# The Impact of Increased Requirements for Verification Measurements of Category III and Category IV Receipts



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Nuclear Nonproliferation Division

## THE IMPACT OF INCREASED REQUIREMENTS FOR VERIFICATION MEASUREMENTS OF CATEGORY III AND CATEGORY IV RECEIPTS

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October 2022

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#### **ABSTRACT**

As part of the regular nuclear material accounting and control process, sites that possess accountable nuclear material are expected to regularly perform measurements of the material. The expectation for the type of measurements and their frequency is set out in US Department of Energy (DOE) Order 474.2, and further detailed in each site's respective material control and accountability (MC&A) plan and associated controlling documents. The measurements of interest in this report are those performed on external (i.e., between DOE sites) transfer receipts of Category III or Category IV accountable materials. In 2020, a revision of DOE Order 474.2 was proposed that would significantly increase the requirement that measurements be performed upon receipt of material, and that those measurements be quantitative verification (rather than qualitative confirmatory) measurements. A survey was performed of DOE sites to discuss the impact on their MC&A measurements program if more rigorous verification measurements requirements for Category III and IV receipts were implemented. An assessment of nondestructive assay (NDA) measurement instruments to meet those needs was also performed. Sites across the board indicated that personnel and budget would have the greatest effect. Sites qualified as Category I and Category II indicated a reduced impact because these sites already have the measurement capabilities and some personnel but would require additional measurement systems. Category III and Category IV sites indicated a much larger impact because these sites generally have smaller MC&A programs. Some smaller sites stated that a measurement program would need to be created to handle the demand. Calorimetry, quantitative gamma spectroscopy, and neutron coincidence counting systems were identified as common systems that could meet the verification measurement requirements. Additionally, calibration standards for the calorimetry and neutron coincidence counting systems are also needed. A comparative study should be performed for the In-Situ Object Counting System (ISOCS) and ISOTOPICS, along with performing a qualification of a commercial-off-the-shelf (COTS) calorimetry system, and development of an active well coincidence counter (AWCC) system that uses a neutron generator.

#### 1. INTRODUCTION

The possession and use of accountable nuclear materials at DOE sites must be in accordance with the requirements of DOE Order 474.2 Chg. 4 (Pg. Chg.), hereafter referred to as DOE O 474.2 [1]. These requirements define what materials are accountable and what measurements are required as part of transferring these materials internally and externally to another material balance area (MBA) or site. As part of a review and revision process initiated in 2020, changes were proposed to the requirements in DOE O 474.2 that would significantly increase how often a quantitative measurement would be necessary as part of the receipt of Category III and Category IV accountable nuclear materials from a different site. It was understood that this requirement would create an impact at the affected DOE sites; thus, more measurements would need to be performed, yet the appropriate qualified instruments for the required measurements might not be immediately available. Accordingly, a survey was conducted of MC&A personnel at DOE sites to understand what the impact would be of the proposed increase in the required number and rigor of measurements. The instrument needs to perform NDA for verification measurements were assessed.

#### 1.1 NUCLEAR MATERIAL CONTROL AND ACCOUNTING

DOE requires sites that possess or use accountable nuclear materials do so in accordance with appropriate regulations. Accountable nuclear materials are specified in two categories: special nuclear materials (SNM) and other accountable nuclear materials (OANM). The requirements for the accounting of nuclear materials are found in DOE O 474.2. In attachment 2 of the order, Tables A and B define SNM and OANM and are reproduced as Figure 1 and Figure 2.

The reportable quantity is listed for each accountable material or isotope. Under DOE requirements, an item that contains less than half (such that it would round down) of a reportable quantity of a given isotope or material is not accountable. However, if a site has numerous nonreportable items, then the total of the items may sum to a reportable quantity for accounting and control purposes.

