Neptunium Holdup Analysis Using the In Situ Object Counting System Calibration Software and the Line Activity Consistency Evaluator Tool

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ABSTRACT

Measurements performed at the Radiochemical Engineering Development Center (REDC) at Oak Ridge National Laboratory were analyzed to determine the mass of ^{237}Np holdup in equipment in support of the laboratory's nuclear material control and accountability program. The Genie 2000 Gamma Acquisition and Analysis Software and the In Situ Object Counting System (ISOCS) calibration software were used for gamma spectroscopy and analysis of these measurements. The equipment measured at REDC included a rotary kiln, which is used in the process of converting neptunium nitrate solution to neptunium oxide, and an additional set of pipes previously used in the rotary kiln. These two pieces of equipment were measured in several different geometries and modeled using ISOCS. Because of the nature of holdup in the equipment, creating the models involved many unknowns and assumptions. To improve the accuracy of the analysis, the Line Activity Consistency Evaluator (LACE) tool in the Genie 2000 software was used to optimize and validate these unknown physical parameters. The average measured ^{237}Np mass of the rotary kiln, with measurements taken 6 months apart, was found to be 49.45 and 40.53 g. The additional set of pipes had an average measured ^{237}Np mass of 27.08 g. This work demonstrates the application and limitations of ISOCS and the LACE tool for measurement of holdup material.

INTRODUCTION

A consequence of the chemical or physical processing of nuclear material, either in a laboratory or an industrial facility, is the loss of control of a portion of the material within the equipment. Intrinsic defects in the equipment or suboptimal operating conditions, among other causes, will cause a fraction of the material moving through the process to become trapped, or held up, in the equipment. Holdup is an ongoing concern to the facility operator for several reasons. It is a safety concern because holdup deposits can lead to dangerous exposure of personnel to the material during repair or maintenance activities and, in extreme cases involving fissile material, can be a criticality hazard. Holdup also interferes with material accounting. Typically, it is an essential requirement of a facility's nuclear material control and accounting commitments that the location and quantities of all the materials onsite are known. The nature of holdup, *i.e.*, accountable material trapped somewhere in the process, impairs material control and accounting compliance. Nondestructive assay methods have been developed to determine the location and quantity of this holdup *in situ* if it is impractical to recover the material for measurement. Gamma spectroscopy is one of the fundamental hold-up methods and, in this case, was combined

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with Genie 2000 and the In Situ Object Counting System (ISOCS) Calibration Software [1], both developed by Mirion Technologies.

ISOCS represents an improvement over the generalized geometry holdup (GGH) modeling method that is the primary method of performing holdup measurements. Analyzing spectra using the GGH model requires treating the holdup of nuclear material as distributed in one of three simple geometries (i.e., as a point source, a line source, or an area source) and assumes that detector placement is orthogonal to the holdup deposit [2, 3]. These simple approximations can fall short when evaluating holdup in more complicated cases and will thus introduce excessive uncertainty into the results. ISOCS incorporates 21 geometry templates and allows for complex attenuation and flexible detector placement. An example of a simple ISOCS model is shown in Figure 1.

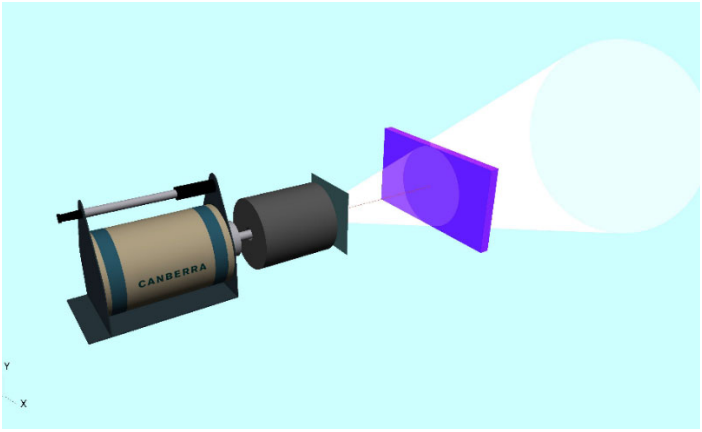

Figure 1. A simple ISOCS geometry model showing the detector, a shield/collimator, and an absorber between the source and the detector, in addition to a cone denoting the field of view through the collimator. In this view, the source is unseen behind the absorber; the software allows the user to rotate the view to any angle as needed.

