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VERIFICATION OF PRIOR MEASUREMENTS BY NONDESTRUCTIVE ASSAY

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Division of Siting, Health, and Safeguards Standards
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1. INTRODUCTION

Paragraph 70.51(f)(3)(ii) of 10 CFR Part 70, "Special Nuclear Material," requires that NRC licensees subject to 70.51(e) establish procedures for verification by remeasuring previously measured, non-tamper-safed special nuclear material (SNM) in process. The purpose of this report is to delineate nondestructive assay (NDA) techniques that may be used to fulfill the above requirement.

Since NDA methodology and applications of the associated instrumentation are discussed extensively in the literature included in the bibliography, emphasis in this report is placed only on the factors affecting NDA measurement accuracies and the selection of the appropriate NDA techniques for certain measurements based on these factors.

Verification of previously measured materials, for which tamper-safing was not employed, is needed for demonstrating at physical inventory (Ref. 1) that the quantities of SNM previously measured are still present. To ensure that there has been no apparent removal of the SNM since the previous measurement, all containers may have to be first reweighed and then scanned by an appropriate NDA technique.

By reweighing each previously measured non-tamper-safed container, followed by NDA scanning of an attribute characteristic of the contained SNM, any significant removal of such material since the previous measurement can be detected. The attribute is selected for being uniquely characteristic of the contained SNM so that alteration of the material to avoid detection of the SNM removal would be difficult. Such unique attributes include the heat output of a plutonium sample, the induced radiation from a fissile sample undergoing active assay, the isotopic composition of a low-enrichment uranium sample, the gamma spectroscopic analysis of a U-235 sample, or the spontaneous fission neutrons from a Pu-240 sample.

It is hoped that the information presented in this report will be helpful in establishing an acceptable framework for using nondestructive assay for the verification measurement of SNM in non-tamper-safed containers.

2. FACTORS AFFECTING MEASUREMENT ACCURACIES

Certain sample characteristics and measurement practices can affect the response of NDA systems. If these are not controlled or properly accounted for in determining the quantity of SNM from the measured activity, errors will be introduced into the final results. This section identifies important sources of measurement error and recommends possible solutions to reduce or eliminate them.

The factors contributing to the overall uncertainty associated with an NDA measurement are identified in the following four areas: (1) The operational characteristics of the NDA systems, (2) the isotopic composition of the SNM sample, (3) the SNM and matrix materials within a sample, and (4) the composition and dimensions of the container.

2.1 Operational Characteristics

The operational characteristics of the NDA system, together with the ability of the system to resolve the desired response from a composite signal, determine the ultimate usefulness of the system. These operational characteristics include (1) operational stability, (2) geometric effects, (3) stimulating radiation uniformity, and (4) energy of the stimulating radiation.

2.1.1 Operational Stability

The ability of an NDA system to reproduce a given measurement may be sensitive to fluctuations in the operational environment. Temperature, humidity, and line voltage variations affect NDA systems to some extent. These effects may be manifested through the introduction of spurious electronic noise or changes in the high voltage applied to the detectors or amplifiers, thereby changing the detection efficiency.

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The environment must be controlled if such fluctuations result in severe NDA response variations that cannot be eliminated through calibration and operational procedures. The sensitivity to background radiations can be monitored and controlled through proper location of the system and the use of radiation shielding, if required.

2.1.2 Geometric Effects

The concern in the geometry of counting is: What portion of the radiations that are emitted by an SNM sample actually enters the sensitive volume of the detector? This portion ranges from a very small fraction, when the distance from sample to detector is large, to 100% for some 4π-detectors, when the SNM sample being counted is completely inside the detector.

Also, for some detectors such as the well-type gamma and neutron counters, the response of the gross count rate is a function of the position of a point source within the well. This implies that the size and distribution of the sample being counted within the well may have an important bearing on the detector response. To make the necessary error corrections, therefore, it is important to predetermine the geometry effect of a counting system.

2.1.3 Stimulating Radiation Uniformity

It is important that the stimulating radiation field (i.e., interrogating neutron or gamma-ray flux) in active NDA systems be designed to be uniform in intensity and energy spectrum throughout the volume of the irradiation chamber. The residual effect can be measured by using an SNM sample that is small in comparison with the dimensions of the irradiation chamber. The response can then be measured with the SNM sample positioned in different locations within the irradiation chamber.

Various methods, including rotating and scanning the container, scanning the source, distributing sources, and combinations of these methods, have been investigated to reduce the response uncertainty attributable to a nonuniform stimulating radiation field. Scanning a rotating container while the detector and source positions are fixed appears to offer an advantage in response uniformity and is therefore recommended.

2.1.4 Energy of the Stimulating Radiation

If the energy of the stimulating radiation is as high as practicable but below the threshold of any interfering reactions such as the neutron-induced fission in U-238, the penetration of the stimulating radiation will be enhanced throughout the volume of the irradiation chamber. A high-energy source providing neutrons above the energy of the fission threshold for fertile constituents such as U-238 and Th-232 can be employed to assay the fertile content of a container.

The presence of extraneous materials, particularly those of low atomic number, lowers the energy spectrum of the interrogating neutron flux in active neutron NDA systems. Incorporating a thermal neutron detector to monitor this effect and thereby provide a basis for a correction to reduce the response uncertainty caused by this effect is recommended.

Active neutron NDA systems with the capability to moderate the interrogating neutron spectrum can provide increased assay sensitivity for samples containing small amounts of fissile material (<100 grams). To enhance the range of usefulness of the system, it is best that this moderation capability be removable.

2.2 Isotopic Composition of SNM Sample

The sample response observed by an NDA system is usually a composite of contributions from more than a single isotope of uranium or plutonium. These sources of radiation are described below.