Table A. Special Nuclear Materials \*\*

Material Type	Reportable Quantity*	Weight Field Used for Element	Weight Field Used for Isotope	Material Type Code
Enriched Uranium	gram	total U	U-235	20
Uranium-233	gram	total U	U-233	70
Plutonium-242 <sup>1</sup> (Pu)	gram	total Pu	Pu-242	40
Plutonium-239-241	gram	total Pu	Pu-239 + Pu-241	50
Plutonium-238 <sup>2</sup>	tenth of a gram	total Pu	Pu-238	83
Uranium in Cascades	gram	total U	U-235	89

<sup>\*</sup>Reportable quantity is the minimum amount of material subject to the requirements of this Order. Facilities with less than a reportable quantity of a material are exempt from the requirements of the Order for that material. Facilities with a reportable quantity of material that exceed a reporting unit or more of material are to report transactions and inventories to NMMSS. A reporting unit is the mass unit that site/facility accounting systems must use for reporting inventories and transactions.

Figure 1. Table A from DOE O 474.2, Attachment 2, showing the elements and isotopes that are considered SNM.

<sup>&</sup>lt;sup>1</sup>Report as Pu-242 if the contained Pu-242 is 20 percent or greater of total plutonium by weight; otherwise, report as Pu-239-241.

<sup>&</sup>lt;sup>2</sup>Report as Pu-238 if the contained Pu-238 is 10 percent or greater of total plutonium by weight; otherwise, report as Pu-239-241.

<sup>\*\*</sup> Although classified as other accountable nuclear material, separated Am-241, separated Am-243, and separated Np-237 must be controlled and accounted for as SNM

Table B. Other Accountable Nuclear Materials

Material Type	Reportable Quantity*	Weight Field Used for Element	Weight Field Used for Isotope	Material Type Code	
Depleted Uranium (U)	kilogram	total U	U-235	10	
Normal Uranium	kilogram	total U	-	81	
Americium-241 <sup>4</sup> (Am)	gram	total Am	Am-241	44	
Americium-243 <sup>4</sup>	gram	total Am	Am-243	45	
Berkelium <sup>3</sup> (Bk)	microgram	-	Bk-249	47	
Californium-252 (Cf)	microgram	-	Cf-252	48	
Curium (Cm)	gram	total Cm	Cm-246	46	
Deuterium <sup>1</sup> (D)	tenth of a kilogram	D <sub>2</sub> O	$D_2$	86	
Enriched Lithium (Li)	kilogram	total Li	Li-6	60	
Neptunium-237 <sup>4</sup> (Np)	gram	total Np	-	82	
Thorium (Th)	kilogram	total Th	-	88	
Tritium <sup>2</sup> (H-3)	gram	total H-3	-	87	

<sup>\*</sup>Reportable quantity is the minimum amount of material reportable to the Nuclear Materials Management and Safeguard System (NMMSS).

Figure 2. Table B from DOE O 474.2, Attachment 2, listing OANM.

Each site must have a documented MC&A or nuclear material accounting and control (NMAC)<sup>1</sup> plan that specifies what materials that site possesses and which programs and processes will be used to ensure compliance with DOE O 474.2. Under the concept of graded safeguards, wherein more certainty is required, and more rigor is applied to materials of greater *attractiveness level*<sup>2</sup>, SNM and OANM items are assigned an attractiveness level based on relative ease of malicious use and a category based on the quantity of accountable material present in the item. From Attachment 2 of DOE O 474.2, Table C defines the graded safeguards attractiveness and category levels and is reproduced in Figure 3.

Category III and Category IV are defined as the less attractive materials and/or small quantities of more attractive materials, except for assembled weapons and test devices. The majority of OANM is assigned to Category IV.

<sup>&</sup>lt;sup>1</sup>For deuterium in the form of heavy water, both the element and isotope weight fields will be used; otherwise, report isotope weight only.

<sup>&</sup>lt;sup>2</sup>Tritium contained in water (H<sub>2</sub>O or D<sub>2</sub>O) used as a moderator in a nuclear reactor is not an accountable material.

<sup>&</sup>lt;sup>3</sup>Berkelium must be accounted for at the site level. It is not required that it be reported to NMMSS.