As part of the ISOCS approach, Mirion Technologies characterizes the energy response of the individual detectors, and this response characterization is used in conjunction with the geometry model to calculate the holdup quantity. The accuracy of the ISOCS model can be evaluated using the Mirion Line Activity Consistency Evaluator (LACE) software tool that is part of the Genie 2000 software package [4]. The LACE tool plots the calculated gamma activities of a nuclide as a function of their energies. Because all gammas of a particular nuclide must necessarily have the same activity, the slope of the plot generated should be zero if all sources of attenuation in the model have been accurately represented. Adjustment of the LACE slope is an iterative process, with the user adjusting the ISOCS model until the optimum (*e.g.*, minimum) LACE slope is achieved. A slope of zero is ideal, suggesting the model provides a realistic representation of the measurement configuration.

IN SITU HOLDUP MEASUREMENT

ISOCS is the tool authorized for the analysis of the holdup measurement of a neptunium solution drying kiln in one of the laboratories in the REDC of the Oak Ridge National Laboratory

(ORNL). An ISOCS characterized Broad Energy Germanium (BEGe) detector was selected for the measurement and installed in a Mirion Technologies ISOCS cart with an InSpector 1000 multichannel analyzer and operated using the Genie 2000 software. The ISOCS shield was fitted with a 30º front plate collimator to narrow the field of view of the detector and the detector was set orthogonal to the kiln. It was also set as close as possible to the glove box to further restrict the field of view to the kiln while minimizing background contributions from other sources in the glovebox.

The kiln is installed in the center of a laboratory glovebox. The glovebox is approximately 122 cm wide and tall. The front window includes four glove ports, approximately 22.5 cm in diameter with approximately 17 cm between each pair (Figure 2). It also includes an extra layer of clear leaded acrylic shielding for personnel safety. This shielding has ports matching the front window and is approximately 111 cm wide by 108 cm tall and 3.5 cm thick.

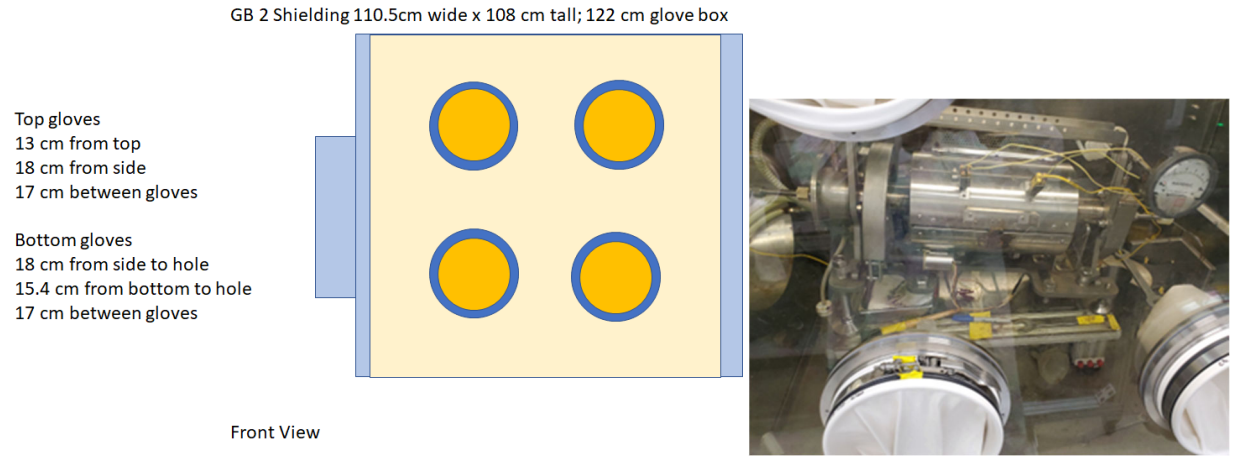

Figure 2. A model of the glovebox (Left) showing the placement and relative dimensions of the glove port openings in the leaded acrylic and a picture of the installed kiln (Right).