2.2.1 Multiple Gamma-Ray Sources

The plutonium isotopes Pu-238 through Pu-241, as well as Am-241 (a daughter product of Pu-241), are spontaneous gamma-ray emitters. The observed spontaneous gamma-ray spectrum from plutonium represents the contribution of gamma rays from these isotopes.

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The uranium isotopes U-233 and U-238 spontaneously emit a gamma-ray spectrum and one gamma ray from U-238 (~766 keV, which is really from the Pa-234 daughter nuclide) is nearly of the same energy as one emitted by Pu-238. Uranium-232, occurring in combination with U-233, has a series of prolific gamma-ray-emitting daughter products, including Th-228.

2.2.2 Multiple Spontaneously Fissioning Isotopes

Plutonium-240 is the principal spontaneously fissioning isotope in the plutonium fuel now used in the nuclear fuel cycle. A few percent of the total fission signal observed is due to Pu-238 and Pu-242.

Uranium has a much lower spontaneous fission rate than plutonium, but in mixed-oxide fuels the spontaneous fission yield from U-238, which is the predominant isotope present, may approach one percent of the Pu-240 yield.

2.2.3 Multiple Thermal Power Source

Each calorimetric measurement response from plutonium is a summation of the spontaneous power emission (in watts per gram) from the isotopes Pu-238, Pu-239, Pu-240, and Am-241. The contribution from each isotope varies as the amount of each isotope varies in the plutonium. Plutonium-238 and Am-241 have much higher specific power (5.6716×10^{-1} and 1.1423×10^{-1} watts/gram) than the other plutonium isotopes.

2.2.4 Multiple Fissile and Fertile Isotopes

In active NDA assay systems, the observed fission response may come from more than one radioisotope. When assaying uranium, if the energy of the interrogating neutrons is above the fission threshold energy for U-238, the response will include a contribution from the fertile U-238 present in the sample, in addition to the response from the fissile U-235, which has a much lower fission threshold.

When a sample of plutonium or uranium mixed-oxide fuel is actively interrogated, the measured response will include contributions from the plutonium and uranium fissile isotopes and, if the energy of the interrogating neutrons is high enough, the response will include contributions from both the fissile and fertile isotopes of plutonium and uranium.

2.3 Response Dependence on the SNM and Matrix Materials Within Sample

If an NDA system is sensitive to the geometry of the detection system and the field of interrogating radiation, variations in the observed sample response may be attributed to the attenuation of emitted gamma rays, neutron multiplication, alpha-neutron reactions, attenuation of the interrogating radiation, and isotopic equilibrium. These factors are discussed below.

2.3.1 Attenuation of Emitted Gamma Rays

The intensity of gamma rays is attenuated as they pass through matter. The magnitude of this attenuation depends on the energy of the gamma rays, as well as the thickness, density, and atomic number of the absorbing material. Low-energy gamma rays are much more severely attenuated than those of high energy. Measuring gamma rays of energies below approximately 80 keV is difficult and usually impractical because of severe attenuation problems.

Uranium and plutonium have high mass absorption coefficients; therefore, self-absorption of gamma rays by these two elements can severely limit the application of passive gamma-ray spectrometry of relatively dense samples.

2.3.2 Neutron Multiplication

Neutrons from spontaneous or induced fission reactions can be absorbed by other nuclei, subsequently causing these nuclei to fission and release additional neutrons. This multiplication results in an increased sample response and therefore should be considered in all active and passive NDA systems.

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2.3.3 Alpha-Neutron (α-n) Reactions

Neutrons are randomly generated through interaction of alpha particles with light elements such as oxygen, fluorine, nitrogen, carbon, and beryllium. Isotopes of uranium and plutonium emit large quantities of alpha particles, which can cause (α-n) reactions and produce neutrons that increase the response of fission measurements.

2.3.4 Sample Self-Shielding

When performing an active assay of dense SNM such as metals or heterogeneous samples containing lumps of SNM, self-shielding of SNM against the interrogating neutrons will occur to varying degrees. Penetrability of the interrogating neutrons into a sample is dependent on the energies of these neutrons, the density of the sample, and the type and amount of matrix material present. Matrix materials containing neutron moderators such as hydrogen will attenuate the interrogating neutrons to lower energy levels and increase the sample self-shielding effect.

2.3.5 Isotopic Equilibrium

Some gamma-ray signatures involve gamma rays emitted from a daughter product of the fissioning isotope of the sample. These signatures can be accurately measured only when the isotopes of the sample and its daughter are in equilibrium. For example, the ingrowth of U-237 in freshly separated plutonium allows one to use the 208-keV gamma ray of U-237 to measure Pu-241 after equilibrium is reached.

2.3.6 Effect of Matrix Materials

Matrix materials, which can be considered as all materials present in the sample other than SNM, can have various effects on the NDA results.

Matrix materials of low atomic number, particularly hydrogenous materials, reduce the energy of neutrons by elastic collision. The lower-energy neutrons are more likely to interact with other nuclei in the sample and thus affect the observed spontaneous fission response.

Matrix materials of high atomic number cause attenuation of gamma rays, particularly gamma rays of low energies. This can cause decreased response during an NDA measurement.

2.4 Composition and Dimension of the Container

Variations in the thickness of the sample container walls and structural composition can significantly affect the accuracy of NDA measurements. Therefore, the same type of container should be used, if possible, for the standard and the unknown samples. Otherwise, a calculated or measured correction must be applied to the measured data to take into account the difference in container wall thickness and structural composition.