<sup>&</sup>lt;sup>4</sup>Americium and Np-237 contained in plutonium as part of the natural in-growth process are not required to be accounted for or reported until separated from the plutonium. If separated, these materials must be controlled and accounted for as SNM.

<sup>&</sup>lt;sup>1</sup> Historically, these programs have been referred to as MC&A programs. Recently, international programs have started using NMAC to refer to the same types of programs in partner countries. For all practical purposes the titles and phrases are identical.

<sup>&</sup>lt;sup>2</sup> Attractiveness level is defined as "a grouping of special nuclear material types and compositions that reflects the relative ease of processing and handling required to convert that material to a nuclear explosive device" (U.S. Department of Energy. "S&S Policy Information Resource: Glossary & Acronyms." DOE O 474.2. Accessed April 1, 2022. Available: https://pir.doe.gov/glossary).

Table	C.	Graded	Safeguards	Table

		Pu/U-233 Category (kg)				Contained U-235/Separated Np-237/Separated Am-241 and Am-243 Category (kg)				All E Materials
	Attractiveness Level	I	п	ш	$IV^1$	I	II	III	$IV^1$	Category IV
WEAPONS Assembled weapons and test devices	A	All	N/A	N/A	N/A	All	N/A	N/A	N/A	N/A
PURE PRODUCTS Pits, major components, button ingots, recastable metal, directly convertible materials	В	≥2	≥0.4<2	≥0.2<0.4	<0.2	≥5	≥1<5	≥0.4<1	<0.4	N/A
HIGH-GRADE MATERIALS Carbides, oxides, nitrates, solutions (≥25g/L) etc.; fuel elements and assemblies; alloys and mixtures; UF <sub>4</sub> or UF <sub>6</sub> (≥50% enriched)	С	≥6	≥2<6	≥0.4<2	<0.4	≥20	≥6<20	≥2<6	<2	N/A
LOW-GRADE MATERIALS Solutions (1 to 25 g/L), process residues requiring extensive reprocessing: Pu-238 (except waste); UF <sub>4</sub> or UF <sub>6</sub> ( $\geq$ 20% < 50% enriched)	D	N/A	≥16	≥3<16	<3	N/A	≥50	≥8<50	<8	N/A
ALL OTHER MATERIALS Highly irradiated <sup>3</sup> forms, solutions (<1g/L), compounds; uranium containing <20% U-235 or <10% U- 233 <sup>2</sup> (any form, any quantity)	Е	N/A	N/A	N/A	Reportable Quantities	N/A	N/A	N/A	Reportable Quantities	Reportable Quantities

The lower limit for Category IV is equal to reportable quantities in this Order

Figure 3. Table C from DOE O 474.2, Attachment 2, showing the matrix used to determine attractiveness level and thus Category for the application of graded safeguards to SNM.

Each site organizes their facilities into one or more MBAs—administrative boundaries typically aligned with room, building, or site boundaries—which define where accountable materials may be stored and used. The site MC&A plan, or associated documentation, will define the MBA boundaries and the procedures needed for accountable material use, inventory, and transfer to other MBAs or sites. All items within a single MBA are summed together to assign a Category to the MBA, using the definitions in Table C of Attachment 2.

## 1.1.1 MC&A Measurements

Measurements are performed on accountable material as part of a site's procedures under its approved MC&A plan. Three types of measurements are defined in Section 6.3.1 of Tech Standard 1194-2019 [2]:

- (1) Accountability measurements shall be used to establish initial values for nuclear materials and to replace existing values with more accurate measured values. These measurements shall be of the highest quality, consistent with the graded safeguards concept for the highest potential material attractiveness level, and the contribution of the uncertainty of the measurement to the overall measurement uncertainty of the inventory or transfer values.
- (2) Verification measurements shall be used to validate the accounting system values when necessary, e.g., at time of physical inventory for non-tamper-indicating items or in response to a security anomaly that could have resulted in a theft or diversion of nuclear material. Verification measurements do not need to use highly accurate or precise methods, however, when these measurements are used to adjust accounting values, they shall have accuracy and precision comparable to, or better than the original measurement method.

