EE-7	-	-	-
Billets - 25 cm	Ext. Mode 1	all	$R = am/(1 + bm)$	1.71EE-1	4.69EE-4	-	-	4.04EE-6	3.82 EE-10	-	-	3.84EE-8	-	-	-
UF ₆	Graphite	80	$R = a + bm + cm^2 + dm^3$	0	3.08EE-1	9.78EE-5	-2.17EE-8	-	2.13EE-4	3.97 EE-10	4.24 EE-17	-	-2.84EE-7	8.92 EE-11	-1.28 EE-13
UF ₆	Graphite	50	$R = a + bm + cm^2 + dm^3$	0	4.15EE-1	-4.87EE-6	2.78EE-9	-	-	-	-	-	-	-	-
UF ₆	Graphite	30	$R = a + bm + cm^2 + dm^3$	0	4.86EE-1	-7.33EE-5	1.91EE-8	-	4.71EE-4	1.62EE-9	3.17 EE-16	-	-8.52EE-7	3.60 EE-10	-7.09 EE-13
UF ₆	Graphite	20	$R = a + bm + cm^2 + dm^3$	0	5.22EE-1	-1.08EE-4	2.73EE-8	-	-	-	-	-	-	-	-

^aAmLi sources N009 and N010 assumed for Mode 1 and sources N009, N010, C171, and C174 for Graphite.

^b9.99EE-9 = 9.99×10^{-9}

^cSee Appendix D for explanation of UF₆ data

unknown samples can be renormalized so that the ^{235}U mass can be found from the precalibration curves as follows:

$$R = k \cdot R_{\text{measured}} = a + bm + cm^2 + dm^3 .$$

By renormalizing the calibration coefficients, the measured rate can be directly used in the calibration curve as follows:

$$R_{\text{measured}} = \frac{a}{k} + \frac{b}{k}m + \frac{c}{k}m^2 + \frac{d}{k}m^3 = a' + b'm + c'm^2 + d'm^3 .$$

Similarly, the same normalization factor k can be applied to calibration curves of the form

$$R = \frac{am}{1 + bm} \text{ yielding}$$

$$R = k \cdot R_{\text{measured}} = \frac{am}{1 + bm}$$

and

$$R_{\text{measured}} = \frac{am}{k(1 + bm)} = \frac{a'm}{1 + bm} .$$

PROCEDURE FOR THE UF_6 AND MODE 1 MEASUREMENTS

Table III lists the parts for the UF_6 and Mode 1 detector configurations. In both configurations, a 1.27-cm-thick aluminum plate is included. This plate must be placed in the bottom of the empty sample cavity before any of the inserts are added, as illustrated in Figs. 2 and 5, so that the samples are positioned properly in the detector.

UF_6 Measurements

For the UF_6 measurements, the graphite insert, cadmium liner, four americium-lithium sources, and four polyethylene source holders are required. Figure 12a shows the positions of each of the sources in the graphite insert along with the orientation of the lids of the sources and the source holders. Note that both sets of lids are pointed down toward the floor. The graphite insert is composed of five sections. We recommend that the bottom three sections be assembled outside the detector when the two lower americium-lithium sources are installed. Place a piece of tape along the outer surface of the three sections to hold them together while the unit is placed in the

TABLE III. Parts List for Each Detector Configuration	
UF ₆	Mode 1
1.27-cm-thick aluminum plate graphite insert (numbered for assembly) cadmium liner (ID 12.7 cm) 4 americium-lithium sources: C171,C174,N009,N010 4 polyethylene source holders	1.27-cm-thick aluminum plate 2 polyethylene, cadmium-lined end plugs cadmium liner (ID 22.5 cm) 2 americium-lithium sources: N009, N010 nickel ring aluminum billet sample holder polyethylene interrogator bottle 3-cm-tall wood blocks (Ext. Mode 1)

Fig. 12. Location and orientation of americium-lithium sources and relevant polyethylene source holders (a only) for both the UF₆ (a) and Mode 1(b) detector configurations (Note: drawing is not to scale).

detector. Next, assemble the remaining two sections with the other two americium-lithium sources in the detector. All measurements are made with the cadmium liner in place.

