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Neutron Assay System for Confinement Vessel Disposition

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Abstract

Los Alamos National Laboratory has a number of spherical confinement vessels (CVs) remaining from tests involving nuclear materials. These vessels have an inner diameter of 6 feet with 1-inch thick steel walls. The goal of the Confinement Vessel Disposition (CVD) project is to remove debris and reduce contamination inside the CVs. The Confinement Vessel Assay System (CVAS) was developed to measure the amount of special nuclear material (SNM) in CVs before and after cleanout. Prior to cleanout, the system will be used to perform a verification measurement of each vessel. After cleanout, the system will be used to perform safeguards-quality assays of $\leq$100-g ^{239}Pu equivalent in a vessel for safeguards termination. The CVAS has been tested and calibrated in preparation for verification and safeguards measurements.

1 Introduction

The Confinement Vessel Assay System (CVAS) was designed to measure the plutonium content of confinement vessels (CVs) before and after debris removal. The CVs are 6-ft diameter spheres remaining from 1970s-era experiments, and have 1- or 2-in thick steel walls . Figure 1 shows several of these vessels at Los Alamos National Laboratory (LANL). Each CV contains $\sim$500 lb of debris, in the form of actinide metals and oxides, including up to 5 kg ^{239}Pu equivalent, mixed with metal, beryllium, lead, powdered silica, graphite, wires, and hardware. Previous campaigns have successfully removed the debris from a few vessels using hand tools. Now, equipment and procedural improvements, including the use of a robotic arm and glove box that can be attached directly to the large port on the CV, have reduced the risks to the workers performing the cleanout. The goal of the Confinement Vessel Disposition (CVD) project is to remove debris from CVs and dispose of it. The CVAS will be used to assay the ^{239}Pu equivalent present in the vessel before cleanout, as a verification measurement that can be compared with records of the experiment. After cleanout, the system will be used to perform

Figure 1: A number of confinement vessels in their current storage location, awaiting cleanout

safeguards-quality assays of the vessel to determine whether the ^{239}Pu equivalent content has been reduced to $\leq$ 100 g, the level required for safeguards termination.

The CVAS hardware includes a detector and an electronics rack. The detector consists of four banks of 6-ft long ^{3}He tubes embedded in high-density polyethylene moderator. A photograph of the detector system configured around an empty vessel is shown in Figure 2. Two of the detector banks are made up of three separate panels. These banks contain 19 ^{3}He tubes in total, with 11 tubes in the central panel and 4 tubes in each of the outer panels. The two single-panel banks both contain 11 ^{3}He tubes. The design of the CVAS is described in greater detail in reference [1]

2 Calibration of the CVAS System

When the verification measurement is performed prior to cleanout, there may be up to 5 kg of ^{239}Pu equivalent mass in each of the vessels. After debris has been removed from the vessels, the amount of ^{239}Pu equivalent should be less than 100 g, and could be as little as a fraction of a gram. Thus, the CVAS must be capable of performing assays over a large range of Pu masses.

Developing a calibration curve for the system using the doubles rates of standard samples was the most ideal approach. Although the singles rate gives better statistical precision, it is also susceptible to variations in single neutron production through (α,n) interactions with light nuclei. The singles rate is also highly dependent on the composition of the material. The doubles rate, in contrast, is relatively impervious to variations in (α,n) production for material with low neutron multiplication (e.g. relatively low mass and low density), and gives a much more accurate measure of the fissionable mass. Therefore, medium- and large-mass samples were calibrated using the doubles rate. However, for very low Pu mass, the poor counting statistics of the doubles rate necessitated the use of the singles rate instead. Linear calibration curves were used for both low- and medium-mass samples, but since multiplication effects cause the calibration curve to curve upwards for very large masses, the calibration curve for the large-mass samples utilized a quadratic fit. Thus, three separate calibration curves were generated for low, medium, and high mass assays. The sources used for the calibration are discussed in

Figure 2: The CVAS positioned around an empty vessel. The polyethylene moderator has been encased in aluminum to reduce fire loading. The ends of the ^{3}He tubes and their preamplifiers can be seen projecting from the tops of the detector banks.

