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GUIDE TO NONDESTRUCTIVE ASSAY STANDARDS: PREPARATION CRITERIA, AVAILABILITY, AND PRACTICAL CONSIDERATIONS

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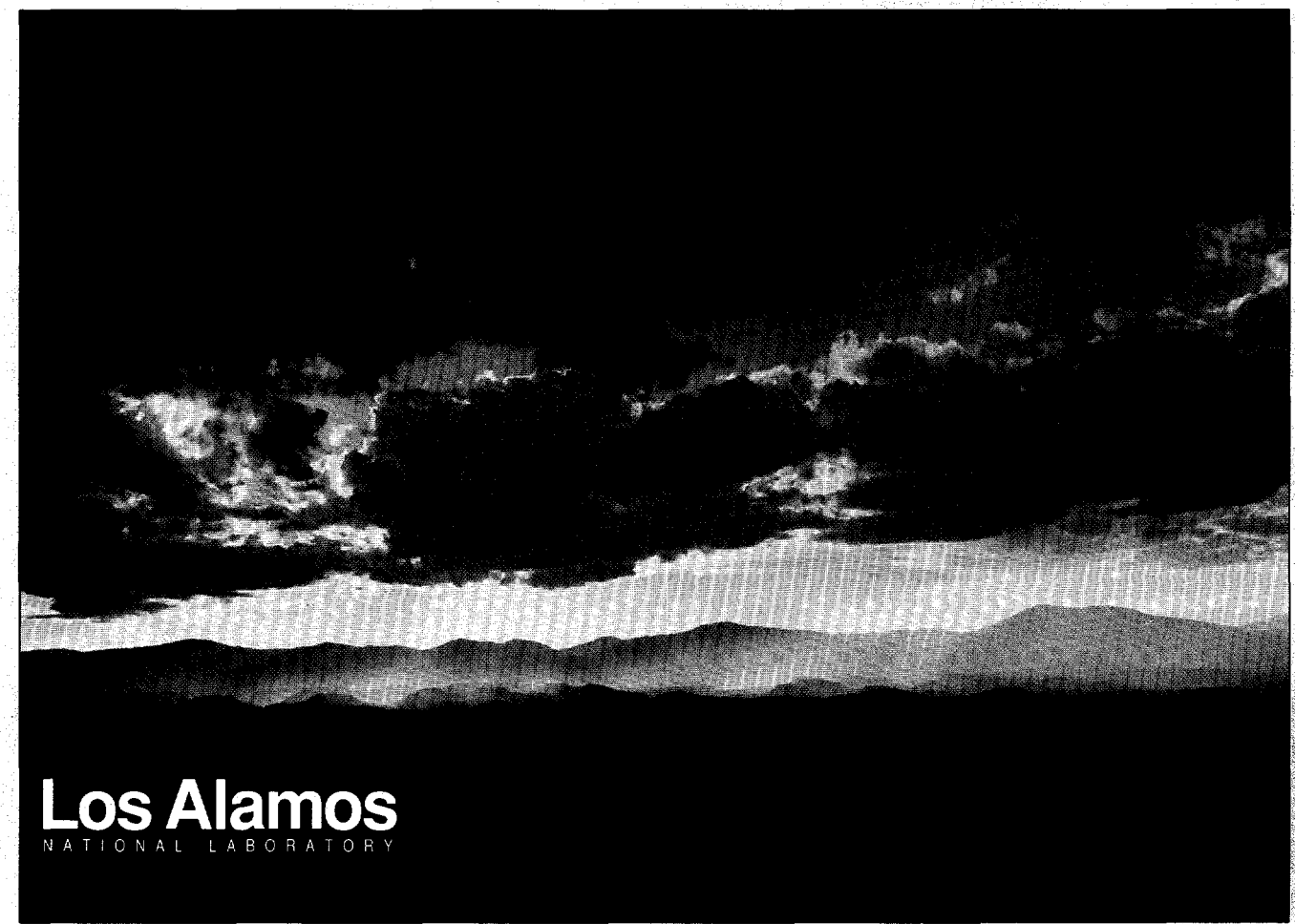

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Guide to Nondestructive Assay Standards: Preparation Criteria, Availability, and Practical Considerations

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Abstract

For certification and measurement control, nondestructive assay (NDA) instruments and methods used for verification measurements of special nuclear materials (SNMs) require calibrations based on certified reference materials (CRMs), or working reference materials (WRMs), traceable to the national system of measurements, and adequately characteristic of the unknowns. The Department of Energy Office of Safeguards and Security is sponsoring production of a comprehensive guide to preparation of NDA standards. The scope of the report includes preparation criteria, current availability of CRMs and WRMs, practical considerations for preparation and characterization, and an extensive bibliography.

In preparing the report, based primarily on experience at Los Alamos, we have found that standards preparation is highly dependent on the particular NDA method being applied. We therefore include sections that contain information specific to commonly used neutron and gamma-ray NDA techniques.

We also present approaches that are alternatives to, or minimize requirements for physical standards. In this section, we describe instrument cross-calibration, where one member of a family of nearly identical detectors is carefully calibrated with WRMs (defining the calibration curve shape), and other detectors are normalized to the reference detector, using a single WRM, or an isotopic source. Another alternative, that is becoming increasingly viable, is the use of Monte Carlo simulations to define shape, and a single WRM, or isotopic source for normalization. Another cost- and effort-saving approach is characterize WRMs with the combination of calorimetry and plutonium isotopics (Cal/Iso). In many practical cases, Cal/Iso values are more accurate than can be obtained with destructive analysis and weighing.

The guide is tailored to a wide audience, US DOE facilities, although considerable interest has been shown from those concerned with materials control and accountability in Russia, Europe, Japan and South America.

I. Introduction

Material control and accountability (MC&A) systems for special nuclear materials (SNMs) are based on material balances for accountable processes. Material balances are based on quantitative measurements. Accordingly, our ability to use material balances to detect significant losses from the process depends greatly on precise and accurate measurements. Nondestructive assay (NDA) techniques have become important tools for quantitative determination of SNM. NDA is often cheaper and more timely than destructive analysis (DA), requires less training of the operator, and does not generate any hazardous or radioactive waste. In some cases, such as scrap and waste from chemical or physical processes involving SNM, NDA may be the only method for determining SNM content with reasonable accuracy. Detailed discussions of the various NDA techniques can be found in references 1 and 2.