Once a UF₆ cylinder is placed in the detector for measurement, the cylinder should be measured in three positions, that is, rotate the bottle 120° twice during the course of the measurement for a total of three measurement positions. Figure 13 is a guide for determining the measurement time necessary to achieve a given statistical error in the measurement. A reasonable default value is 9 • 100 s for a total of 900 s. For some cases with low ²³⁵U mass, a longer measurement might be required to meet IAEA error requirements. A multiplicative weighting factor of 1.08 is also included in the error value to account for the difference between the calculated error and the observed error (see Appendix C for further discussion of the weighting factor). This weighting

Fig. 13. Estimate of count time necessary to achieve a certain statistical error in the count rate for UF_6 .

factor may need to be updated in the data collection software from the standard value of 1.20 that is appropriate for passive plutonium counting.

Mode 1 Measurements

The standard end plug detector configuration is used for the non- UF_6 material categories. Two americium-lithium sources, N009 and N010, are used for these measurements. Figure 12b shows the positions of each of the sources in the end plugs along with the orientation of the source lids. Note that the lids point away from the sample cavity. A 1-in. polyethylene disk in the bottom end plug has been removed to achieve the Mode 1 configuration. All of the measurements for this material category will be made in fast mode, so the cadmium liner needs to be installed for all cases using the end plugs.

Figures 14 through 17 are guides for determining the measurement time necessary to achieve a given statistical error for the various materials measured in the Mode 1 configuration. A multiplicative weighting factor of 1.08 is also included in the error value to account for the difference between the calculated error and the observed error (see Appendix C). This weighting factor may need to be updated in the data collection software from the standard high-level neutron coincidence counter (HLNC) value of 1.20.

COMPARISON OF MEASURED AND CALCULATED DETECTOR PARAMETERS

Several uranium oxide samples (see Table IV) were measured in the AWCC with the graphite inserts. Monte Carlo calculations were also performed for the same measurement cases so that the coincidence rates obtained from the two methods could be compared. Table V lists the count rates

(a)

(b)

Fig. 14. Estimate of count time necessary to achieve a certain statistical error in the count rate for cylindrical uranium metal ingots enriched at 46% (a) and 90% (b).

Fig. 15. Estimate of count time necessary to achieve a certain statistical error in the count rate for uranium metal finger ingots.

Fig. 16. Estimate of count time necessary to achieve a certain statistical error in the count rate for uranium billets.

Fig. 17. Estimate of count time necessary to achieve a certain statistical error in the count rate for cylindrical uranium-aluminum alloy scrap.

obtained from measurements and Monte Carlo calculations for the AWCC with the graphite insert. The order in which the sample numbers are listed in the first column of Table V corresponds to the order in which the samples were placed in the detector; the first sample was placed on the bottom and any additional samples were stacked on top of one another in the order indicated. Table VI compares the measured and calculated count rates for the AWCC in the Mode 1 configuration. For both cases, the calculated rates are biased high by about 33%.

TABLE IV. LANL Uranium Oxide Standards⁴

Sample	Oxide Mass (g)	U Mass (g)	²³⁵ U Mass (g)	Enrich. (wt%)	Approx. Oxide Density (g/cm ³)	Approx. Oxide Depth (cm)	Can Height (cm)	Can Diameter (cm)
A1-324-2	1169	987	99.5	10.08	3.1	4.1	12.3	10.8
UIISO 12	1170	990	117.0	11.80	2.4	5.4	12.3	10.8
UIISO 13	1171	991	128.4	12.96	2.4	5.4	12.3	10.8
UIISO 17	1169	989	171.0	17.24	2.6	5.0	12.3	10.8
UIISO 27	1174	991	265.0	26.75	2.1	6.2	12.3	10.8
UIISO 38	1172	991	372.1	37.55	2.1	6.0	12.3	10.8
UIISO 52	1171	989	515.4	52.12	2.2	5.8	12.3	10.8
UIISO 66	1171	990	658.8	66.04	2.3	5.6	12.3	10.8
UIISO 91	1178	990	904.0	91.34	1.7	7.7	12.3	10.8