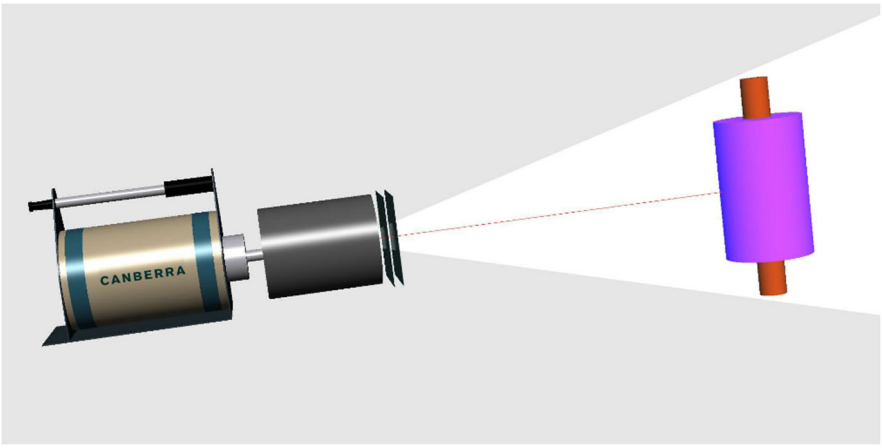

Figure 3. The ISOCS model of the kiln within the glovebox that was used to measure the neptunium holdup.

An ISOCS model of the kiln was developed from the manufacturer's drawings as shown in Figure 3. Absorber layers were placed in the model between the source and the detector, to simulate the glove box window and shielding, and then varied to obtain the optimum LACE slope. The inner pipes of the kiln are removeable for maintenance or replacement, and a previously used set of these pipes is present in another area of the glovebox and was also measured and modeled using ISOCS. Two separate measurements of the kiln were taken 6 months apart and one measurement was taken of the pipes. The quantity of neptunium was reported by the ISOCS calculations as shown in Table 1.

Table 1. Calculated values of neptunium holdup as reported by the ISOCS model. Uncertainties are assigned separately and are not reported here.

ISOCS Reported Values for Neptunium Mass	
First Kiln Measurement	49.45 g
Second Kiln Measurement	40.53 g
Pipes Measurement	27.08 g

The measurement and analysis were expected to be straightforward as the kiln is geometrically simple and, as a commercially available system, its design was well documented. However, the analysis proved more difficult than anticipated. All evaluations using the LACE tool indicated under-attenuation of the characteristic neptunium energies. Further investigation revealed significant portions of the lower two glove ports were within the detector FOV despite efforts to avoid their inclusion. It is certain that these ports, "shielded" by only the operator's rubber gloves, provided a path for unattenuated gammas to reach the detector and make the ISOCS models appear under-attenuated when analyzed with LACE. There was no better mechanical means of positioning the detector available and no better location to set the detector because of space constraints. There is also no way within ISOCS to model an absorber with open holes. To improve the results additional measurements were performed to see if a correction could be developed for the presence of the glove ports.

GLOVEPORTS MEASUREMENTS SETUP

It was not practicable to set up a full duplicate of the glove box and its shield, so a proxy was developed. A square piece of leaded glass was acquired to substitute for the leaded acrylic shield of the REDC glove box. The glass had one large and three small circular holes cut into it with matching plugs of the same material. This larger section could be rotated, permitting the rotation of the smaller holes around the interior of the square (Figure 44). Each smaller hole was assigned a number, 1 - 3. Rotating the larger circular section permitted two foreground measurements through each smaller hole, one on the left side of the bundle and one on the right. Removing the plugs from holes 2 and 3 also permitted a measurement with two holes open.