NDA of SNM involves the measurement of the radiation emitted by samples of the material to determine the amount of material present, its isotopic composition or both. These measurements must be carefully calibrated or verified in the case of plutonium isotopic or neutron multiplicity measurements. The calibration procedure involves establishing or verifying the response of the measurement instrument to known quantities of nuclear material, by applying the NDA technique in question to thoroughly characterized samples that contain well-known quantities of the nuclear material. The SNM samples used in this way are called working reference materials (WRMs), or certified reference materials (CRMs), if the samples have been characterized according to established national or international procedures and goals.

NDA standards are generally of a physical size, SNM mass, and chemical form that is applicable to the NDA technique being calibrated, but they do not have to exactly match the type of material being measured. There is the mistaken impression that the standards must match the unknowns in all aspects of physical and chemical composition; this is overly restrictive and would require an inordinate number of standards. The knowledge of measurement physics is sufficient that many factors affecting the measurement can be corrected for during the calibration and measurement process, thereby reducing the number and types of standards required. NDA standards should have properties that are representative of the unknowns in all significant characteristics that affect the measurement. For NDA standards, the emphasis should be on calibrating the technique. For example, plutonium oxide standards for neutron coincidence counting should contain a minimum of impurities that contribute to the (α,n) reaction, whereas standards containing substantial amounts of these impurities are perfectly adequate for neutron multiplicity counting. Understanding the physical principles of all the NDA techniques offers the opportunity to minimize the number of sets of standards required. CRMs with the best pedigree (high-quality documentation that provides traceability to the national system of measurements) will not necessarily be suitable for measurement of unknowns different in some characteristic important to the measurement. The report that this paper is drawn from explains the important factors required for each of the NDA techniques discussed.

II. Example of NDA Standards - Uranium Enrichment Measurements

Determination of uranium enrichment is a key measurement in uranium enrichment and fuel fabrication facilities and for international safeguards.

A. Measurement principles

^{235}U decays by alpha emission to excited levels of ^{231}Th, which in turn emit gamma rays of various energies, the most prominent of which has an energy of 185.7 keV. If the uranium sample is large enough, the gamma rays originating from deep within the sample are completely absorbed and do not contribute to the gamma-ray intensity observed at the surface of the sample. Thus, with increasing sample thickness, the 186-keV gamma-ray intensity at the surface reaches an equilibrium value, which is almost independent of the physical form of the sample. For pure uranium compounds this value is proportional to the ^{235}U enrichment of the sample, and in general, only small corrections for chemical composition need be applied. This is the "enrichment meter" principle, and its successful application requires that the sample being assayed is thick enough to be opaque for 186-keV gamma radiation (see Ref. 1, Ch. 7). All measurement parameters that affect the observed counting rate must be carefully controlled and corrected for and are shown schematically in Figure 1.

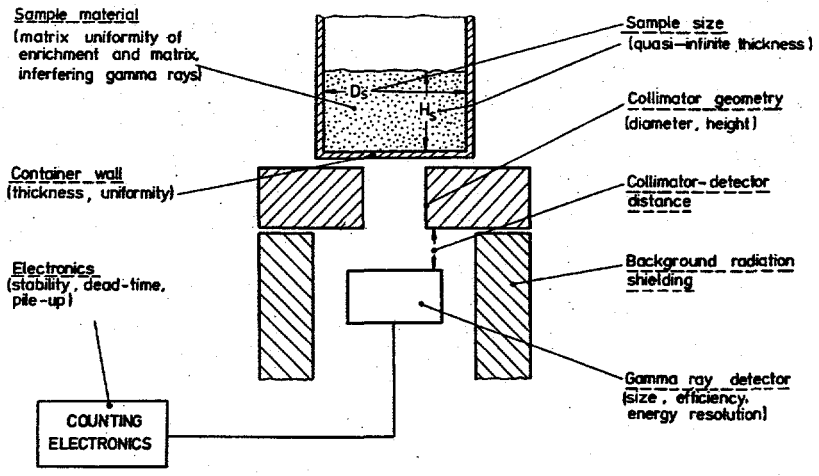

Figure 1. Parameters affecting the accuracy of gamma-ray-spectroscopic ^{235}U enrichment measurements for the "enrichment meter" principle.

The relation between the ^{235}U enrichment E_5 and the net peak counting rate N_{186} of 186-keV photons, observed with a gamma-ray detector is influenced by many factors, as shown in Eq. 1 (E_5 is given in atom %):

$$E_5 = 100 \bullet N_{186} \bullet \frac{4 \bullet \sigma \bullet T_{1/2}}{\ell n\ 2 \bullet P_{186}} \bullet \frac{1}{F \bullet \Omega \bullet \varepsilon} \bullet C_{Ma} \bullet C_{Wa} \bullet C_{El} \bullet C_{Int}, \qquad (1)$$

where

N_{186} = observed net peak counting rate of 186-keV photons
σ = photon attenuation cross section at 186 keV for uranium
$T_{1/2}$ = half-life of ^{235}U
P_{186} = branching ratio of 186-keV photons
F = collimator cross section
Ω = solid angle formed by collimator and detector
ε = intrinsic detector efficiency
C_{Ma} = correction for gamma attenuation in sample matrix materials
C_{Wa} = correction for gamma attenuation in sample container wall
C_{El} = correction for counting rate losses caused by counting electronics
C_{int} = correction for interference due to photons from isotopes other than ^{235}U

A detailed discussion of this equation can be found in reference 3.

B. Physical criteria for enrichment standards

An enrichment standard has the following requirements:

1. The sample must be "quasi-infinitely" thick for 186-keV gamma rays. This condition inherently restricts the method to relatively thick samples. A discussion of this is given in reference 4.

2. The sample must be uniform with respect to ^{235}U enrichment. When mixtures of differently enriched materials are assayed, the sample material must be carefully homogenized before measurement.

3. The container wall thickness must be reasonably thin so that the correction for attenuation in the wall is relatively small. If the container wall is thick, the gamma attenuation must be experimentally determined.

Enrichment measurements using the "enrichment meter" principle will give accurate results only if the sample is sufficiently thick to be opaque for 186-keV gamma rays. For 7-cm diameter samples, the approximate minimum values of the sample masses for UO_2 and U_3O_8 powders are: M = 202.5 g for UO_2 powder and M = 209.7 g for U_3O_8 powder. The CRM EC-NRM-171/NBL-SRM-969 is based on this calculation with 200 g of U_3O_8 in each standard, with a sample height of 2 cm. This is to minimize the amount of SNM required.