<sup>&</sup>lt;sup>2</sup>The total quantity of U-233 = (Contained U-233 - Contained U-235). The category is determined by using the Pu/U-233 side of this table.

<sup>&</sup>lt;sup>3</sup>In this Order "highly irradiated is defined in Attachment 4(Definitions).

(3) Confirmation measurements shall be used to validate the presence of nuclear material for transfers, and to determine nuclear material presence under anomalous conditions. To validate nuclear material presence, a characteristic of the nuclear material, not just the item, shall be measured. However, when an item has been stored in a Category I or II area under TID<sup>3</sup> protection, and an effective material surveillance program for that area, item gross weight may be used for as a confirmation measurement.

Measurements are performed when an accountable item is created, when it is transferred to a new MBA, and as part of periodic inventories. An item may also be measured as part of the response to discovery of an anomaly and as part of the recovery actions. The items that were measured, measurement instruments that were used, and the frequency of required measurements are documented in the site MC&A plan in compliance with DOE O 474.2.

Instruments used to perform measurements must be qualified and approved to be used for MC&A purposes. Although the methodology for measurement method qualification is not specified in DOE O 474.2 or in DOE-STD-1194, best practices for qualifying a method are to document the

- technical basis for selection of the instrument and method,
- materials or items for which the method is applicable,
- performance testing,
- procedures,
- standards,
- uncertainty analysis,
- acceptable level of accuracy and precision,
- measurement control methodology, and
- staff training.

#### 1.1.2 Transfers—External Shipments and Receipts

Transfers are defined as movement of an accountable item across MBA boundaries. They are divided into internal and external.

- Internal transfer—transfer of an accountable item across MBA boundaries within the same site.
- External transfer—transfer of an accountable item across MBA boundaries from two different sites; or rather a transfer across site boundaries.

Internal transfers are handled according to the site's MC&A plan. Requirements for TIDs, measurements, packaging, or other procedures are documented in the site's MC&A plan.

External transfers, also referred to as shipments (i.e., external transfers out of a site) and receipts (i.e., external transfers into a site), are also handled according to the site's MC&A plan. The transfer is documented on Form 741 [3], along with a shipper/receiver agreement (SRA). This process documents the items, their book value on the shipper's accountability system, information about packaging, TID serial numbers, and any other information that needs to be shared between the two different sites for a successful transfer. Which measurements are required by the receiver upon receipt of the accountable items are controlled by the SRA, the site's MC&A plan, and DOE O 474.2. Procedures for the event of discrepancy between the receiver's measured values and the shipper's Form 741 stated values (i.e., a shipper/receiver difference) are also documented in the SRA.

<sup>&</sup>lt;sup>3</sup> TID stands for tamper indicating device, which is a device that "reveals violations of containment integrity."

The proposed revision to DOE O 474.2 in 2020 included language that would have significantly increased how many receipts are required to have a verification measurement performed. The potential impact of this change to the requirements is the focus of this report.

## 1.2 CURRENT MEASUREMENT REQUIREMENTS FOR EXTERNAL RECEIPTS

The current measurement requirements for external transfers from DOE O 474.2 originate from Attachment 3 that supplies metrics with which to assess a site's MC&A plan. Metric 3a. states "the quantity of all nuclear material types present on inventory is determined using identified measurement systems, measurement services, technically justified values or accepted shipper's values when approved by the DOE line management." Each site's MC&A plan includes documentation on which measurements will be performed, which items can be evaluated using technical justification, and which received items can be added to the inventory by accepting the shipper's values.

This has led to a large variation in practices among sites for receiving Category III and Category IV items. In all cases, an appropriate book value is entered onto the inventory record. The variation shows up through what measurement is used to allow the shipper's values to be accepted or the conditions under which a verification measurement is required instead.