[2].

Prior to performing calibration measurements, background measurements were performed with the empty vessel in place. The standards were then placed in a Hagen container and suspended in the center of the sphere to provide a yield similar to that of a uniformly distributed source on the inner surface of the vessel. The left side of Figure 3 shows the calibration curve for low-mass samples, taking the singles rate to be a function of $^{240}\mathrm{Pu}_{eff}$ mass in the range from 7 mg to 0.61 g, which corresponds roughly to between 0.1 g and 10 g weapons grade Pu (WGPu, ^{239}Pu $> 93\%$). A linear fit was performed on these data, and the result is also shown in Figure 3 as a straight line. The low mass calibration curve has the functional form: Singles Rate $= 3.536 + 202.666 \times m(^{240}\mathrm{Pu}_{eff})$. Medium-mass calibration measurements were performed in the same manner as the low mass calibration measurements, but because of the improved counting statistics, the doubles rate was used for the calibration. The right side of Figure 3 shows the doubles rate as a function of $^{240}\mathrm{Pu}_{eff}$ mass from 0.34 g to 4.1 g (5 g to 60 g WGPu) along with the linear fit. The corresponding doubles range from 1.2 counts per second (cps) to 8.1 cps above background. The medium mass calibration curve has the functional form: Doubles Rate $= 0.388 + 1.877 \times m(^{240}\mathrm{Pu}_{eff})$.

High-mass calibration measurements were performed without an empty vessel. Standards were placed on a stand in the center of the detector system at a location corresponding to the center of a vessel during assay measurements. In order to adjust for the moderating effect of the 1-in thick steel vessel, a correction was applied to the doubles rate prior to calibration curve fitting. This correction factor was determined from previous measurements of a ^{252}Cf

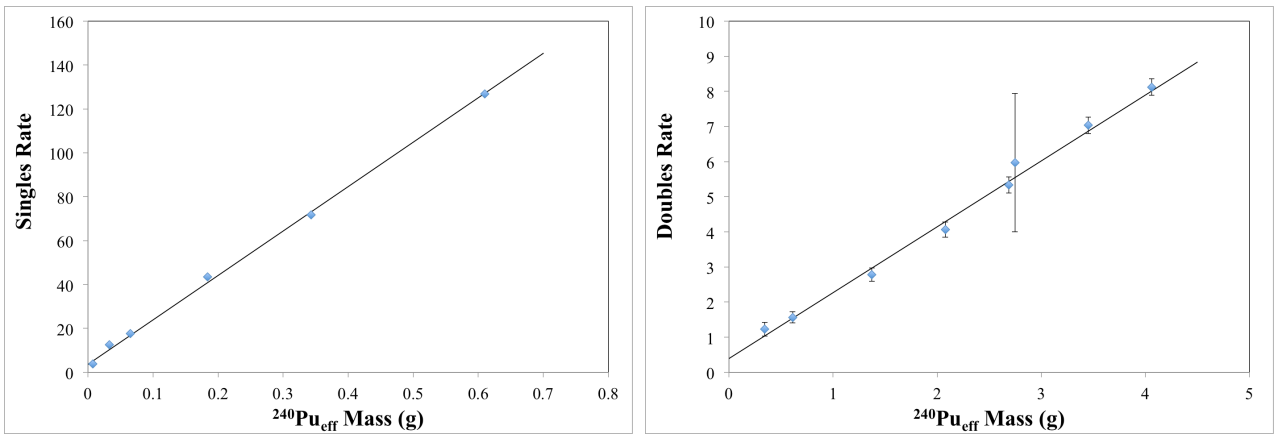

Figure 3: The low- and medium-mass calibration curves both use linear fits to relate rates and $^{240}\mathrm{Pu}_{eff}$ mass. The medium-mass curve uses the doubles rate, which is preferable, while the low-mass curve uses the singles rate, since the counting statistics for the doubles rate in these small samples were poor. For the low-mass curve, error bars are within the polymarkers.