C. Preparation and examples of enrichment standards

Two sets of enrichment standards should be mentioned: one is the EC-NRM-171/NBS-SRM-969, which was jointly certified by the CBNM* in Geel, Belgium, and by the US National Bureau of Standards (NBS). This well-documented set has enrichments from 0.3% to 4.5%, which is adequate only for the low-enrichment fuel cycle (Ref. 3). We have been told that IRMM and NBL are preparing additional standards with higher enrichments. Details of this CRM are discussed in Chapter IV of reference 4.

The second set is a suite of WRMs at Los Alamos. This set has nineteen 1-kg cans of uranium oxide with 12 enrichments between normal and 91 at.% that were acquired over a period of many years. They were packaged in various containers; isotopic ratios and uranium fractions were only routinely determined and based on a single sample. In 1985, Los Alamos repackaged and recharacterized these 19 uranium oxide samples (Ref. 5). Of the 19 WRMs, seven were found to be homogeneous and could be declared as primary enrichment standards.

From each of the 19-kg lots of uranium oxide, six 2-g samples were collected from different parts of the oxide. Of the six samples, three were analyzed by Los Alamos, two were submitted to NBL for similar analyses, and one sample was put into archival storage for possible future analyses. The most important analysis is of course the isotopic distribution; the uranium fraction is of secondary importance. The isotopic distributions and other important parameters of the seven primary standards are shown in Table I.

These seven uranium oxide standards span the range from 91% to 10% and complement very well the CRM EC-NRM-171/NBS-SRM-969 set. To perform enrichment measurements over a wide enrichment range, both sets of standards should be used.

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* The Central Bureau of Nuclear Measurements (CBNM) in Geel, Belgium has changed its name to the Institue of Reference Materials and Measurements (IRMM). The US National Bureau of Standards (NBS) has changed its name to the National Institute of Standards and Technology (NIST). The agent that sells the CRM standards for NIST is the New Brunswick Laboratory (NBL) at Argonne, Illinois.

Table I.
Parameters of Primary Enrichment Standards at Los Alamos.

Standard ID	^{235}U	Std. Dev.	U mass	Approx. Oxide	Approx. Oxide
	(at.%)	+/-	(g)	Depth (cm)	Density(g/cm^3)
UISO-91	91.419	0.011	990	7.7	1.7
UISO-66	66.317	0.032	990	5.6	2.3
UISO-52	52.426	0.004	989	5.8	2.2
UISO-38	37.848	0.015	991	6.0	2.1
UISO-13	13.098	0.008	991	5.4	2.4
A1-324-1	10.202	0.003	987	4.6	2.8
A1-324-2	10.200	0.001	987	4.1	3.1

III. Example of NDA Standards - Active Neutron Coincidence Counting

A. Measurement principles

When SNM isotopes fission (either spontaneously or by being induced), the fission process produces prompt neutron radiation with average multiplicities between 2 and 4, depending on the fissioning isotopes. Electronics for neutron assay instrumentation has been developed to recognize multiple (time-correlated) neutron detection events. Thus, the observed neutron coincidence count rate will be related to the amount of fissioning material in the sample. The response of the measurement system is sensitive to several effects in addition to the amount of fissioning material:

- Moderating materials in the sample and in the detector will reduce the average neutron energies and will usually increase the response of the system [examples of moderating materials are plastics (especially CH_2) and moisture (water)].
- Alpha-particle emissions by the SNM isotopes in the sample can interact with low-Z matrix materials and produce neutrons [so-called "(α,n) reactions"].
- Neutrons in the sample can be absorbed, without causing further fissions, and reduce measurement system response [so-called "self-shielding effect"].
- Neutrons in the sample can induce further fissions, adding to the response of the system as the amount of SNM increases [so-called "multiplication effect"].

When all of the above effects are present in neutron assays, the calibration curves are seldom linear and must be defined with several standards.

Neutron coincidence assays of plutonium samples is possible because of the passive signal from spontaneous fissions of the even-mass isotopes. For uranium samples, the passive fission rates are usually insignificant, and so neutron coincidence assays must be performed by inducing fissions in the sample. This is done with random (i.e., not time correlated) neutrons from an external neutron source, usually AmLi. The active neutron coincidence assays therefore measure the sample's fissile (odd-mass) isotopes.

B. Design of standards for the Active Well Coincidence Counter (AWCC)

A set of enriched uranium oxide standards was fabricated in 1991-1992[6] for use with the AWCC. The AWCC has two modes of operation: fast and thermal. In the fast mode, the sample cavity has a cadmium liner that absorbs thermal neutrons; this mode is intended for the assay of large uranium samples. In the thermal mode, the cadmium liner is removed to allow fissions induced by thermal neutrons; this mode is intended for low-mass uranium samples. It was decided that 15 standards could cover the range of sample sizes. These 15 standards can be grouped into low-mass and high-mass sets. The low-mass standards set covers the ^{235}U mass range up to 250 g and the high-mass set covers the range of 250 g up to 4 kg. Samples below 250 g are to be assayed in the thermal mode and those above 250 g are to be assayed in the fast mode.

Approximately 18 kg of U_3O_8 from four batches were blended into four new bottles by taking approximately the same amount from each batch. These four new bottles were mixed in a blender for one hour. Samples were drawn from each bottle for chemical characterization. The characterization included uranium mass fraction, uranium isotopic distribution, weight loss-on-ignition (LOI), and impurity determination.

The standards were packaged into food pack cans; two sizes of containers were used to accommodate all the different masses of U_3O_8. The smaller cans (A) were 12.75 cm in diameter by 10.16 cm tall. The dimensions of the larger cans (B), which used double containment for the higher-mass standards, were 11.43 cm in diameter by 19.05 cm tall for the inner can, and 12.75 cm in diameter by 20.32 cm tall for the outer can. The masses and the container types for this set of WRMs are shown in Table II.

One aspect of this set of AWCC WRMs is less than desirable. For some of the low-mass standards, the fill height is less than 0.5 cm. Because the AWCC is sensitive to the position of the AmLi activation source relative to the sample, any changes in the distribution of the sample in the can could affect the assay result. In retrospect, for the low-mass standards (mass < 50 g), a smaller can with the uranium oxide fixed inside the can would be more desirable.

Table II

Certified ^{235}U mass, Container, and Estimated Fill Height of This Set of AWCC WRMs.