TABLE V. Comparison of Coincidence Rates Measured in AWCC with the Graphite Insert to those Calculated from Monte Carlo

Sample	^{235}U (g)	T_{meas} (c/s)	R_{meas} (c/s)	R_{calc} (c/s)	$\frac{R_{\text{calc}}}{R_{\text{meas}}}$
UIISO 17	170.6	46,865	83.1	98.8	1.19
UIISO 27	265.1	47,358	119.3	142.6	1.20
UIISO 27,17,13	564.1	47,590	247.4	309.6	1.25
UIISO 17,27,13	564.1	47,568	245.5	313.4	1.28
UIISO 12,52,17,13	931.6	47,347	332.8	463.5	1.39
UIISO 52,12,17,13	931.6	47,407	325.0	461.0	1.42
UIISO 52,27,17	951.3	47,396	340.0	464.4	1.37
UIISO 17,27,52	951.3	47,303	341.0	468.6	1.37
UIISO 52,27,17,13	1079.1	57,284	378.6	526.4	1.39
Average					1.32

TABLE VI. Coincidence Rates Measured in AWCC Using Mode 1 Configuration Compared to those Calculated from Monte Carlo

Sample	^{235}U (g)	R_{meas} (c/s)	R_{calc} (c/s)	$\frac{R_{\text{calc}}}{R_{\text{meas}}}$
UIISO 13	128.4	26.9	33.8	1.25
UIISO 17	171.0	34.7	44.1	1.27
UIISO 27	265.0	46.8	61.9	1.32
UIISO 38	372.1	59.4	81.6	1.38
UIISO 52	515.4	72.4	105.5	1.46
UIISO 66	658.8	87.1	124.6	1.43
UIISO 91	904.0	118.7	162.9	1.37
Average				1.35

Figure 18 compares the efficiency profiles for the AWCC with the graphite inserts obtained from measurements using a californium source (CR-7) and Monte Carlo calculations. The largest deviation between the calculated and measured efficiencies is 5%, which occurs at the center.

SUMMARY

Monte Carlo calculations were used to design a graphite/polyethylene insert for the AWCC that provides a relative uniform interrogation for full or partially loaded UF_6 cylinders. Four americium-lithium sources were required to extend the uniform interrogation over the full sample fill height of ~40 cm. The variation of the AWCC response to non-uniform UF_6 fill geometries (for example, all the UF_6 on one side of the cylinder) was investigated and measurement procedures were developed to give accurate results for all fill geometries.

Calibration curves were generated using the Monte Carlo calculations for UF_6 , HEU aluminum alloy scrap, HEU billets, and HEU ingots. The UF_6 curves correspond to the special graphite insert and all of the other material types correspond to the standard AWCC end plugs in the Mode 1 configuration (see Fig. 5).

The Monte Carlo calculations were used to get the functional relationship between the coincidence response and the mass with subsequent normalization to a benchmark standard of uranium.

Fig. 18. Measured and calculated efficiency profiles for the AWCC with the graphite inserts obtained using a californium source.

For those materials where no benchmark standard was available, we used an intermediate standard that had similar geometry to the actual material. For example, a stack of four cans of UO_2 with enrichments ranging from 12 to 20% was used to simulate a cylinder of UF_6 . The Monte Carlo calculations were performed for both the UO_2 cans and the UF_6 cylinders, and the measured normalization factor for the UO_2 cans was applied to UF_6 cylinders where no benchmark standard was available for the preliminary calibration.

Subsequent application of the calibration technique to actual UF_6 cylinders gave results that agreed with the destructive analysis of the same cylinders within 2 to 3%. The Monte Carlo calculations can be used to greatly reduce the future requirements for physical standards, and the benchmark standards can be of a different material than the target material.

APPENDIX A

Optimization Calculations for Existing End Plugs

The design of the existing end plugs in the AWCC was reviewed to determine whether the existing configuration would also benefit from the addition of graphite around the americium-lithium sources. The goal of these calculations was to determine whether adding graphite to the polyethylene end plug holding the americium-lithium sources would increase the absolute response of the system without significantly increasing the error in the measured reals rate.