source placed inside a small steel shell of 1-in thickness with an inner diameter of 6 in and an outer diameter of 8 in. The ratio of the doubles rate with the source in the shell to that of a bare source was 1.07 ± 0.02. Figure 4 shows the corrected doubles rate as a function of $^{240}\mathrm{Pu}_{eff}$ mass for masses between 2.7 g and 249.0 g (40 g to 4.151 kg WGPu). Because of sample multiplication in the higher mass standards, the data are fit with a quadratic equation: Doubles Rate = $1.629 \times m(^{240}\mathrm{Pu}_{eff}) + 0.031 \times m(^{240}\mathrm{Pu}_{eff})^2$.

3 Systematic Uncertainty and Backgrounds

The total uncertainty in an assay will depend on the following:

1. The statistical uncertainty associated with the background measurement,
2. The statistical uncertainty associated with the assay measurement,
3. The statistical uncertainty of the isotopic composition. The uncertainty in the isotopics determination is expected to be small ($\sim 6\%$) compared to the other sources of uncertainty,
4. The systematic uncertainty for position bias, and
5. The systematic uncertainty for fluctuations in cosmic ray spallation, unless the background measurement is performed with an empty vessel.

The two sources of systematic error, position bias and cosmic ray spallation, were explored through simulation and experiments in order to quantify the expected uncertainties.

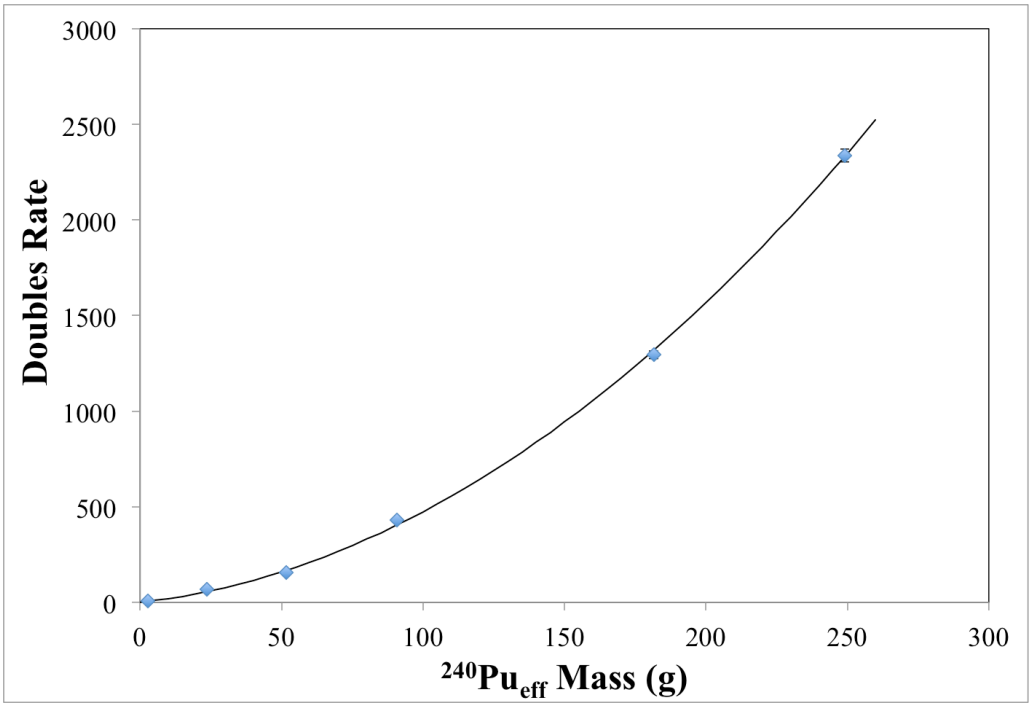

Figure 4: The high-mass calibration curve uses the doubles rate as a function of $^{240}\mathrm{Pu}_{eff}$ mass, and employs a quadratic, rather than a linear, fit to account for multiplication effects