3. NDA TECHNIQUE SELECTION

The NDA techniques selected for verification of prior measurements should reflect a consideration of the accuracy requirements for the assay and the type of SNM and containers in the inventory. No single NDA method is capable of universal application, and frequently more than one NDA technique is required to perform the job.

In this section, general applications of SNM measurements by NDA and examples of different techniques are discussed.

3.1 SNM Applications

3.1.1 Plutonium

Calorimetry determinations are the least sensitive to matrix effects but rely on a detailed knowledge of the Am-241 content and the plutonium isotopic composition to transform the measured heat flux to grams of plutonium (Ref. 2).

Gamma-ray spectroscopy systems complement the potential of other assay methods by providing the capability to nondestructively determine, or verify, the Am-241 content and the plutonium isotopic composition (with the exception of Pu-242). High-resolution gamma-ray systems are capable of extracting the maximum amount of information (isotopic composition, isotopic content, presence of extraneous gamma-ray sources) from an assay, but content density severely affects the accuracy of quantitative predictions based on that assay method.*

Passive coincidence detection of the spontaneous fission yield of Pu-bearing systems provides an indication of the combined Pu-238, Pu-240, and Pu-242 sample content. With known isotopic composition, the Pu content can be computed (Ref. 3). Neutron multiplication effects become severe at high Pu sample loadings. Plastic scintillation coincidence detection systems are often designed in conjunction with active neutron interrogation source systems. Operated in passive and active modes, such systems are able to provide an assay of both the spontaneously fissioning content and the fissile content of the sample. The spontaneous background can be subtracted from an active NDA response to provide a yield attributable to the fissile SNM content of the container.

3.1.2 Uranium

Active neutron systems can provide for both high-energy and moderated interrogation spectrum capabilities. Operation with the high-energy neutron source will decrease the density dependence and neutron self-shielding effects and will therefore significantly enhance the uniqueness of the observed response. To extend the applicability of such a system to small fissile loadings, a well-moderated interrogating spectrum can be used to take advantage of the increased U-235 fission probability for neutrons of low energy.

The number and energy of the gamma rays emitted from the uranium isotopes, with the exceptions of the minor isotopes U-232 and U-237, are generally lower than those for plutonium. The 185-keV transition observed in the decay of U-235 is frequently used in uranium applications. However, the penetration of this U-235 primary gamma ray is so poor that the gamma-ray NDA technique is applicable only with low-density, homogeneous samples.**

Calorimetry is not applicable to the assay of uranium enriched in the U-235 isotope because of the low specific power. Also, the intense activity of the daughter products of U-232 imposes a severe complication on the use of calorimetry.

3.2 Examples of SNM Measurements by NDA

The following examples are some typical NDA procedures used frequently in measuring SNM. Table 1 provides a cross-reference of various NDA techniques used with satisfactory results in measuring some common forms of SNM.

3.2.1 Gamma Spectroscopy

1. Thallium-activated sodium iodide detectors are used for measuring uranium-235 enrichment (Ref. 4); the sample is measured through an appropriate collimator and the 185-keV activity is detected with a NaI(Tℓ) crystal to obtain the U-235 enrichment of the sample. Using the collimator and a simple method for proper subtraction of the Compton continuum permits measuring a signal that is truly proportional to the U-235 enrichment. The net counts in the 185-keV peak are proportional to the percent enrichment of the sample.

This technique may even be used to measure samples of uranium-bearing materials that are opaque to the 185-keV gamma rays since the measured 185-keV activity per unit is proportional to the U-235 enrichment. The measurement is nearly independent of matrix material ($Z \leq 30$) and

*Although techniques do exist that can provide quite accurate assay on isotopic ratios that are not sensitive to density and matrix effects, the techniques are still under active development to make them more generally applicable.

**It should be noted that by using the Segmented Gamma Scan (SGS) procedure, homogeneous matrices are no longer a limiting factor for the measurement of U-235 material.

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Table 1

NDA TECHNIQUE SELECTION
FOR SOME COMMON SNM SAMPLES

SNM	Technique: Calorimetry	Gamma Spectroscopy: NaI(Tℓ)	Gamma Spectroscopy: Ge(Li)	Neutron Coincidence Counting	Active Assay
Low-enrichment U-235 Fuel Pellet		(Ref. 4)[a]	X[b]		(Ref. 5)
UF_6 Product (enriched)		(Ref. 6)	X		
UO_2 (>80%)		(Refs. 7, 8)	(Refs. 7, 8)		(Ref. 9)
U-235 Ash			(Ref. 10)		
U-238 Incinerator Ash			X	(Ref. 11)	
UO_2 Power			X		(Refs. 5, 12)
U/Pu Mixed Oxide	(Ref. 13)		(Refs. 14, 15)	(Ref. 16)	
Pu Metal Alloy	X		(Refs. 15, 17, 18)	X	
PuO_2 Feed Material	(Ref. 19)		X	(Refs. 20, 21)	
Pu-239 Mixed Oxide	X		X		(Refs. 5, 22)
Pu-238 Metal and Metal Oxides	X		X	(Ref. 23)	
Pu Mixed Oxide Scrap	X		X		(Ref. 15)
Pu Nitrate			(Refs. 16, 24, 25)	(Ref. 26)	
Pu-239 (Homogenously distributed in ash or graphite matrix	X	(Ref. 27)	X	(Refs. 23, 26)	
Pu-238 and Pu-239 Scrap and Waste Cans	X	X	X	X	(Ref. 28)
Pu High-Density, High-SNM-Content Samples	(Ref. 2)		X		

[a]The references represent some typical applications.

[b]The NDA technique is being used routinely in the nuclear industry.

relative densities of sample components (Ref. 6). Corrections can usually be made, when necessary, for effects of cladding thickness and the relative densities of uranium and matrix material.