Standard ID	^{235}U (g)	Uncertainty (^{235}Ug)	Container	Est. Fill Height (cm)
STDUO10	9.98	0.02	A	0.1
STDUO25	25.00	0.04	A	0.14
STDUO50	49.95	0.08	A	0.27
STDUO75	75.00	0.12	A	0.79
STDUO100	99.99	0.16	A	1.27
STDUO125	125.00	0.20	A	1.43
STDUO150	149.99	0.23	A	1.69
STDUO250	250.00	0.39	A	2.54
STDUO500	500.00	0.78	A	2.90
STDUO750	750.0	1.2	A	5.08
STDUO1000	1000.0	1.6	A	5.87
STDUO1500	1500.3	2.3	B	7.62
STDUO2000	2000.3	3.1	B	10.2
STDUO2800	2800.2	4.4	B	14.0
STDUO3600	3600.5	5.6	B	18.4

IV. Traceability of Working Reference Materials to National System

We will now present some generalized procedures for the preparation of NDA standards; if the procedures are followed diligently, technically defensible standards that are traceable to the national measurement system will be produced. Most of these procedures follow the recommendations of the American Society for Testing and Materials (ASTM) Standard Guide for Preparation of Working Reference Materials for Use in the Analysis of Nuclear Fuel Cycle Materials,[7] which covers the preparation of both DA and NDA WRM standards. In this paper we will point out the specifics applicable only to the preparation of NDA WRM standards. We would like to emphasize that the basis of any NDA WRM is a high-precision chemical analysis that utilizes appropriate CRMs to validate the analytical procedure. A recommended approach to producing WRM standards is given in Fig. 2. The activities in each of the preparation steps should be well documented, as part of the facility measurement control program.

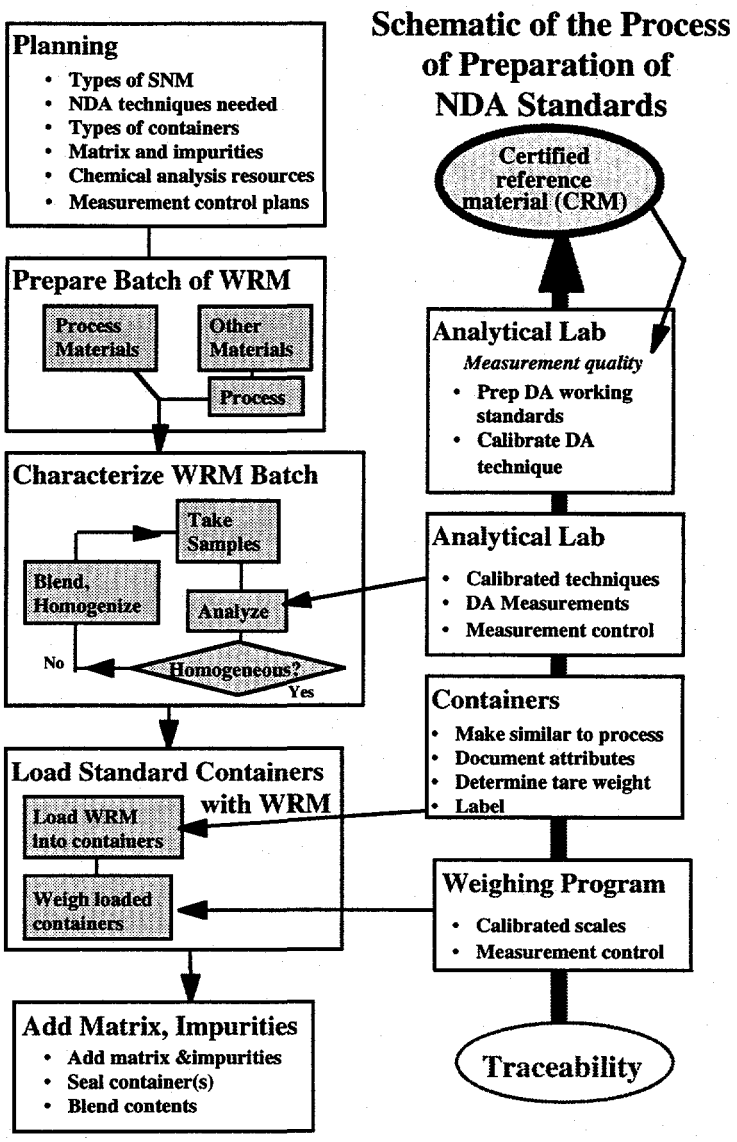

Figure 2. Schematic of the Process of Preparation of NDA WRMs. The standards preparation process is depicted on the left, and the elements of the traceability of the standards to the national measurement system are indicated on the right.

A. Planning the standards fabrication process

The preparation of WRM standards requires careful planning and a written procedure before the work starts to ensure the credibility of the completed WRM. The written procedure should cover the entire project in detail and it should be carefully reviewed to minimize the potential problem areas. It should include at least the following: the intended use of the WRM; the selection and preparation of the SNM; the plans for sampling and sample characterization; containers and packaging for WRM; and the plans for verification measurements and statistical analyses to establish accurately the WRM reference value and its uncertainties.

B. Preparation

The starting materials for the preparation of a WRM standard might be SNM material that is already in the desired WRM form. For example, one could take a batch of uranium dioxide pellets or MOX pellets directly from a process run, appropriately sample and characterize it, and then package the batch as a WRM standard. When the starting SNM is not in the desired WRM form, then a different approach must be used to produce the form desired. For example, one could dissolve high-purity uranium metal in acid, add given amounts of impurities to the solution, and then chemically convert the mixture back into the desired uranium dioxide form (after thorough mixing) to produce a WRM standard with a specific level of impurities.

The homogeneity of SNM in a batch of nuclear material used to prepare a WRM standard is very important for some obvious reasons. First of all, it is important for the whole batch to be homogeneous in SNM so that all samples selected for chemical analysis (DA) truly represent the entire batch. Secondly, it is important because it means that any small portion of the standard can be used for a characterization measurement (for example, an isotopic distribution measurement), and the results will represent the whole standard. Normally, homogeneity can readily be accomplished either by chemical methods, such as dissolving the SNM in acid followed by an appropriate precipitation, or by physical methods, such as blending a batch of oxides for an extended period of time (several hours).