Figure A-1 shows how adding graphite in the end plugs affects the count rates. The results in this figure are based on a fixed source strength from two americium-lithium sources. The calculations corresponding to 2.5, 5.1, and 7.6 cm of graphite have the graphite added from the outside going towards the center of the end plug (Disk A in Fig. 5) such that the americium-lithium source was always in a ring of polyethylene. The "C-donut" case corresponds to the polyethylene disks labeled B (see Fig. 5) being replaced with graphite and the disks labeled A being composed of polyethylene. Based on these calculations, the highest response is achieved with 7.6 cm of graphite, which corresponds to a 1.8-cm-thick polyethylene ring around the sources.

The second criterion was to look at the error associated with the reals rates for each of the end plug designs. This data is included in Table A-1 based on a 1000-s measurement. As graphite replaces the polyethylene, the reals-rate error increases. This increase is caused by the increase in the accidentals rate from the reduced shielding of the ^3He tubes from the americium-lithium sources. Because adding graphite to the end plugs did not significantly improve the error, the standard end plug design was not changed for this application. For the active counter designs that have a larger sample-cavity diameter, the combination of polyethylene and graphite in the end plugs could be beneficial.

Fig. A-1. Absolute reals-count rates for various standard end plug designs from Monte Carlo calculations.

TABLE A-1. Calculated Reals-Rate ^a Errors in Various End Plug Designs	
Case Description	Error (% per 1000 s)
all poly	1.2
2.5 cm graphite	1.3
5.0 cm graphite	1.6
7.5 cm graphite	2.0
all graphite	3.6
graphite donut	1.3

^a246 g ²³⁵U sample

APPENDIX B

Self-Interrogation Method

An alternative method for measuring the fissile content of UF_6 is to use a passive self-interrogation technique.⁵ In this case, we take advantage of the neutrons produced from the $(\alpha\text{-n})$ reaction that occurs with fluorine. Alpha particles produced primarily from the decay of ^{234}U interact with the fluorine in the sample to produce an internal neutron interrogation source (see Table B-1) that is better coupled to the assay sample than the external americium-lithium sources. As shown in Fig. B-1, the ratio of the reals to the totals rate is used in generating calibration curves so that the $(\alpha\text{-n})$ source strength dependence is removed from the calibration. The $(\alpha\text{-n})$ source strength is proportional to the amount of ^{234}U in the sample, which is also proportional to the amount of ^{235}U present. Consequently as the ^{235}U mass increases, so does the source strength. By using the ratio of R/T , we can remove the ^{234}U dependence from the calibration because the $(\alpha\text{-n})$ source terms cancel in the R/T ratio.

Two principal factors preclude the use of the self-interrogation method for this measurement situation. The first factor is the slope of the calibration curves in Fig. B-1. As the slope of the curve approaches zero for high masses, particularly for the 30% enrichment case, a small percentage error in R/T translates into a large error in the ^{235}U mass. Table B-2 further illustrates this effect by examining the propagated error in the ^{235}U mass, σ_m , and the effect a 1% change in the R/T ratio has on the mass uncertainty, $R(\pm 1\%) \rightarrow \sigma_m(\%)$, for the 30% enrichment case. This information is presented for both the passive assay and the active assay techniques. For active assay, the calculation is based on R , not R/T , because the ratio of the rates does not result in the source term canceling out. The active data shows a uniform mass uncertainty over the sample mass range; however, for the passive data, the mass uncertainty increases as the sample mass increases.

The second factor is the nonuniform passive axial profile in the AWCC with the graphite insert. Figure B-2 illustrates the count rate profiles using active and passive assay modes for the UF_6 sample used to obtain the data in Fig. 3. The active assay profile is more uniform over the fill height region of the UF_6 cylinder and the efficiency at the bottom of the cylinder drops less significantly, which will improve the measurement error for low-mass, low-fill-height samples.