2. Thallium-activitated sodium iodide detectors are used with an external transmission technique for measuring plutonium (Ref. 27); gamma rays in the 385-keV complex from plutonium are counted by an NaI(Tℓ) detector. The window of a single-channel analyzer is centered at the energy complex. Transmission measurements are made to correct for sample attenuation. The plutonium content of the sample is then determined from the measured gamma activity.

3. Segmented gamma scanners employing lithium-drifted germanium (Ge(Li)) detectors are used for measuring plutonium or uranium ash (Ref. 10); the 414-keV Pu-239 gamma activity, the 766-keV Pu-238 gamma activity, or the 185-keV U-235 gamma activity is measured with a collimated Ge(Li) detector. The detector, collimated so as to view about a 1/2-inch slice of the sample at any one time, scans the sample in the vertical direction. The data are corrected for self-absorption by measuring the transmission of an external source (for U-235, use a Yb-169 source; for Pu-239, use an Se-75 source; for Pu-238, use a Cs-137 source) through the sample correction applied as a function of vertical height of the sample to minimize errors due to sample inhomogeneities. The measured activities are used to determine the amount of fissile material in the sample.

4. Automated Ge(Li) scanners are used for measuring uranium/plutonium mixed-oxide fuel pins (Refs. 14 and 15); a Ge(Li) detector is used to measure the intensity of the 414-keV gamma rays from Pu-239, the 185-keV gamma rays from U-235, and gamma rays from other heavy isotopes. To increase the efficiency for measuring a large throughput of the same type of samples, an automated sample changer scans the samples. The measured 414-keV gamma activity determines the amount of Pu-239 content; other isotopes from plutonium can also be assayed by measuring their respective gamma activities (see Appendix A).

5. High-resolution Ge(Li) detectors are used for measuring plutonium nitrate (Ref. 24); a high-resolution Ge(Li) detector is used to measure the intensity of the 129-keV (Pu-239), 149-keV (Pu-241), 152-keV (Pu-238), 160-keV (Pu-240), and 414-keV (Pu-239) gamma rays from plutonium nitrate solutions. The ratio of the measured intensities of the 129-keV and 414-keV gamma-ray lines is used to correct for attenuation in the sample. The measured activities determine the total plutonium content of the sample and the Pu-240, Pu-241, and Pu-238 content relative to Pu-239.

3.2.2 Calorimetry Method

1. Portable dry calorimeters are used for measuring mixed-oxide fuel rods (Ref. 13); a calorimeter measures the amount of heat generated by the natural radioactive decay of nuclear materials. For example, Pu-239 nuclei decay by alpha-particle emission, with each decay producing about 5.5-MeV kinetic energy plus the energy associated with gamma radiation. Most of this energy is absorbed in the sample itself and is converted into heat (for Pu-239, the natural rate of heat production is about 2 milliwatts per gram of material). The portable dry calorimeter measures the amount of additional electrical heating power required to maintain a fuel rod at a specific temperature higher than that which could be maintained by the heat produced in radioactive decay of nuclear material within the rod. If the isotopic composition of the rod is known, the nuclear material content of the sample can be determined from the difference in electrical heating power required to maintain a fuel rod at the same temperature as a dummy rod not containing fuel.

2. An isothermal twin-bridge calorimeter is used for measuring plutonium oxide feed material (Ref. 19); this calorimeter measures the difference in heat output between the known output of a standard or electric heater and the output from the unknown sample. There are several different techniques used to make this measurement, all of which employ a measurement of the temperature difference between the sample chamber and the chamber containing the known sample or reference heater. This temperature difference is measured with thermoresistive elements placed inside each chamber and connected in a Wheatstone Bridge configuration. If the isotopic content of the sample is known, the quantity of nuclear material in the sample can be determined from the measured heat output from the reference chamber and the Wheatstone Bridge measurement of the temperature difference between the chambers.

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3.2.3 Neutron Coincidence Counting

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An He-3 neutron coincidence counter is used for measuring U-238 incinerator ash (Ref. 11); neutrons emitted spontaneously from fissionable material samples are moderated and detected by moderated He-3 neutron detectors. Detector coincidence logic, designed to distinguish between fission neutrons and (α,n) neutrons, operates on the principle that each fission event produces

two to three neutrons while an (α,n) event only produces a single neutron. The fission neutrons detected from a single fission event are correlated in time; on the average, they will be detected within a time that is roughly equivalent to the neutron lifetime in the detector moderator. The number of coincidence events* is therefore related to the amount of spontaneously fissioning material in the sample. A correction is measured for the accidental coincidences that occur because of the detection of two (α,n) neutrons within the coincidence time interval. Additional measurement made with an external fission source placed adjacent to the sample can be used to correct for changes in the effective counter efficiency due to the moderating effects of different sample matrices. A known isotopic content allows the total amount of fissionable material in the sample to be determined from the measured neutron coincidence count rate.

3.2.4 Active Assay

1. An automated active coincidence counting system is used for measuring Pu-239 mixed-oxide light-water reactor (LWR) fuel rods (Ref. 22,; a low-energy Pu neutron source or a moderated Cf-252 source is used to irradiate the fuel rod (the source used depends on the scan speeds required). The multihead plastic scintillating detectors count the emissions of induced fission gamma rays and neutrons from neutron-induced fission primarily occurring in the fissile material in the rod. The detectors use nanosecond coincidence logic to sort out fission events (multiple emissions of gamma rays and neutrons) from background radiation. The coincidence count rate is directly related to the amount of fissile material within the rod.