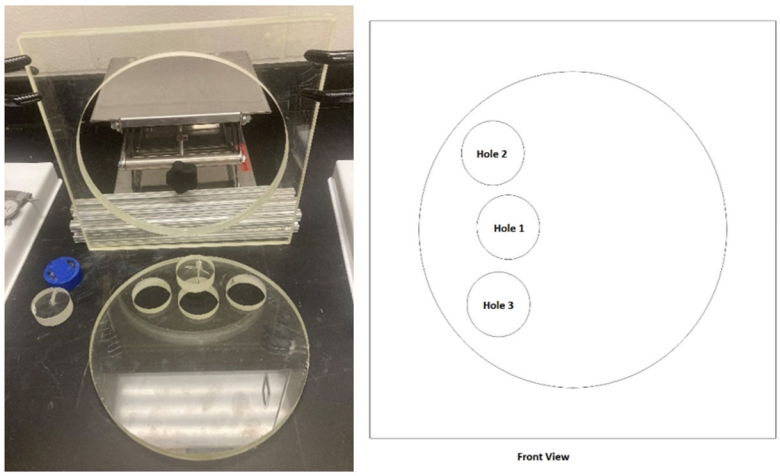

Figure 44. The leaded glass used to measure the effect of opening in the absorber on ISOCS measurements. Left: A picture of the leaded glass square showing the large plug in the foreground and the smaller plugs resting nearby. Right: A drawing with the large plug rotated 90º, showing that the orientation of the smaller openings or plugs can be adjusted.

Radiological safety restrictions did not permit use of the same nuclide as encountered in the glove box, instead ^{235}U was chosen as the surrogate source material. A collection of enriched uranium lab samples was selected. These samples included a traceable sealed source containing highly enriched uranium metal and 10 plastic tubes containing highly enriched uranium oxide (U_3O_8). The thin polyethylene tubes contained a slurry of uranium oxide powder and epoxy. After curing and sealing, each source was approximately 33 cm long. The uranium content of each tube was provided by the manufacturer, but they were not considered traceable standards. Each one also varied somewhat in content, active length, and consistency in the distribution of the material.

The properties of the leaded glass were unknown. Its dimensions and mass were measured to calculate the density. A handheld X-Ray fluorescence device was used to determine the elemental makeup. The X-Ray fluorescence analysis of the leaded glass indicated it had a content of 45.5% lead and 55.5% glass (SiO_2). A long count of the bare ^{235}U metal source and the same source fully shielded by the glass was performed, and the attenuation coefficient calculated from this was compared to the attenuation coefficient for the same mix of materials as calculated by the NIST XCOM Photon Cross Sections Database [5]. These were found to agree satisfactorily, so the X-Ray fluorescence results were used to model the leaded glass as an absorber layer in ISOCS, which also used the XCOM database.

For these test measurements, the same BEGe detector and ISOCS shield used to assay the glove box was used. The system was set up with the sources placed at the same measurement distance from the detector face as for the kiln measurements. The leaded glass was set between the source location and the detector, and entirely within the field of view of the detector. The detector, leaded glass, and sources were then adjusted such that all three were on the same vertical and horizontal centerlines, as shown in Figure 5.

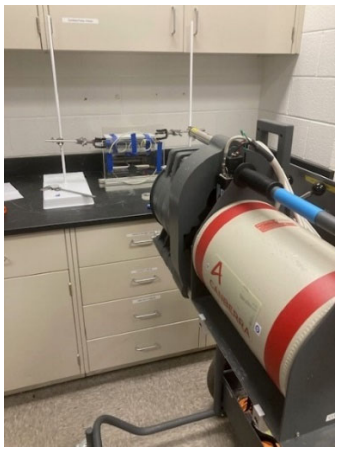

Figure 55. The BEGe detector and the leaded glass were setup to allow measurement of the source tubes through the leaded glass and the various small openings.