## 1.3 PROPOSED CHANGES TO MEASUREMENT REQUIREMENTS

An early proposed version of the revised DOE O 474.2 incorporated language that would be interpreted as requiring verification measurements on more received items than what is now practiced. The proposed language appeared to reduce the likelihood of obtaining DOE line management approval to use technical justifications or acceptance of the shipper's values to generate book values for inventory upon receipt of items. The exact wording is not significant, especially because it has been discussed, reviewed, questioned, and rewritten since the beginning of 2020, and is currently still in draft form. What is significant is that such a change was not simply requiring sites to perform more measurements than before; this proposed change could require sites to perform measurements for which they do not currently have appropriate qualified instruments and trained staff.

Accordingly, this project was tasked with surveying various DOE sites to discuss the impact of a more rigorous interpretation of verification measurement requirements for accountable material receipts and to determine the impact if they had to perform a verification measurement on every Category III or Category IV item received.

#### 2. DOE SITE SURVEY

A survey was conducted with the goal of talking with MC&A personnel, specifically NDA measurements personnel, at DOE sites. The purpose was to discuss the impact that would be caused by a more rigorous verification measurement requirement for receipts of Category III and Category IV items. Another goal of the survey discussions was to identify any needed measurement instrumentation that influences a site's compliance status with such a verification measurements requirement.

#### 2.1 SURVEY METHOD AND MANNER

The survey method was to set up online discussions that allow face-to-face conversation. However, many sites have restrictions on the use of cameras; thus, personnel at those sites either connected without video (which still allowed them to see a shared screen from the discussion facilitator) or connected via phone call. One site used a series of emails to answer questions, and two other sites hosted the discussion entirely over email or phone calls.

Every site was sent a simple agenda to establish the purpose of the conversation and direct the questions to accomplish the survey goals. The agenda was organized as follows:

- Discussion of the Upcoming MC&A Requirement to Perform Category III and Category IV Verification Measurements upon Receipt
- High Level Discussion of Impact (number of Category III and Category IV receipts; manpower impact, etc.)
- Current Measurement Equipment used for Category III and Category IV Verification Measurements
- Measurement Gaps to Meet New Requirement
- Equipment/Procedure Needs to Meet Gaps.

The emphasis was on understanding which Category III or Category IV accountable materials the sites possessed, how often they had an external transfer to their site, and what the impact would be of requiring a verification measurement on each item vs. their current practice.

#### 2.2 LIST OF SITES

The sites operated by DOE that were contacted for this survey are listed below. The discussions and needs assessments have been generalized in this report to the extent possible.

Brookhaven National Laboratory

Fermi National Accelerator Center

Idaho National Laboratory

Lawrence Berkeley National Laboratory

Lawrence Livermore National Laboratory

Nevada National Security Site

Oak Ridge National Laboratory

Pacific Northwest National Laboratory

Princeton Plasma Physics Laboratory

Sandia National Laboratories

Savannah River Site/Savannah River National Laboratory

SLAC National Accelerator Laboratory

Thomas Jefferson National Accelerator Facility

Y-12 National Security Complex

At the time of writing, Ames Laboratory, Argonne National Laboratory, Los Alamos National Laboratory, and none of the Naval Nuclear Laboratories have been reached for discussions.

## 3. RESULTS OF SITE SURVEY

The results of the survey discussions have been organized into general observations, general impact, notable specific impacts, and discussion of the instruments that would be appropriate to meet the identified measurements needs. The results are discussed as broadly and generally as possible; therefore, the overall impact is the point of interest as opposed to specific needs at a particular DOE site.

#### 3.1 GENERAL OBSERVATIONS

Some general observations from the site survey are discussed here.

Wide variation of current practices—the current practices at sites are widely varied. Each site follows the MC&A plan as approved by the appropriate DOE site office. Because of differences in materials and historic processes, the sites have developed different MC&A practices. For example, some current site practices include the following:

- Performing a verification measurement for Category III or Category IV external receipts, with few exceptions.
- Performing two confirmation measurements (e.g., gamma spectroscopy and recording the mass for items with a TID).
- Performing confirmatory gamma spectroscopy measurements and accepting the shipper's book value.
- Performing confirmatory gamma spectroscopy measurements and quantitative measurements after the item is processed.
- Performing no measurements and accepting the shipper's values.