The homogeneity of a batch of SNM can be determined to roughly 1% by the NDA technique of measuring the isotopic distribution of a small portion of the batch with a collimated detector, or by neutron counting of small samples. A more precise test of homogeneity can be performed by DA techniques, or, if the sample is plutonium, by calorimetry. One can use either of these methods to determine the SNM concentration to a few tenths of a percent. Usually the batch can be considered homogeneous if the results from five or six sample analyses agree within statistical limits.

The form of the nuclear material in a WRM standard can be any stable form of the SNM of interest, but special attention must be given to potential causes of instability, such as problems with chemical reactions, problems with changes in stoichiometry, and problems with radiation damage or radioactive decay. The forms most commonly used for nuclear materials have been oxide powder, pellets, or metals. Once the SNM has been characterized and packaged as a WRM standard (usually in the form of oxide powder, pellets, or in metallic form), it is important that its characteristics remain stable for the estimated shelf life of the standard. This is an important concern because it is well known that regular plutonium oxide is deliquescent and readily absorbs moisture. However, if the oxide has been fired to a relatively high temperature (950°C to 1100°C), then the plutonium oxide is relatively stable and will not absorb moisture.[8] The moisture content of a sample can be determined by performing the weight LOI test. The LOI test should be performed on every sample taken to determine the plutonium concentration. For the high-fired plutonium oxide, the LOI weight loss usually amounts to only a few tenths of one percent. Once the sample is prepared and sealed (normally under a dry nitrogen atmosphere), any subsequent weight gain has been found to be negligible. This obviously means that

the standard must be very well sealed to prevent moisture or other impurities from seeping into the container.

C. Weight measurement

The final determination of the amount of SNM in a WRM standard is carried out by weight measurements because they constitute such a high-precision measurement method. Therefore, it is important that the weight be determined accurately and correctly. The scale or balance should be properly chosen to accommodate the range of weights to be determined and it should be at least a factor of ten more accurate than the final requirements. The balances should be verified with National Institute of Science and Technology (NIST)-traceable certified weights both before and after the weighing of the SNM.

An important aspect of weighing in the preparation of WRM standards is to determine the appropriate time in the procedure to make the weight measurements. Let us take the case of preparing a diluted plutonium oxide WRM standard for the NDA technique of segmented gamma scanning. There are two potential methods of determining the weight of SNM. In the first method, the empty inner can is initially weighed, an appropriate quantity of plutonium oxide is transferred to the can, and then the can is weighed again to determine the net weight of plutonium oxide. Then the diluent material (typically graphite or diatomaceous earth) is added to the can, and after the can is properly sealed, the mixture is blended by shaking to create a homogeneous mixture. This method requires that the plutonium concentration of the oxide be determined by DA techniques. In the second method, an already diluted and homogeneous plutonium oxide mixture is transferred into the pre weighed inner can. This method requires that the plutonium concentration of the diluted oxide be determined by DA techniques, which is potentially a much harder problem, especially if the diluent material cannot be readily dissolved. Graphite and diatomaceous earth are commonly used as diluents in the preparation of such standards; both are difficult to dissolve by standard chemical techniques.

D. Container and packaging

It is important that the container material be chemically compatible with the WRM matrix and that the container material not affect signals from the WRM. For example, plastic bottles should not be used for neutron standards because plastics tend to moderate the neutron spectrum and thereby affect neutron measurements. Some plastics also contain fluorine, which will increase the (α,n) signal from the standard.

Usually, aluminum, steel, or stainless steel containers should be used to package the WRM standards. Steel (food) cans that can be sealed by a food packing process are sometimes used, but food pack cans are nominally, relatively thin and tend to deform with usage. Aluminum and stainless steel are better choices for container materials, but both require welding to seal the container, which is cumbersome and costly if it has to be done inside a glove box (for plutonium WRM). Few facilities can weld inside glove boxes. Whenever a stainless steel container is used for a gamma-ray WRM, the attenuation of the gamma rays due to the container should be taken into account in the calibration of the NDA instrument being used.

After the container is packaged, it should also be leak tested to minimize any possibility of radioactive contamination leaking out or moisture and other impurities leaking into the standard.

E. Sampling and characterization

Of all the steps in the preparation of WRM standards, the sampling and subsequent sample characterization is the most important step and it needs to be planned carefully. It is the combination of sampling and characterization that enables one to determine both the uniformity of the batch of

WRM and the amount of nuclear material in each standard. Samples characterized by a trained analyst are traceable to the national system of measurements.

Sampling is relatively simple if the WRM is uranium, plutonium, or mixed oxide. Typically, samples are taken from different parts of a mixed and blended oxide batch to test homogeneity. Sampling can become complicated if the WRM is in the form of fuel pellets. But one can select fuel pellets from the same batch, then assure the uniformity from pin to pin by measuring the U/Pu ratio or the oxygen to heavy metal ratio, and finally select several fuel pins for DA.

Because the available types of CRMs for the calibration of NDA techniques are limited, the traceability of the WRM has to rely mainly on DA techniques. Where possible, DA CRM standards are used to calibrate the methods used to establish the concentration values (reference values) assigned to the WRMs, thus providing the traceability to CRM standards. Uranium- and plutonium-concentration CRM standards are available from NBL as well as CBNM in Europe. Uranium and plutonium isotopic CRM standards are also available from NBL and CBNM.[9] The analytical laboratories should validate their measurement techniques by assaying the CRM standards before or after assaying the unknown nuclear material. The analysis method selected should be a consensus standard method for the sample to be analyzed. For example, mass spectrometry should be used to determine the ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu isotopic distribution, and alpha spectrometry should be used to put Pu in to determine the ^{238}Pu and ^{241}Am isotopic distribution. The uranium concentration of a sample can be determined by means of the Davis-Gray technique. Because of the importance of DA and its traceability to the national measurement system, it is desirable to have two or more independent laboratories performing analyses on identical samples from the same batch of WRM. This is necessary to prevent any potential systematic bias from one laboratory. However, the desire to minimize systematic bias must be balanced with the additional cost and the time delay.

There is also the question of how accurate the DA should be. A general guideline is that the results of the DA should be more precise by a factor of 3 to 5 than and less biased compared to the normal accuracy of the NDA technique with which the WRM will be used. For example, in preparing a neutron standard for an NDA technique where the measurement precision is typically 1%, the DA analysis to quantify the WRM should have a precision of ~0.2% so that the uncertainty contributed by the DA to the overall NDA measurement error is relatively small, 0.02% in this case. In the case of waste WRM standards, the DA can be more precise by a factor of 3. For example, an analytical DA precision of 1% to 2% is acceptable for segmented gamma scanner (SGS) WRM standards because the SGS assay precision and bias are typically around 5%. In light of this, it is quite possible that a combination of calorimetry and isotopic distribution measurements will be sufficient to quantify WRMs intended for NDA measurements on waste nuclear material. The advantage of this approach is that it is relatively inexpensive compared to DA techniques.