Several measurements were taken with a variety of source configurations and attenuation combinations. The goal was to take spectra of the bare sources, then of the sources behind a uniform, solid leaded glass shield. Finally, spectra would be collected of the tube sources behind the leaded glass shield with one or two of the holes uncovered in a variety of locations along the length of the tubes. It was hoped that a single tube could be used to ensure a simple ISOCS model and an analysis with low error and uncertainty. The initial measurement of a fully shielded tube proved futile because the glass so effectively attenuated the 185.7 keV signal that even very long count times could not provide enough counts for ISOCS to calculate a material quantity or even reliably attribute an activity level to the correct nuclide.

All the available tube sources were then combined to increase the signal at the expense of a simpler model. This increased the 185.7 keV signal through the uniform shield and permitted workable count times but still not enough to yield a usable activity level for calculating a material mass. The tube bundle was tightly packed and secured into the smallest cross section possible. This did not align well with either a pipe model or box model in ISOCS but came closest to the box model, so that model was chosen. The ISOCS model required a value for the length of the source and the bundle of 10 sources had to be treated as a single source, so the overall active lengths of the tubes were measured and averaged to yield a source length and combined with the average length and width of the bundle for the model. The density of the material was calculated as though it were uniform and was used as a starting point in the model. The model was then modified to reduce the LACE slope as close as possible to zero. No provision could be made for bubbles and voids except modifications to the material density.

The planned series of measurements with one or two of the glass plugs removed were then completed and the spectra were then analyzed in accordance with the method authorized for the kiln measurements. In every case, the bare source was measured first and evaluated using ISOCS. The same ISOCS model developed for the bare sources was then modified to include an absorber modeled after the leaded glass with no other parameters changed. These shielded measurements were then analyzed using ISOCS and only the density or thickness of the leaded

glass absorber varied independently until the LACE slope of the ^{235}U energies flattened and the calculated mass quantities were recorded.

The uncertainties arising from the physical inconsistencies of the tube sources and the constraints of the model created a positive measurement bias in the result, as shown in Table 2.

Table 2. A comparison of the actual mass of the source (bundled tubes of U_3O_8) to the measured value, as calculated by the ISOCS model.

^{235}U Mass	
Declared	15.155 g
Measured	16.701 g
Error (%)	10.20%

The measured results were retained and used as the accepted results for the subsequent shielded measurements so that the effect of only the absorber adjustments could be evaluated. The shield density and thickness were both varied independently until the LACE slope came as near as possible to zero. Table 3 shows the results for each measurement position when the absorber density was varied, and Table 4 shows the same measurement results when the absorber thickness was varied.

Table 3. Measured ^{235}U mass when the leaded glass absorber density was varied to obtain optimal LACE slope.

Hole(s) Open	**Position**	**Absorber Density (g/cm^3)**	**Lace Slope**	**Measured ^{235}U Mass (g)**	**Error (%)**
3	Left	0.024	-0.001	3.2	- 429
1	Left	0.075	0.063	3.7	- 352
2	Left	0.040	0.012	3.4	- 393
3	Right	0.160	0.006	3.8	- 340
1	Right	0.129	0.002	3.7	- 351
2	Right	0.088	0.000	3.3	- 404
2 + 3	Left and Right	0.760	0.002	6.3	- 163

The calculated masses were much smaller than the accepted value. There was also more variation of the calculated masses than would be expected if the sources were uniform in activity and active length.

The gamma energies penetrating the glass shield were assumed to be negligible, and if the source was uniform throughout its length, then a reasonable holdup value might be

obtained by ratioing the total mass value with the fraction of the source visible through the openings in the leaded glass. The diameter(s) of the open holes was taken to be the amount of the

Table 4. Measured ^{235}U mass when the leaded glass absorber thickness was varied to obtain optimal LACE slope.

Hole(s) Open	Position	Absorber Thickness (cm)	Lace Slope	As found ^{235}U Mass (g)	Error (%)
3	Left	0.008	0.005	3.2	- 430
1	Left	0.025	0.081	3.7	- 355
2	Left	0.012	0.032	3.4	- 397
3	Right	0.054	0.040	3.7	- 346
1	Right	0.046	0.005	3.7	- 351
2	Right	0.031	0.006	3.3	- 405
2 + 3	Left and Right	0.027	0.004	6.3	- 163

Table 5. Average bundle length for the source and the diameters of the three holes in the leaded glass.