2. An active isotopic source assay system is used for measuring high enrichment U-235 oxides (Ref. 9); a collimated beam of neutrons from an external source is used to stimulate fissions in the assay sample. When a fission event occurs in the sample, two to three neutrons and about seven gamma rays are emitted simultaneously. An array of fast-response, plastic scintillation radiation detectors detects two or more of the emitted fission neutrons and gamma rays in coincidence. The coincidence requirement discriminates against other reactions occurring within the sample that produce fewer particles per event, as well as direct interactions of the source neutrons with the detectors. The coincident count rate is related to the amount of fissionable material in the sample.

4. ACCURACY OF NDA MEASUREMENTS

The relative accuracy of NDA techniques used for attribute scanning may vary widely depending on the sample type and the measurement system. However, it is important to select the appropriate NDA technique for an application and to optimize the instrument-operating conditions so that the best relative accuracy of the measurement may be obtained.

As shown by investigators referenced in Table 1, relative accuracy of ±0.17% is obtainable for measuring low-enrichment U-235 fuel pellets with a 2-in x 2-in sodium iodide detector, while relative accuracies of 4 to 5% are obtainable for assaying plutonium nitrate solution with a high-resolution lithium-drifted germanium detector. Relative accuracies of less than ±1% are obtainable for assaying plutonium with high-performance calorimeters. As for the neutron coincidence counting technique, relative accuracy of a few percent is obtainable for assaying plutonium oxides, while relative accuracy of about 3% is obtainable for assaying uranium ash. When using active assay techniques, relative accuracies of ±1% and ±0.5% are obtainable for assaying LWR plutonium mixed-oxide and LWR U-235 fuel pellets, respectively.

5. CONCLUSION

General conclusions regarding the considerations in selecting and using NDA techniques for verification of the quantity of SNM stored in non-tamper-safed containers are given below.

5.1 Technique Selection

It is important that the NDA technique selection be based on attainable accuracy after considering the characteristics of the SNM sample. The measurement performance required may be such that, when the uncertainty corresponding to the remeasurement is combined with the

*A coincidence event is one in which a pair of neutrons are detected within a specified time interval.

uncertainties corresponding to the other inventory components, the quality constraints on the total limit of error of the material unaccounted for (LEMUF) will be satisfied. However, it should be realized that in some cases no single technique appears capable of meeting all the requirements. When more than one type of information is required to separate a composite response, more than one NDA technique may be required to provide that information.

5.2 NDA System Specifications

It is important that NDA systems selected for verification measurements be designed to meet the following objectives:

1. Operational characteristics are essentially independent of external background radiation, temperature, humidity, and line voltage, or these variables must be closely controlled.

2. System response is essentially independent of the isotopic concentration, matrix of the SNM sample, measurement geometry, and the stimulating source intensity and energy.

3. Systems are designed to allow reproducible positioning of the detector and item being assayed.

4. With a choice of high-resolution Ge(Li) gamma-ray detection systems, the system has the performance specifications described in Regulatory Guide 5.9, "Specifications for Ge(Li) Spectroscopy Systems for Material Protection Measurements."

5. With a choice of high-efficiency NaI(Tl) gamma-ray detection systems, the system has a resolution of Full-Width-Half Maximum less than 16% at the 185.7-keV energy peak of uranium-235, which is generally considered to be adequate for measuring uranium enrichment.

6. With a choice of neutron coincidence counters, the system has high-detection efficiency and is capable of operating in the presence of gamma radiation. In addition, it is important that the instrument supplier provide the performance information that is required for estimating errors regarding the precision of the coincidence response as a function of the real coincidence counting rate, the background to real coincidence ratio, and the relative coincidence response from a point of source of fission radiation as a function in the counting chamber.

5.3 Other Considerations

To obtain the optimum results in verification measurements, it is also important that:

1. SNM containers be selected consistent with the capability and sensitivity of the NDA system, with uniformity of wall thickness and material composition.

2. Adequate collimators be used for both gamma and neutron coincidence counting systems.

3. In implementing the verification or prior measurement program, the statistical aspects of the program also be considered. Various sampling plans and statistical tests may be used to ensure the validity of prior measurements within the original limits of error. Verification measurements should be performed at a level of confidence equivalent to the original measurement for the type of material; for example, a plan for sampling fuel pellets should be equivalent to the original sampling plan. The application of such statistical principles and concepts to inventory verification is discussed in a proposed Regulatory Guide entitled "Statistical Aspects of Prior Measurements Verification."

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REFERENCES

1. Regulatory Guide 5.13, "Conduct of Nuclear Material Physical Inventories," U.S. Nuclear Regulatory Commission (Nov. 1973).

2. O'Hara, F., et al., "Calorimetry for Safeguards Purposes," Mound Laboratory Report MLM-1798 (Jan. 1972).

3. Sher, R., "Operating Characteristics of Neutron Well Coincidence Counters," Brookhaven National Laboratory Report BNL-50332 (Jan. 1972).

4. Gunderson, G., et al., "Enrichment Measurement in Low Enriched ^{235}U Fuel Pellets," Proc. 13th Annual INMM Meeting (1972).

5. Miller, H., et al., "Results of Typical Nondestructive Measurements of Fissionable Material in Various Forms Using NNC Machines," Proc. 12th Annual INMM Meeting (1971).