At least one of the samples should be analyzed for impurities to limit the influence of the impurities on the certification of the WRM. Typical low-Z impurities that can have a marked effect on neutron NDA are beryllium, boron, fluorine, lithium, sodium, magnesium, aluminum, silicon, chlorine, carbon, and oxygen. Some samples should also be archived to facilitate the resolution of any future questions concerning the standard ("referee" samples).

F. Verification

For a set of WRM standards that covers a range of masses (or concentrations), it is necessary to perform verification measurements to check the internal consistency of the set. This is particularly important for solution WRM standards. Solutions are ideal samples for NDA systems because solutions can achieve an analytical precision and bias of 0.1% to 0.2%. We have found that NDA techniques can determine the gross inconsistencies within a set of solution WRM standards that range from low to high concentrations. The total corrected counts/g of SNM should be internally consistent

for all of the WRM standards in a set of standards; we have found that some samples from a set of concentration WRM standards can deviate from the average value for the amount of SNM by several percent. An example of the verification measurement of a set of SGS can standards is given in[10].

V. ATS

A. Detector family techniques - cross calibration

For both domestic and international inspections of nuclear materials, it is often necessary to employ NDA instruments based on neutron time-correlation counting. For quantitative NDA, the neutron-measuring instruments must be calibrated using physical standards representative of the unknowns. For four reasons, calibration requirements and constraints are significantly different for international-inspector-controlled NDA equipment than for in-plant operator equipment: 1) the physical standards representative of the assay samples are generally not available to the inspector at the plant; 2) the inspector normally covers a much wider range of nuclear materials than does the plant operator because the inspector visits many different facilities; 3) the inspector typically uses a family of similar NDA instruments rather than a single, specialized detector; and 4) the inspector routinely transports equipment between facilities, making it necessary to frequently renormalize prior calibrations.

The traditional calibration approach is to develop physical standards representative of the unknowns and then measure them on the same detector that will be used for the unknowns. However, this approach is frequently impractical for field verification, e.g., because the standards must be sampled and the samples destructively analyzed for certification. Calibration and normalization procedures have been developed for the Uranium Neutron Coincidence Collar (International Atomic Energy Agency (IAEA) designation UNCL) [11] to overcome the above-mentioned problems. These UNCL calibration procedures can be generalized for the other instrument families such as the AWCC. [12] Both the AWCC and the UNCL are active systems and hence they are more complex to implement than the passive systems such as the High-Level Neutron-Coincidence Counter (HLNC-II)[13].

The basic idea of cross-calibration[14] is to carefully calibrate <u>one</u> member of the instrument family for an important category of material [for example, boiling-water-reactor (BWR) fuel assemblies] covering a wide range of mass loadings. The calibration parameters for this <u>reference detector</u> are then <u>fixed</u>, and the responses of other members of the instrument family are normalized to these fixed calibration parameters.

This technique of fixing calibration parameters (curve shape) assumes that the nonlinear shape is primarily a characteristic of the nuclear material items, and any detector-related effects are the same for all members of that instrument family.

The primary benefit of this calibration approach is that it reduces the requirements for physical standards as well as the in-field time required to completely calibrate <u>all</u> members of a detector family. For example, there are presently more than 20 UNCLs, and it is very difficult to find a nuclear facility with a <u>wide enough</u> range of BWR or pressurized-water-reactor (PWR) mass loadings. Also, full use is made of historical calibration data. This tie-in with historical data enhances the quality assurance of measurements. Another more subtle benefit is that detector-to-detector consistency can be determined easily using the cross-reference approach.

The following section gives a specific example and recommended calibration function for the Inventory Sample Counter (INVS)[15]. The case of the INVS counter is considered here, because of its simplicity. There are only two material categories. Multiplication corrections for both are insignificant and therefore unnecessary.

The material categories of interest are

(1) PuO_2 powder, mixed oxide (MOX) powder, and pellets; and
(2) plutonium nitrate and other solutions.

In all cases, the sample plutonium masses should be <20 g for solids and <10 g for liquids. It is possible to lump several material types into one category because the neutron-multiplication differences are negligible.

The preferred calibration function is

$$R = a_1 m + a_2 m^2 \quad ,$$

where R is the real coincidence count rate (reals), $a_1 m >> a_2 m^2$, and m is the effective ^{240}Pu mass.

Note that the term $a_2 m^2$ represents the small multiplication effects.

A set of physical standards covering the full mass range of interest is used to obtain the constants a_1 and a_2 for the reference INVS. The reals rate for a reference ^{252}Cf spontaneous-fission neutron source is measured at the same time as the calibration of the reference INVS. The precision of the ^{252}Cf reals measurement should be 0.5%, or less. The ^{252}Cf data, based on measurements made with the Los Alamos source CR-5, are given in Reference 14 (Table I, p. 4) for seven INVS counters. The calibration parameters a_1 and a_2 are also given for MOX pellets and powders measured in the reference INVS.

For in-field applications of the reference INVS, the measured response R is corrected for possible electronic drifts by remeasuring the reference ^{252}Cf source (or one whose relative neutron intensity is known), making decay corrections, and computing the electronics normalization constant. Table XIX of Reference 14 gives absolute and relative yields for 43 ^{252}Cf sources used routinely by Los Alamos, the IAEA, and EURATOM.

A different INVS can be cross-calibrated by counting the same reference sample (or samples) in both the reference and new detectors at approximately the same time and computing the cross-calibration constant. Occasionally, a container correction is required.

The complete correction factor is

$$k = (\text{electronics}) \bullet (\text{cross reference}) \bullet (\text{container changes}), \text{ or}$$

$$k = [R_0(Cf)/R_{new}(Cf)] \cdot [R_0(\text{INVS-ref})/R_0(\text{INVS-X})] \cdot (\text{container changes}) .$$

The cross-reference term $R_0(\text{INVS-ref})/R_0(\text{INVS-X})$ can be measured using either a plutonium sample or a ^{252}Cf source. The counting precision should be a few tenths of a percent, or less, because any error in this factor will appear as a bias in the assay.