Hole Diameter (cm)		Bundle Length (cm)
1	5.124	28.950
2	5.092	
3	5.092	

source that was unattenuated by the leaded glass. These dimensions are shown in Table 5. The total mass was then calculated based on the ratio of the length of the source to the sum of the diameters of the open holes, multiplied by the visible ^{235}U mass as reported by ISOCS. This was then compared to the measured value in Table 2. The results are shown in Table 6 and Table 7.

This assumption yielded better agreement, but still with a positive bias.

RESULTS AND DISCUSSION

The *in situ* measurement of the neptunium holdup was performed using the ISOCS modeling and software calculations.

The measurement results with the open holes in the leaded glass absorber could be adjusted to reasonable values by assuming that the only material contributing to the calculation is that which was visible through the absorber openings, coupled with knowledge of the total source distribution, in this case the dimensions of the tube sources. The results in Table 6 and Table 7

would be an acceptable outcome in routine field NDA measurement. In the field, it is better to overestimate holdup than to underestimate it and acceptable measurement error limits of up to 50% are common.

Table 6. Extrapolated mass calculated by using the ratio of visible length through openings to total source length when the leaded glass density was varied. The difference column is calculated with respect to the accepted value from Table 2.

Hole(s) Open	Position	Hole(s) Diameter (cm)	Diameter to Length Ratio	Measured ^{235}U Mass (g)	Extrapolated ^{235}U Mass (g)	Difference (%)
3	Left	5.092	0.176	3.158	18.0	7
1	Left	5.124	0.177	3.693	20.9	22
2	Left	5.092	0.176	3.388	19.3	14
3	Right	5.092	0.176	3.797	21.6	26
1	Right	5.124	0.177	3.703	20.9	22
2	Right	5.092	0.176	3.314	18.8	12
2, 3	Left and Right	10.184	0.352	6.346	18.0	8

Table 7. Extrapolated mass calculated by using the ratio of visible length through openings to total source length when the leaded glass thickness was varied. The difference column is calculated with respect to the accepted value from Table 2.

Hole(s) Open	Position	Hole(s) Diameter (cm)	Diameter to Length Ratio	Measured ^{235}U Mass (g)	Extrapolated ^{235}U Mass (g)	Difference (%)
3	Left	5.092	0.176	3.152	17.9	7
1	Left	5.124	0.177	3.668	20.7	22
2	Left	5.092	0.176	3.358	19.1	13
3	Right	5.092	0.176	3.746	21.3	24
1	Right	5.124	0.177	3.699	20.9	22
2	Right	5.092	0.176	3.308	18.8	12
2, 3	Left & Right	10.184	0.352	6.340	18.0	8

The measurement error of the bare bundle of tube sources confirmed that the physical properties of the source were not optimally defined in the model. The source documentation indicated that each tube contained a slightly different amount of material, and the material length and distribution (bubbles, voids, *etc.*) varied as well. Finally, although a box-shaped geometry of the bundle was the closest fit within the ISOCS model, it could not perfectly represent the bundle.

These inconsistencies decrease the accuracy and increase the final uncertainty of the calculated quantities.

CONCLUSION

The results confirmed the effectiveness of the leaded glass as an absorber of the ^{235}U energies. Similarly effective shielding on the kiln glovebox, as it is placed there for personnel safety, would be expected. The real-world scenario that prompted this study illustrated the difficulties inherent in these measurements. Despite this, the results indicate that reasonably accurate holdup estimations can be calculated if the ratio of open-to-shielded geometry is known; if it is accepted that the response of the detector to the shielded material was negligible; and if the size and extent of the holdup, as well as the degree of homogeneity, are known. This could be determined through destructive analysis and the operator's knowledge of the process. Alternatively, additional measurement modalities, such as gamma imaging, could be used to understand the source properties to improve the model and calculations.

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