3.1 Position Bias

When a confinement vessel is assayed, the exact distribution of material within the vessel will be unknown. The system has been calibrated assuming a uniform distribution of material. In the worst-case scenario, all of the material may be located at one particular location (e.g. close to a 19-tube assembly), biasing the assay results. To account for such a bias, an MCNPX [3] model was used to estimate the systematic uncertainty due to the variability of the response as a function of source position.

The detector response to a ^{240}Pu source distributed uniformly on the inner surface of the sphere was simulated for a 1-in thick vessel, and then a series of simulated results were generated with a ^{240}Pu point source located at various positions along the inner surface of the sphere. Data were generated for points along the meridian that passes through the point nearest to the 19-tube assembly and the top and bottom of the sphere. The largest deviations occur at the point nearest the 19-tube assembly and at the top of the sphere.

Since the calibration process assumed a uniform distribution of material throughout the vessel, if a vessel were assayed in which all of the material was located at the top of the vessel, the assay would underestimate the amount of material present. The measured singles rate for a source at the top of the vessel is 0.694 times that of a uniformly distributed source. Therefore, the amount of material actually present would be 1/0.694 times, or 44% greater than, the amount assayed. Similarly, if all of the material was located at the point nearest the 19-tube assembly, the measured singles rate would be 1.612 times higher than that of a uniformly distributed source, and the amount of material present would be 38% less than the assay

	Singles	Doubles
Minimum	-26%	-41%
Maximum	+30%	+85%

Table 1: Systematic uncertainty due to position bias

	Singles Rate		Doubles Rate	
	Difference	Error	Difference	Error
Meas. Set 1	24.83	0.53	5.41	0.13
Meas. Set 2	29.71	0.49	5.48	0.14
Average	27.27	5.45*	5.45	1.09*

Table 2: Singles and doubles rates with and without the sphere present. Average errors (starred values) are 20% of the average rates, reflecting fluctuations in the background in addition to variations in cosmic ray spallation.

amount. Given that it is highly improbable that all of the material would be concentrated at the top or against the side of the vessel, a more reasonable estimate of the systematic uncertainty would lie between +44% and -38%. Assuming a uniform distribution of probabilities across the range of relative singles and doubles rates, and that 68% likelihood falls within 68% of the extreme values of +44% and -38% or +30% and -26% respectively. Table 1 shows the asymmetric systematic uncertainties for singles and doubles rates estimated in this manner. After taking the position bias uncertainty into account, the results in Table 1 for material placed at the bottom of the vessel, the assay results agree with the declared mass within 1σ, systematic error.

3.2 Background Subtraction

Cosmic ray spallation occurs when a cosmic ray particle or secondary particle impacts a nucleus, resulting in the expulsion of large numbers of neutrons (as well as protons). The confinement vessel presents a large amount of dense high-Z material, and therefore, cosmic ray spallation makes a sizeable contribution to the background. To estimate the background component due to cosmic ray spallation, we performed measurements of an empty vessel (1-in thick wall) and compared the results to measurements in the same location with no sphere present. Table 2 shows the differences in the singles and doubles rates with and without the sphere present for two sets of measurements. The errors quoted for the average values are 20% of those values, which should accommodate natural fluctuations in the background rate, as well as environmental and seasonal fluctuations in cosmic ray spallation.

Prior to assay of a vessel, a background measurement will be performed in a nearby location. Ideally, this will be performed with the detectors placed around an empty vessel and this background will simply be subtracted from the assay measurement. However, in some situations this may not be possible. In this case, a background will be performed without a vessel present, and the average values in Table 2 will then be added to the no-sphere background rates and used to determine the background error values.