The last term in k, which represents container or matrix changes or both, is needed only if the unknown samples are packaged in a way that alters their INVS response relative to the original standards. Usually, this term is unity.

Given the stability of present-day coincidence electronics and power supplies, the electronics term will be near unity under most conditions.

After k has been determined, the new calibration equation is

$$kR = a_1 m + a_2 m^2 \quad .$$

The significant aspects of using the cross-calibration procedure are to introduce the term R_0(INVS-ref)/R_0(INVS-X) and to fix the calibration constants a_1 and a_2 of all INVS detector heads for a given material category. More examples of implementing the cross-calibration method are given in References 4 and 14.

B. Use of Monte Carlo simulations to minimize the required number of standards

In several important cases, sufficient information is known about the items to be assayed so that accurate simulations of instrument response can be constructed. A good example of these cases is that of unirradiated fast breeder reactor (FBR) fuel elements. One of the primary assay techniques for this important material category is passive neutron coincidence counting (PNCC). Other cases are described in Reference 4.

In Reference 16, a calculational model is applied to the two-parameter (singles and doubles) PNCC assay of fresh, finished FBR subassemblies and the results are compared with calibration measurements. Two assay instruments were considered: the Universal Fast Breeder Reactor Subassembly Counter (UFBC) and the Capsule Counter installed in the Japanese Plutonium Fuel Production Facility. Passive UFBC assays of four Fast Flux Test Facility (FFTF) and six German SNR-300 FBR fuel subassemblies were simulated. Calculated results are shown in Fig. 3, along with measured results for the FFTF assemblies. Passive Capsule Counter assays of five FFTF, three JOYO assemblies, and one MONJU assembly were simulated. Calculated and measured results are shown in Fig. 4.

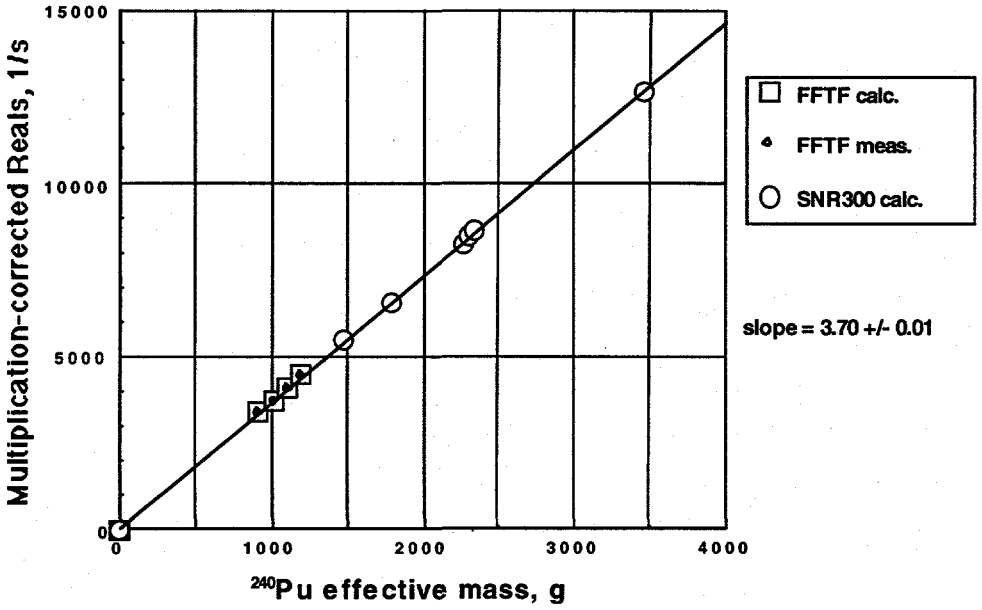

Fig. 3. Plot of calculated and measured multiplication-corrected real-coincidence count rates versus effective ^{240}Pu mass for FFTF and SNR300 FBR fuel subassemblies measured in the UFBC. The linear fit (through the origin) is that for the calculated points and its slope is 3.70 ± 0.01.

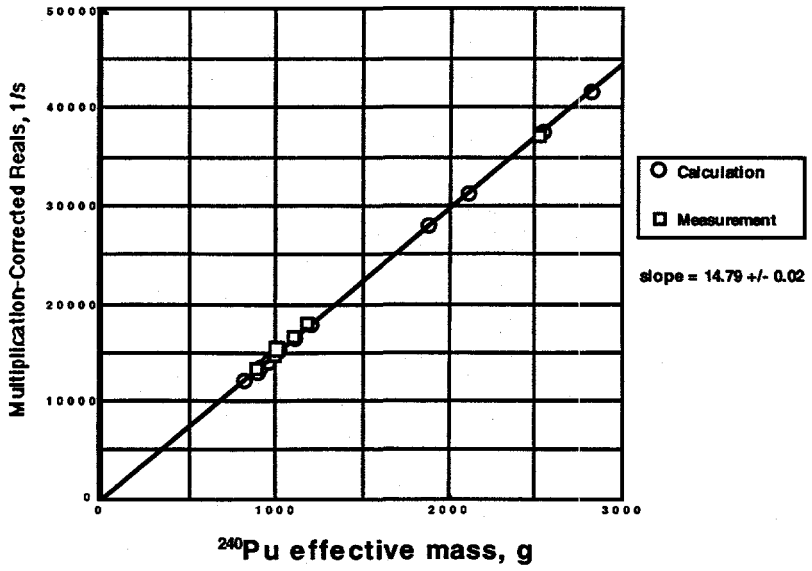

Fig. 4. Plot of calculated and measured multiplication-corrected real-coincidence count rates versus effective ^{240}Pu mass for FFTF, JOYO and MONJU FBR fuel subassemblies measured in the PFPF Capsule Counter. The linear fit (through the origin) is that for the calculated points and its slope is 14.79 ± 0.02.

For the case of the US FFTF fuel, the absolute ratio of calculations to measurements for the multiplication-corrected-reals calibration constant is +1.1 ± 1.0% (average of four subassemblies) for the UFBC and -1.3 ± 0.6% (average of five subassemblies) for the PFPF Capsule Counter. For initial measurements of Japanese fuel in the Capsule Counter, the absolute ratio is -1.0 ± 0.7% for three JOYO assemblies and +0.8 ± 0.7% for the one MONJU assembly. The calculated results are very accurate and precise and offer more effective and less costly inspector verification of FBR fuel elements by reducing reliance on physical standards (as well as costly and time-consuming sampling and DA) to expand the cross-calibration database.