Calorimetry used for verification measurements—multiple sites reported using calorimetry for verification measurements for accountable materials such as <sup>238</sup>Pu, plutonium and tritium. This is notable because properly conducted calorimetry is capable of more accuracy and precision than what is required for verification measurements. Thus, calorimetry is typically associated with accountability measurements. Additionally, calorimetry is a slow measurement, whereby a single data point can take hours or days of instrument time because of the necessity to allow the system to reach thermal equilibrium. Several sites suggested that if they were required to perform more verification measurements than currently practiced, then they would opt for calorimetry for some of their anticipated items.

Shipper's book value accepted pending processing—the sites that perform processing reported that the current practice is to perform confirmatory gamma measurements and then accept the shipper's book values for Category III and Category IV items. Depending on the programmatic need, those items may be stored for years before the processing and measurement occurs. This leads to potential issues for items of questionable pedigree.

Intact fresh fuel is treated as intrinsically tamper indicating—all sites that currently receive fuel elements (i.e., research reactor and commercial) indicated that intact fuel elements are considered intrinsically tamper indicating and therefore are performing confirmatory measurement only.

Quantifying the accountable material in neutron sources—broadly speaking, all sites have isotopic neutron sources, such as americium beryllium (AmBe), plutonium beryllium (PuBe), or americium lithium (AmLi), and can anticipate needing to acquire additional sources in the future. It is difficult to perform a verification measurement of the accountable SNM because of the nature and construction of these sources. Therefore, a more rigorous measurement requirement would be a potential issue.

Category IV is a broad category—The items considered Category IV have a wide variation in material type, physical configuration, and measurability. For high-purity SNM, Category IV items have small masses, but LEU items are defined as Category IV regardless of mass. Items that are difficult to measure, such as irradiated reactor fuel, are also defined as Category IV. A majority of OANM is Category IV, and of this, deuterium (being both widely used and nonradioactive) would present issues with developing a qualified NDA verification method that uniquely verifies deuterium mass. Additionally, the standard NDA method to measure tritium is calorimetry, which includes the challenges mentioned here.

#### 3.2 GENERAL IMPACT AREAS

An increased requirement for performing verification measurements led to broad impact areas.

Need for quantitative gamma spectroscopy measurement methods—almost every site currently uses gamma spectroscopy for confirmatory measurements. Although theoretically any gamma detector can be used to perform quantitative measurements, the necessary corrections and calculations could make it difficult to achieve acceptable results. To obtain accuracy and precision that is sufficient for verification measurements, it is preferred to acquire and qualify a gamma measurement system with a well characterized detector and a supported software package that aids in the calculations and corrections. Many sites proposed that ISOCS, a gamma assay system marketed by Mirion (or the similar system, ISOTOPIC with the ISO-CART, sold by ORTEC), would be appropriate for their gamma measurement needs for additional verification measurements.

Need for uranium measurement methods: neutron coincidence counting—many sites expressed that an AWCC would be appropriate for meeting their measurement needs. Neutrons are typically more penetrative than gammas and are therefore an appropriate measurement tool for quantifying uranium, especially for larger mass or higher density items. Some of the sites already employ AWCCs but would likely need additional instrument acquisitions to meet the increased measurement demands of a more rigorous verification measurement requirement. An AWCC does not provide isotopic knowledge of the item; such knowledge is often obtained by a gamma spectroscopy measurement.

Calorimetry—many sites expressed that they would plan to employ calorimetry if additional verification measurements were required. Calorimetry is best suited to plutonium, especially <sup>238</sup>Pu, and tritium measurements. Some sites already employ calorimeters, but instrumentation capacity would need to be expanded by purchasing new units. Some sites would need to purchase new units to achieve the necessary reliability to keep up with the higher measurement frequency caused by a stronger verification measurement requirement. Some sites do not currently use calorimetry but have proposed it as the measurement technique they would want to employ. Using calorimetry for plutonium requires isotopic information about the item; therefore, this is often obtained by a corresponding high-resolution gamma spectroscopy measurement. Using calorimetry also requires proper matching of the calorimeter heat range, appropriate heat standards, and the heat output of the items to be measured.