6. Reilly, T. D., et al., "Nuclear Safeguards R&D," Los Alamos Scientific Laboratory Report LA 4705-MS(1971), p. 16.

7. Reilly, T. D., et al., "Nuclear Safeguards R&D," Los Alamos Scientific Laboratory Report LA-4605-MS(1970), p. 19.

8. Reilly, T. D., et al., "Nuclear Safeguards R&D," Los Alamos Scientific Laboratory Report LA-4605-MS(1970), p. 37.

9. Gallagher, W. J., "Evaluation of ISAS Under Commercial Operating Conditions," Brookhaven Office Report BHO-67-4.

10. Reilly, T. D., "Assay Experience with Monal at Oak Ridge," Proc. 13th Annual INMM Meeting (1972).

11. Foley, J. E., "Nuclear Safeguards R&D," Los Alamos Scientific Laboratory Report LA-5705-MS(1970), p. 18.

12. Bramblett, R. L., et al., "Applications of Photoinduced Reactions to Nuclear Materials Safeguards Problems," General Atomic RT Report Gulf-RT-A-10914 (Dec. 1971).

13. Beyer, N. S., et al., "A Portable Dry Calorimeter for the Nondestructive Assay of Mixed Oxide Fuel Rods," Proc. 13th Annual INMM Meeting (1972).

14. Beyer, N. S., et al., "Four Passive Assay Techniques Applied to Mixed Oxide Fuel," Argonne National Laboratory Report ANL-7907 (1972).

15. Perry, R. B., et al., "Measurement Reliability of Destructive and Nondestructive Techniques of Assaying a Reactor Fuel Inventory," Proc. 11th Annual INMM Meeting (1970).

16. Lovett, J. E., et al., "Plant Instrumentation Program," Third Quarterly Report, Brookhaven Office Report BHO-69-3 (April 1970).

17. Perry, R. B., et al., "Nondestructive Assay of ZPPR Fuel," Proc. 9th Annual INMM Meeting (1968).

18. Beyer, N. S., et al., "Controlling Large Quantities of Plutonium Fuel," Proc. 10th Annual INMM Meeting (1969).

19. O'Hara, F., et al., "Calorimetric Determination of Plutonium in Bulk Reactor Materials," Mound Laboratory (June 1971).

20. Bishop, D. M., et al., "Plant Instrumentation Program," Third Quarterly Report, General Electric Company Report GEAP-12114, Oct.-Dec. 1970.

21. Wadelsamper, D. C., et al., "Integrated Safeguards Experiment," Fourth Quarterly Report, General Electric Company Report GEAP-12114-7, April-June 1971.

22. Miller, H., "LWR Fuel Rod Nondestructive Assay Measurement Using NNC Production and Portable Systems," Proc. 13th Annual INMM Meeting (1972).

847 ~~267~~ 269

23. Menlove, H. O., et al., "Nuclear Safeguards R&D," Los Alamos Scientific Laboratory Report LA-4456-MS (1970), p. 26.

24. Cline, J. E., et al., "A Technique for Assay of L-10 Bottles of Plutonium Nitrate," Idaho Nuclear Corporation Report IN-1433 (1970).

25. Cline, J. E., "A Nondestructive Technique for Assay of Load-out Bottles of Plutonium Nitrate Solution," International Meeting on Nondestructive Measurements and Identification Techniques in Nuclear Safeguards, Italy, Sept. 1971.

26. Limpert, J. C., "Plant Instrumentation Program," Final Quarterly Progress Report, Westinghouse Cheswick Report WCAP-7562-4 (May 1971).

27. Lawless, J. L., and Chanda, R., "A Plutonium Waste Counter," Trans. Am. Nucl. Soc. 13, p. 748 (June 1970).

28. Bramblett, R. L., et al., Application of Photoinduced Reactions to Nuclear Material Safeguards Problems," General Atomic RT Report Gulf-RT-A 10826 (1971).

29. Reilly, T. D., and Parker, J. L., "A Guide to Gamma-Ray Assay for Nuclear Material Accountability," Los Alamos Scientific Laboratory Manual LA-5700 (March 1975).

847 ~~268~~ 270

BIBLIOGRAPHY

General References

American National Standard, "Guide to Calibrating Nondestructive Assay Systems," ANSI N15.20-1975.

American National Standard, "Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Control," ANSI N15.22-1975.

Auguston, R. and Reilly, T., "Fundamentals of Passive Nondestructive Assay of Fissionable Material," Los Alamos Scientific Laboratory Manual LA-5651-M (Sept. 1974).

Bingham, C. D., et al., "Nondestructive Assay Measurements Can Be Traceable," Nuclear Materials Management, Vol. V, No. II, p. 32 (1976).

Dragnev, T., "Non-Destructive Assay Techniques for Nuclear Safeguards Measurements," Atomic Energy Reviews 11, No. 2, p. 341 (1973).

George, R. S., et al., "Inspector Measurement Verification Activities," Proc. 16th Annual INMM Meeting (1975).

Kull, L. A., "Catalogue of Nuclear Material Safeguards Instruments," Brookhaven National Laboratory Report BNL-17165 (August 1972).

Reilly, T. D., and Evans, M. L., "Measurement Reliability for Nuclear Material Assay," Los Alamos Scientific Laboratory Report LA-6574 (1976).

Smith, D. B., and Forster, R. A., "NDA Measurement Error Due to Instrument Calibration," Proc. 15th Annual INMM Meeting (1974).

Gamma-Ray Spectrometry

Cline, J. E., et al., "Gamma Rays Emitted by the Fissionable Nuclides and Associated Isotopes," Aerojet Nuclear Company Report ANCR-1069 (Supplement To IN-1448) (July 1972).

Cline, J. E., "Gamma Rays Emitted by the Fissionable Nuclides and Associated Isotopes," Idaho Nuclear Corporation Report IN-1448 (Jan. 1971).

Dragnev, T., and Scharf, K., "Nondestructive Gamma Spectrometry Measurement of $^{239}Pu/^{240}Pu$ and $Pu/^{240}Pa$ Ratios," Int. J. App. Rad. and Isotopes, 26, p. 125 (1975).