	WGPu	bkg. w/vessel		bkg. w/o vessel	
	Mass(g)	Lower	Upper	Lower	Upper
Singles	0.1	74%	76%	391%	391%
	0.5	30%	34%	86%	87%
	1	37%	31%	49%	52%
	3	26%	30%	30%	33%
	5	26%	30%	27%	31%
Doubles	5	53%	91%	177%	192%
	10	44%	86%	104%	128%
	20	42%	85%	59%	95%
	30	41%	85%	50%	90%
	40	41%	85%	46%	88%
	50	41%	85%	44%	87%
	60	41%	85%	44%	86%

Table 3: Total calibration uncertainty for low- and medium-mass items. The singles-based calibration was used for smaller masses (less than 5 g) and the doubles-based calibration for medium masses (5 g through 60 g).

3.3 Total Uncertainty

As mentioned at the beginning of Section 3, the total uncertainty in an assay depends on two sources of statistical uncertainty – that of the background measurement and that of the assay itself – and on two sources of systematic uncertainty, which have been discussed in subsections 3.1 and 3.2. The relative importance of these sources of uncertainty depends on the mass of Pu measured. For large mass items, the uncertainty is dominated by the position bias, but for very low mass items, the cosmic ray spallation uncertainty can have a large impact. As an illustration, we have calculated the uncertainties for measurements of the low and medium mass WGPu oxide standards used for calibration, taking into account both the systematic errors discussed above and typical values for the statistical errors associated with the assay and its background measurement. Table 3 shows the total uncertainties for Pu masses between 0.1 g and 60 g, using the appropriate calibration for each regime. These calculations assume that the material conforms to the standards used to build the calibration curves. The uncertainties given in Table 3 should be considered a worst-case scenario. As the CVD project proceeds, we expect to reduce the systematic uncertainties. Information about the location of material in a given vessel will allow us to correct for the position bias in the assay, and recent work on background estimation suggests that our estimate of 20% uncertainty is higher than necessary.

4 Conclusion

The CVAS was developed to measure the amount of ^{239}Pu equivalent in spherical confinement vessels before and after cleanout. Prior to cleanout, the system will be used to perform a verification measurement of each vessel. After cleanout, the system will be used to perform safeguards-quality assays of $\leq$ 100-g ^{239}Pu equivalent in a vessel for safeguards termination.

The system was calibrated in three different mass regions in order to cover the entire Pu mass range that will be assayed. The low-mass calibration curve is based on singles rates and covers $^{240}\mathrm{Pu}_{eff}$ masses from 7 mg to 0.61 g (corresponding roughly to between 0.1 g and 10 g WGPu). The medium- and high-mass calibration curves are based on doubles rates; the medium-mass calibration is valid for 0.34 g to 4.1 g $^{240}\mathrm{Pu}_{eff}$ mass (5 g to 60 g WGPu), and the high-mass calibration is valid for 2.7 g to 249 g $^{240}\mathrm{Pu}_{eff}$ mass (40 g to 4.151 kg WGPu). Assayed masses agreed with declared masses within to 3σ, statistical error.

When a low-mass item was assayed with the standards positioned at the bottom of the vessel, the assayed mass was lower than the declared mass by more than three times the statistical error. The systematic uncertainty due to position bias was estimated using an MCNPX model to simulate the response of the system to material localized at various points along the inner surface of the vessel. The systematic uncertainty was taken as 68% of the deviation from the true mass that would be assayed if the material were located at a single point of lowest and highest response. Including the positional bias uncertainty, the assay results for the material placed at the bottom of the vessel agree with the declared mass within 1σ.

Ideally, a background will be performed with the CVAS placed around an empty vessel prior to an assay measurement. If this is not possible, a background may be performed with no vessel in place and a background spallation component added to the measured background prior to assay. The background component due to cosmic ray spallation was determined by performing measurements of an empty vessel and comparing to measurements in the same location with no vessel present. For very low mass items, this will add a sizeable uncertainty.

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