Staff impact vs. Instrument Impact—many sites expressed this explicitly, while others revealed their thoughts through discussion. A requirement that increases the number of measurements performed at a site will potentially require more instruments. The capital expense could be substantial but would effectively be a one-time expense. The impact on staff commitment was of greater concern. Over time,

the ongoing expense of maintaining trained personnel to perform and analyze the measurements, as well as the potential programmatic impact of delaying use of materials until the measurements have been completed, will be the more concerning issue. This was especially true of sites using, or who would plan to use, calorimetry for their verification measurements.

Existing Category I and Category II sites have greater current measurement capability—because of the required verification measurements currently in place for Category I and Category II items, sites with existing Category I and Category II MBAs typically have the appropriate instruments and staff to perform verification measurements of Category III and Category IV items. The biggest concern for these sites was the staffing or scheduling impact of performing more measurements at more MBAs. Sites that currently only have Category III and Category IV MBAs would experience a more noticeable impact because such sites are more likely to need new instruments and different or higher levels of staff experience and training augmentation. Some sites would need to dramatically enlarge, or build in, their MC&A measurements program.

#### 3.3 NOTABLE SPECIFIC IMPACT DISCUSSIONS

Two specific challenges identified included the impact of using calorimetry for these verification measurements along with the difficulty of performing qualified measurements on items with challenging isotopic mixtures.

Calorimetry measurements take time—when used for verification or accountability measurements, one of the measurement requirements is to measure an appropriate standard before and after performing the measurement of interest. This ensures that the instrument is working correctly during the measurement and provides data for control charts to track long term statistical trends. Calorimetry measurements require waiting for thermal equilibrium to be achieved. Depending on the isotope and mass of the item, thermal equilibrium can take hours or days until it is reached. Accordingly, a calorimeter system can only perform measurements on a few items per day, at best. The impact of requiring more measurements, when those measurements are to be performed by calorimetry, is that it dramatically increases the number of qualified instruments needed. It can also significantly impact the schedule of the program using the material being measured. Although this can affect any measurement program, current programs with <sup>238</sup>Pu or tritium would likely see the most impact. Programs using Pu that contain smaller amounts of <sup>238</sup>Pu would also see an impact but have other choices available, such as neutron coincidence counting.

Microcalorimetry for items with unusual isotopics—Gamma spectroscopy is often performed using sodium iodide (NaI) or lanthanum bromide (LaBr) detectors. Their energy resolution is sufficient for confirmatory measurements and simple isotopic measurements, such as uranium enrichment. When more detailed isotopic information is needed, typical for verification or accountability measurements, high purity germanium (HPGe) detectors are the standard because of their higher resolution. However, for many unusual isotopic mixes, the HPGe resolution is not sufficient for quantitative measurements to the desired degree of accuracy and precision. Notably, <sup>242</sup>Pu has peaks that interfere with peaks from other Pu isotopes, making it very difficult to perform accurate measurements of the <sup>242</sup>Pu content of an item. It was suggested that the microcalorimetry system developed at Los Alamos National Laboratory—with its energy resolution for gamma measurements that outperforms the HPGe—would be a potential solution to this issue. If verification measurements of <sup>242</sup>Pu items become required with greater rigor, then such a system may become necessary. This is not a COTS product, but it has recently become a product that LANL can provide externally vs. a laboratory-only experimental system.

## 3.4 MEASUREMENT EQUIPMENT NEEDED TO MEET IMPACT

Based on the feedback from the sites surveyed, and the scope of this project, there are a few pieces of measurement equipment that would reduce the impact to site MC&A organizations.

Quantitative gamma spectroscopy measurement methods—as stated in this report, most sites stated that a quantitative gamma spectroscopy method would be needed to fulfill the requirement for verification measurements. In an earlier part of this project, ORNL qualified ISOCS for verification measurements. One site has qualified ISOTOPIC for waste and holdup measurements. There has been minimal work comparing ISOCS with ISOTOPIC other than a paper published by Savannah River National Laboratory [4].