Gunnink, R., and Tinney, J. F., "Analysis of Fuel Rods by Gamma-Ray Spectroscopy," Lawrence Radiation Laboratory Report UCRL-51086 (Aug. 1971).

Gunnink, R., "Plutonium Isotopic Measurements by Gamma-Ray Spectrometry," Proceedings of the Symposium on Calorimetric Assay of Plutonium, Mound Laboratory Report MLM-2177, p. 45 (1973).

Gunnink, R., "A Simulation Study of Plutonium Gamma-Ray Groupings for Isotopic Ratio Determinations," Lawrence Livermore Laboratory Report UCRL-51605 (1974).

Gunnink, R., et al., "A System for Plutonium Analysis by Gamma-Ray Spectrometry: Part 1: Techniques for Analysis of Solutions. Part 2: Computer Programs for Data Reduction and Interpretation," Lawrence Livermore Laboratory Report UCRL-51577, Parts 1 and 2 (1974).

Gunnink, R., "Status of Plutonium Isotopic Measurements by Gamma-Ray Spectrometry," Lawrence Livermore Laboratory Report UCRL-76418 (1975).

Gunnink, R., et al., "A Re-evaluation of the Gamma-Ray Energies and Absolute Branching Intensities of ^{237}U, $^{238,239,240,241}Pu$, and ^{241}Am," Lawrence Livermore Laboratory Report UCRL-52139 (1976). (This is an update of UCRL-51087, 1971.)

Haas, F. X., "Nondestructive Measurement of Plutonium Isotopic and Americium Concentrations Using Gamma-Ray Spectrometry," Proceedings of the Symposium of Calorimetric Assay of Plutonium, Mound Laboratory Report MOM-2177, p. 60 (1973).

847 ~~269~~ 271

Helmer, R. G., et al., "Gamma-Ray Energy and Intensity Measurements with Ge(Li) Spectrometers," The Electromagnetic Interaction in Nuclear Spectroscopy, Chapter 17, North-Holland (1975).

Higinbotham, W., "Passive Gamma-Ray Scanning of Plutonium Recycle Fuel Rods," Brookhaven National Laboratory Report BNL-50351 (June 1972).

Kraner, H. W., "On the Use of Gamma-Ray Spectroscopy To Determine Pu Isotopic Abundances in Plutonium Sources," Brookhaven National Laboratory Report BNL-50237 (May 1970).

Kull, L. A., et al., "Guidelines for Gamma-Ray Spectroscopy Measurements of ^{235}U Enrichment," Brookhaven National Laboratory Report BNL-50414 (March 1974).

Kull, L. A., "An Introduction to Ge(Li) and NaI Gamma-Ray Detectors for Safeguards Applications," Argonne National Laboratory Report ANL/ACEA-103 (Sept. 1974).

Reilly, T. D., and Parker, J. L., "A Guide to Gamma-Ray Assay for Nuclear Material Accountability," Los Alamos Scientific Laboratory Manual LA-5794-M (March 1975).

Umezawa, H., et al., "Gamma-Ray Spectrometric Determination of Isotopic Ratios of Plutonium," Japanese Atomic Energy Research Institute Report JAERI-6295 ((1975).

Calorimetry

Brumbach, S. B., et al., "Plutonium Calorimetry and SNM Holdup Measurements," Argonne National Laboratory Report ANL-77-8 (NUREG-0182) (1976).

Ditmars, D. A., "Precision Measurement by Bunsen Ice Calorimetry of the Total Decay Power of a Plutonium Standard Heat Source," a paper presented at the 30th Annual Calorimetry Conference (1975). (D. A. Ditmars, National Bureau of Standards, Washington, D.C. 20234).

Fellers, C. L. and Seabaugh, P. W., "Predication of Calorimeter Equilibrium," Proc. 17th Annual INMM Meeting (1976).

Gunn, S. R., "Radiometric Calorimetry: A Review," Nuclear Instruments and Methods, Vol. 29, pp. 1-24 (Sept. 1964).

Hansen, L. D., et al., "A Heat Conduction Calorimeter for the Measurement of Solid Heat-Producing Samples," a paper presented at the 31st Annual Calorimetry Conference (1976); (L. D. Hansen, Tronac, Inc., Orem, Utah).

O'Hara, F., et al., "The Use of Calorimetry in Nuclear Materials Management," Mound Laboratory (June 1970).

O'Hara, F., et al., "Calorimetry for Safeguards Purposes," Mound Laboratory Report MLM-1798 (Jan. 1972).

O'Hara, F., et al., "Calorimetric Determination of Plutonium in Bulk Reactor Materials," Mound Laboratory (June 1971).

Perry, R. B., et al., "An Analytical Model for a Fast Response Calorimeter - With Applications," Proc. 17th Annual INMM Meeting (1976).

Stephens, H. P., "A Refrigerant-11 Calorimeter for Measurement of Steady-State Powers," a paper presented at the 31st Annual Calorimetry Conference (1976); (H. P. Stephens, Sandia Laboratories, Albuquerque, N.M. 87115).

Strohm, W. W., and Havenstein, M. F., "Proceedings of the Symposium on the Calorimetric Assay of Plutonium, October 24-25, 1973," Mound Laboratory Report MLM-2177 (Oct. 1974).

Neutron Measurements (Passive and Active)

Atwell, T. L., et al., "Assay of Plutonium Metal Buttons with a Portable Neutron Counter," Los Alamos Scientific Laboratory Report LA-5431-PR, p. 20 (1973).