TABLE B-1. Neutron Yields (n/s•g) from Various Uranium Isotopes in UF_6 (Ref. 6)

Isotope	(α, n)	Spontaneous fission	Total
^{234}U	$5.80 \cdot 10^2$	$5.02 \cdot 10^{-3}$	$5.80 \cdot 10^2$
^{235}U	$8.00 \cdot 10^{-2}$	$2.99 \cdot 10^{-4}$	$8.03 \cdot 10^{-2}$
^{236}U	$2.90 \cdot 10^0$	$5.49 \cdot 10^{-3}$	$2.90 \cdot 10^0$
^{238}U	$2.80 \cdot 10^{-2}$	$1.36 \cdot 10^{-2}$	$4.16 \cdot 10^{-2}$

Fig. B-1. Precalibration curves for UF_6 in the AWCC with the graphite insert using passive assay.

TABLE B-2. Effect of the Calibration Curve for UF_6 with Near Zero Slope on the Calculated Mass Error

^{235}U (g)	Passive Only				Active and Passive			
	R/T	$\sigma_{R/T}$ (%)	σ_m (%)	$R(\pm 1\%) \rightarrow \sigma_m$ (%) ^a	R	σ_R (%)	σ_m (%)	$R(\pm 1\%) \rightarrow \sigma_m$ (%) ^a
362.3	0.0106	3.6	6.0	1.7	235.6	9.9	9.9	1.05
561.8	0.0132	2.8	5.5	2.0	381.1	6.1	6.7	1.07
762.0	0.0153	2.4	5.8	2.4	501.0	4.7	5.1	1.08
962.1	0.0168	2.2	6.1	2.8	611.4	3.9	4.2	1.08
1163.3	0.0185	2.0	7.4	3.5	726.0	3.3	3.5	1.08
1362.5	0.0188	2.0	6.8	3.6	847.6	2.8	3.1	1.08
1571.2	0.0192	1.9	6.6	3.8	968.0	2.5	2.7	1.07

^a $R(\pm 1\%) \rightarrow \sigma_m$ (%) $\equiv$ a 1% change in R results in a σ_m percent change in the mass m .

Fig. B-2. Comparison of response profiles in the AWCC with the graphite insert for active and passive assay of UF_6 .

APPENDIX C

Observed Error vs Calculated Error

The reported error from neutron coincidence collection software has typically contained a 1.20 weighting factor to increase the calculated error using the simple square root of the $(R + A)$ and A counts by 20% so that it corresponds more closely with the observed error.⁷ The 20% factor currently used was determined for a purely passive coincidence counting situation, that is, a system with a small accidentals background. For the case of measuring UF_6 in an AWCC with a graphite insert, a large accidentals background is generated because of (1) the reduced shielding of the 3He tubes by replacing polyethylene with graphite and (2) the addition of two americium-lithium sources, making a total of four sources in the detector. Consequently, a series of measurements was performed with the AWCC in the UF_6 configuration with the americium-lithium sources in place to determine whether this factor needs to be adjusted for the new counting configuration. Table C-1 shows the results from these measurements. Based on the weighting factors for R , which are defined as the ratio of the standard deviation to the calculated error, shown in Table C-1, an average value of 1.08 was chosen for the weighting factor for the real error. The weighting factor, standard deviation, and calculated error are defined as follows:

$$\text{weighting factor} = \frac{\text{standard deviation}}{\text{calculated error}} ;$$

$$\text{standard deviation} = \sqrt{\frac{n \sum x^2 - (\sum x)^2}{n(n-1)}} , \text{ where } x \text{ is either } R \text{ or } T \text{ and } n \text{ is the number of runs} ;$$

$$\text{calculated error in } R = \sqrt{(R + A) + A} ; \text{ and}$$

$$\text{calculated error in } T = \sqrt{T} .$$

TABLE C-1. Measurement Data used to Calculate the Weighting Factor in the Error Calculation

Sample	Count time (s)	Number of runs	Std. dev. for R	Ave. calc. error for R	Weighting factor for R	Std. dev. for T	Ave. calc. error for T	Weighting factor for T
CR-5	100	50	6326	5900	1.072	2700	2283	1.183
A1-324-2	200	991	785	722	1.087	1066	948	1.124

This reduced error-correction factor for the cases with highly uncorrelated counting rates is to be expected and is quantified in Dytlewski's paper.⁷ New versions of the neutron coincidence counting software calculate the correct error and no weighting factor is required.

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