Calorimetry—as stated by several sites, calorimetry would be the primary verification method. However, the calorimeters at those sites are generally decades old. To replace or supplement the existing calorimeters, COTS calorimeters could be purchased from SETSAFE or SETARAM, both brands of KEP Technologies. There are no domestic suppliers of calorimeters for nuclear material measurements. Furthermore, the existing calorimeter software is either DOS-based or requires Windows XP. That is onerous for the sites to maintain and not an ideal long-term solution.

Neutron Coincidence Counting—several sites also mentioned an AWCC would be useful for verification measurements. These systems can be custom built by a few companies and software does exist for their use with modern operating systems. However, the largest barrier to implementing AWCCs is obtaining the AmLi interrogation sources. These AmLi sources are no longer manufactured and existing sources are difficult to find. An experimental study has shown that a deuterium-deuterium neutron generator could be used in the AWCC and would match or exceed the precision of an AWCC using AmLi sources [5]. Other work has been investigated using <sup>252</sup>Cf sources as replacement interrogation sources [6]. Both options have the potential to replace AmLi sources but have not been implemented domestically.

Calibration Standards—with the interest in expanding calorimetry and using the AWCC, a follow-on issue is the availability of calibration standards. Currently, heat standards and AWCC uranium standards are available through the New Brunswick Laboratory Program Office. Calorimeters can be calibrated using certified electrical heat standards, but good practice dictates using at least one nuclear material heat standard for calibration validation. For the AWCC, unless the site already has a set of calibration standards, they would need to be calibrated by simulation, such as by Monte Carlo techniques or by obtaining calibration standards from other sites.

A possible solution to this would be to establish, or revive, the calorimetry exchange (CALEX) program or something similar. This program was administered by the New Brunswick Laboratory and would provide participant sites with calorimetry heat standards. The goal of the program was to identify measurement uncertainties and differences, but the standards would also be used by some participants for calibration of their calorimetry systems. Although transport security would entail cost, this type of program could alleviate the requirement that every site has to have its own full set of calibration standards.

Deuterium—Several sites regularly use and receive accountable quantities of deuterium. There is no known non-destructive method to confirm it is D vs. H. The only method possible is destructive analysis. Correlated measurements are used instead, such as pressure, temperature, and tank volume.

## 4. NEXT STEPS

Based on the information gathered from each of the sites mentioned in this report, the next steps are as follows:

- Acquire software for ISOTOPIC and repeat qualification process to compare performance with ISOCS
- Purchase a COTS calorimeter and work through the qualification process.
- Begin a development project to adapt one of ORNL's AWCCs to use a neutron generator and associated operational software.

These three steps would assist in decision-making across the sites on what are the most cost and time efficient methods to meet verification measurement needs. The work regarding the calorimeter and AWCC would lay the groundwork for other sites to acquire new equipment and qualify a new neutron counting methodology.

#### 5. SUMMARY

In 2020, proposed revisions to DOE O 474.2 pushed for verification measurements of all Category III and Category IV items upon receipt. This project was begun based on that proposed language; however, the proposed requirement has since been softened. Regardless, this work surveyed the DOE complex to understand the impact of verification measurements of all receipts on MC&A programs.

Across the board, sites indicated that personnel and budget would cause the largest impact. Sites which are qualified as Category I and Category II indicated a reduced impact as these sites already have the measurement capabilities and some personnel but would require the acquisition of additional measurement systems. Category III and Category IV sites indicated a much larger impact as these sites generally have smaller MC&A programs. A few smaller sites stated a measurement program would need to be created to handle the demand.

In terms of new equipment, calorimetry, quantitative gamma spectroscopy, and neutron coincidence counting systems were identified as common systems that could meet the verification measurement requirements. Another major issue is the need for calibration standards for the calorimetry and neutron coincidence counting systems.

A comparative study should be performed for ISOCS and ISOTOPICS, qualification of a COTS calorimetry system should be performed, as well as development of an AWCC system that uses a neutron generator.

#### 6. REFERENCES

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