Bramblett, R. L., "Fuel Rod Scanner for Quality Control and Safeguards Transaction," IEEE Nuclear Science Symposium (1974).

847 ~~270~~ 272

Bramblett, R. L., "Passive and Active Measurement of SNM in 55- to 83-Gallon Drums," Proc. 16th Annual INMM Meeting (1975).

Foley, J. E., "4π-Neutron Counter for 55-Gallon Barrels," Los Alamos Scientific Laboratory A-1 Progress Report, LA-4605-MS, p. 25 (1970).

Forster, R. A., and Menlove, H. O., "Neutron Coincidence Detector for Fuel Pins: Description and Operating Procedures Manual," Los Alamos Scientific Laboratory Manual LA-5156-M (Feb. 1973).

Menlove, H. O., "Nondestructive Assay of HTGR Fuel Rods," Proc. 16th Annual INMM Meeting (1975).

Menlove, H. O., et al., "A Multi-Spectra Neutron Irradiation Technique for the Nondestructive Assay of Fissionable Materials," Nucl. Technology 10, p. 366 (1971).

Sampson, T. E., "Neutron Yields from Uranium Isotopes in Uranium Hexafluoride," Nucl. Engr. Sci. 54, No. 4, p. 470 (1974).

Sher, R., "Operating Characteristics of Neutron Well Coincidence Counters," Brookhaven National Laboratory Report BNL-50322 (Jan. 1972).

Smith, D. B., et al., "An Automated Nondestructive Assay System for the Measurement of Irradiated High-Enriched Uranium Fuel," Proc. 16th Annual INMM Meeting (1975).

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APPENDIX A

MAJOR GAMMA-RAY SIGNATURES FOR THE FISSIONABLE ISOTOPES*

Isotope	Energy (keV)	Intensity $(g\text{-}s)^{-1}$	Comments
^{235}U	185.72	4.3×10^{4}	Only intense gamma ray. Resolved with NaI as well as Ge(Li). Useful for enrichment and quantitative measurements. Several much weaker peaks are seldom useful.
^{238}U	1001.10 766.40	1.0×10^{2} 3.9×10^{1}	These actually arise from the ^{234m}Pa daughter of ^{238}U. After chemical separation, about 100 days are required for the activity to come into equilibrium at the levels stated. Plutonium-238 gives rise to the same 766.40-keV gamma and would produce interference in U-Pu mixtures. Useful for work with Ge(Li) or NaI.
^{238}Pu	766.40 152.77	1.5×10^{5} 6.5×10^{6}	Most useful for quantitative assay. Ge(Li) or NaI. Useful for isotopic determinations with Ge(Li).
^{239}Pu	413.69	3.4×10^{4}	The 413.69 usually provides the basis for Ge(Li) assays. The 413.69 plus the 375.02 and its weak neighbors form a complex upon which NaI assays are based.
^{239}Pu	129.28	1.4×10^{5}	Useful for isotopic determinations with Ge(Li). Plutonium-239 has over 100 gamma rays, some of which are useful for careful work with Ge(Li).
^{240}Pu	--	--	Several weak gamma rays, but all suffer bad interference from gammas of other isotopes. Requires very careful work with high-resolution detector to make use of any of them.
^{241}Pu	207.98	2.0×10^{7}	Actually from ^{237}U daughter and requires about 25 days after chemical separation to come into equilibrium at stated value. May also have a few percent interference from ^{241}Am, which emits same gamma. Nevertheless a good, clean strong gamma useful with both NaI and Ge(Li).
	164.59	1.8×10^{6}	Useful with Ge(Li). Also from ^{237}U.
	148.60	7.5×10^{6}	Useful with Ge(Li). Direct from ^{241}Pu.
^{241}Am	59.54	4.6×10^{10}	Very strong gamma but attenuation problems. Useful with Ge(Li) or NaI. Has several other much less intense gammas sometimes useful for Ge(Li) work.
^{242}Pu	--	--	No useful gamma rays at all. Nature failed us at this point.

*This table is taken from Reference 29.

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APPENDIX B

ASSIGNED HALF-LIVES AND SPECIFIC POWERS
OF PLUTONIUM ISOTOPES AND AMERICIUM-241

Radionuclide	Half-Life* (years)	Decay Constant (day^{-1})	Specific Power* (mW/g)
Pu-238	87.79 ± 0.08 (0.09%)	2.1617×10^{-5}	567.16 ± 0.57 (0.10%)
Pu-239	24 082 ± 46 (0.19%)	7.830×10^{-8}	1.9293 ± 0.0053 (0.27%)
Pu-240	6 537 ± 10 (0.2%)	2.903×10^{-7}	7.098 ± 0.015 (0.2%)
Pu-241	14.35 ± 0.02 (0.14%)	1.322×10^{-4}	3.390 ± 0.002 (0.06%)
Am-241	434.1 ± 0.6 (0.14%)	4.372×10^{-6}	114.23 ± 0.16 (0.14%)

*In the table, the value of the constant is reported ± the standard deviation. The standard deviation relative to the mean, in percent, is given in parentheses.

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APPENDIX C

NEUTRON EMISSION RATES OF SOME NUCLEAR MATERIALS

Isotope	Neutron Emission Rate ($n.g^{-1}.s^{-1}$) Spontaneous Fissions	(α,n) Reactions
U-235	7.6×10^{-4}	
U-238	1.7×10^{-2}	
Pu-238	2.6×10^{3}	
Pu-239	3.0×10^{-2}	
Pu-240	1.0×10^{3}	
Pu-242	1.7×10^{3}	
Cm-242	2.3×10^{7}	
(Pu-239) F_4		4.3×10^{3}
(Pu-238) O_2		1.4×10^{4}

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