C. Use of calorimetry/gamma-ray spectroscopy to reduce the need for destructive analysis

The combination of calorimetry and plutonium gamma-ray spectrometry is a powerful approach to characterizing plutonium-bearing WRMs. The precision and bias are not as good as chemical analysis, but the cost is considerably less and the results are much more timely. With sufficient counting time, precision and bias of better than 0.3% can be achieved for homogeneous and relatively pure plutonium samples. This is certainly sufficient for waste assay systems where the precision and bias are in the several percent range. The precision and bias are probably adequate for certifying secondary WRMs. In addition, in the US the calorimetry exchange program has been on-going since 1981. The data from this program provide assurance of the accuracy of the approach at several facilities.

The combination of calorimetry and plutonium gamma-ray spectrometry should certainly be used for the reverification of standards on a scheduled basis.

VI. Recommendations

Following are six recommendations based on issues we identified in preparing this Reference 4. We also give the rationale for each recommendation.

1. ***Establish, within the United States, a resource center [such as the Safeguards Performance Laboratory (PERLA) in the European Economic Community (EEC)] housing an extensive collection of well-characterized NDA standards and laboratories in which NDA systems are characterized, calibrated, and used for training.***

The US has no dedicated center where users can bring their NDA instruments for evaluation and calibration. We recommend establishing, in the US, a resource center such as PERLA in the EEC. The purposes of this US center would be as follows:

a.) The user community could validate their NDA instruments and simulation techniques using the wide range of types and masses of standards available at this center. The standards at the center must all be well-characterized.

b.) Fully calibrating the majority of neutron NDA instruments for bulk SNM requires a set of relatively large plutonium- or uranium-bearing standards or both for each material type. Many of the developed neutron NDA instruments have been transferred to the commercial sector and, therefore, some standard instrument models are widely available. Each instrument from a given family of a standard model should have the same calibration curve shape. The "universal" calibration curve of a family of counters could carefully be established for a given material type and reference detector at this center. Also, the user could normalize the response of the individual counter to the reference detector(s) by using the universal curve and one or a few CRMs or WRMs.

c.) The well-characterized standards would further serve the user community in a variety of NDA workshops and training courses.

2. ***Request that NBL/IRMM develop several CRMs.***

Several CRMs needed by the user community should be produced. Because of the difficulty in preparing CRMs, they should be selected carefully in conjunction with the development effort to reduce the number of standards required to calibrate NDA systems. If NBL and the IRMM have difficulty in fabricating some of these CRMs, national laboratories can provide support. The plutonium-bearing CRMs may be issued in limited sets because of the difficulty in shipping. Two needs stand out:

a.) Plutonium metal standards. These will be used to normalize the response of individual counters and can use the universal calibration curve established for these families of instruments. We find that 3-5 thin disks of pure plutonium metal in the mass range 0.5 to 10 g would be very useful.

b.) Low-density standards. These are useful in many ways. They can be used to calibrate SGS systems, which are designed to assay low-density wastes. Because of the relatively low density of these standards, the self-attenuation can be easily corrected and these standards can be used for holdup calibration and testing. One to two standards of plutonium and uranium in the mass range of 10 to 50 g would be adequate. Only a small number of CRMs are required because the SGS calibration is linear as a function of SNM mass.

3. ***Develop bulk-plutonium WRMs.***

Several WRMs needed by the user community at the resource facility should be produced. These items should not be shipped from site to site. Three needs have been identified:

a.) Impure oxide set- Develop a set of impure plutonium oxide standards to be used in studying the effects of impurities on neutron coincidence and multiplicity counting. Six standards will be made; each will have a mass of 1 kg and a constant isotopic composition (~6% ^{240}Pu). The impurities and their (α,n) yields relative to spontaneous fission (the alpha value) will be as follows:

1.) none
2.) silicon with $\alpha = 1$
3.) silicon with $\alpha = 1.5$
4.) fluorine with $\alpha = 2$
5.) boron with $\alpha = 2.5$
6.) magnesium with $\alpha = 3$

b.) Pure plutonium metal set- Develop a set of pure plutonium metal standards to study the effects of neutron multiplication on neutron coincidence and multiplicity counting. Six standards will be made in the form of right circular cylinders and constant isotopic composition (~6% ^{240}Pu). The plutonium masses will be 30, 60, 125, 250, 500, and 1000 g.

c.) MOX and wet plutonium oxide set- Develop a set of three wet oxide standards and a set of three MOX standards to use in studying the effects of moisture and induced fissions in uranium on neutron coincidence and multiplicity counting. The wet oxide standards will contain 1 kg of plutonium with constant isotopic composition (~6% ^{240}Pu). The moisture contents will be 1%, 2%, and 3% by weight. The MOX standards will have the same plutonium isotopic composition, will contain natural uranium, and will all have a combined uranium and plutonium mass of 1 kg. The plutonium masses will be 100, 200, and 300 g.

4. ***Establish the combination of calorimetry and plutonium gamma spectrometry as an acceptable, routine method to certify plutonium-bearing WRMs.***

The combination of calorimetry and plutonium gamma spectrometry is a powerful approach for characterizing plutonium-bearing WRMs. The combination is less precise and has a larger bias than chemical analysis, but the combination is cheaper and more timely. With sufficient counting time, precision and bias can be better than 0.3% for homogeneous and relatively pure plutonium samples. This is certainly sufficient for waste assay systems where the precision and bias are in the several percent range and probably adequate for certifying secondary WRMs. In addition, in the US the calorimetry exchange program has existed since 1981. Data from this program assures accuracy and precision of the approach at several facilities.

The combination of calorimetry and plutonium gamma spectrometry should certainly be used for the scheduled reverification of standards in measurement control procedures.

5. ***Publish a reference manual on the standards (RMs) that cannot and should not be shipped from site to site.***

Some NDA standards, because of physical size, cannot be shipped from site to site. Also, some low-level waste standards that are used to flag disposable waste at the 100-nCi/g level should not be shipped to other sites because of ease of preparation. It will be very useful to publish a practical guide to illustrate how these standards are made.

6. ***Establish the calculational approach as an acceptable, routine method of NDA instrument calibration.***

   Some standards cannot be characterized without being partially or completely destroyed, for example, spent reactor fuel-elements. These standards would also be difficult to store and maintain. For spent-fuel assay systems, a credible calculational method should be developed, which must be acceptable to the regulatory agency.

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