NONDESTRUCTIVE CO-ASSAY OF U AND PU FISSILE TRACES BY COMBINED DELAYED NEUTRON AND DELAYED GAMMA RAY FISSION PRODUCT NEUTRON ACTIVATION ANALYSIS

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ABSTRACT

The Delayed Neutron Delayed Gamma (DNDG) technique provides a new analytical capability to the International Atomic Energy Agency (IAEA) for detecting undeclared nuclear activities. The US National Nuclear Security Administration (NNSA) is sponsoring research aimed at expanding the capabilities of rapid nondestructive safeguards measurements by using the combination of delayed neutron (DN) and delayed gamma (DG) analysis techniques at the US Department of Energy's Oak Ridge National Laboratory High Flux Isotope Reactor (HFIR). The methods that are being developed use pneumatic tubes located at the HFIR Neutron Activation Analysis (NAA) facility to conduct rapid irradiations and subsequent measurements of samples containing fissile material. The NAA facility at HFIR has supported the IAEA's Network of Analytical Laboratories for nearly a decade by providing mass and enrichment characterization on pre-inspection check (PIC) samples collected by IAEA inspectors in the field. The measurement of short-lived fission products opens the possibility for both U and Pu to be characterized on these samples within a short analysis period (less than 15 min). The distribution profile of heavy fission products (atomic mass 125–145) remains fairly invariant for the fissile isotopes such as ^{235}U and ^{239}Pu, whereas the distribution of light fission products (atomic mass 85–105) varies from one isotope to another. By measuring the ratio of the net full energy peaks using High Resolution Gamma Spectrometry (HRGS), from a low mass isotope (^{104}Tc) and a high mass isotope (^{141}Ba) and using calibration data from pure U and Pu standards, the relative fraction of fissile isotopes in an unknown sample can be determined. Additionally, combining this gamma ray method with delayed neutron activation analysis (DNAA) provides quantification of trace fissile mass. The combination of these two measurement techniques provides picogram detection limits along with the ability to rapidly and nondestructively discriminate U and Pu isotopes. The DN and DG results were combined to demonstrate that fissile mass can be quantified, and the presence of Pu can be flagged in a binary mixture of U and Pu. By irradiating the material in the pneumatic transfer (PT) facility PT-1 at HFIR, with neutron fluxes an order of magnitude higher, the DNDG method was successfully tested using fissile masses approaching the masses of PIC swipe samples. In addition, the signal processing chain of the DN counter at the HFIR NAA laboratory has been upgraded, lowering the uncertainty in the ^{235}U equivalent mass by one-third. The results of these multifaceted approaches to improve safeguards are discussed and results are presented.

Keywords: Delayed neutrons, delayed gamma spectrometry, fission products, nondestructive assay

INTRODUCTION

The International Atomic Energy Agency's (IAEA's) Long-Term R&D plan has a stated high urgency need to develop elemental and isotopic signatures of nuclear fuel cycle activities and processes.[1] Trace levels of fissile material may be present in pre-inspection check (PIC) swipe samples collected by IAEA inspectors prior to entering a facility to be inspected. Each year, the IAEA sends hundreds of swipe samples in plastic capsules to the High Flux Isotope reactor (HFIR) at Oak Ridge National Laboratory (ORNL) for routine irradiation and delayed neutron (DN) counting. This provides a low cost and rapid means (compared to traditional chemical analysis) to detect the presence of fissile materials. The current process is nondestructive and involves straightforward physical measurement: a pneumatic target holder called a *rabbit* is used to transfer the sample to a position close to the HFIR core; then the sample is irradiated, returned, and counted in a shielded array of moderated ^{3}He neutron counters. The detection of neutrons emitted following the decay of neutron-rich fission products, which were created in the sample during irradiation, indicates the presence of fissile material. This fissile material is quantified in terms of the equivalent mass of ^{235}U that would produce the same DN count. Note that the equivalent mass does not provide isotope-specific information. The nominal 15 pg detection limit for the delayed neutron activation analysis (DNAA) technique at HFIR is much lower than the 50 pg requirement defined by the IAEA.[2] This difference is mainly a result of the high neutron flux available in HFIR (up to 4×10^{14} n·cm^{-2}s^{-1}), and the relatively high neutron counting efficiency (~35%).

Although the DNAA technique reports fissile content in units of equivalent mass of ^{235}U, ^{235}U is not the only fissile material of safeguards interest; Pu is also of high safeguards interest. Detection of Pu where it is not expected would be reason for concern. Being able to distinguish between ^{235}U and Pu isotopes would therefore augment and strengthen the DNAA technique and provide detection and deterrence for certain kinds of undeclared activities. The delayed neutron delayed gamma (DNDG) method currently being developed at ORNL complements DN counting with delayed gamma (DG) spectrometry. The DNDG method uses two signatures from active neutron interrogation to determine fission type and to characterize a swipe sample containing trace amounts of U, Pu, or a mixture of the two. Provided a continuous neutron flux, such as one from a nuclear reactor, DN emissions are proportional to the total fission rate within an irradiated material. DGs provide information on the type of fission by taking the ratio of low to high atomic mass fission products. Both of these measurement techniques are completed using comparative analysis, eliminating the need for careful detector and neutron spectrum calibration and measurements. The DNDG method was proven to work at HFIR, and it hopefully can be deployed more widely in the future.

As shown in Figure 1, the yields for high mass number (>120) fission products for the odd mass-numbered actinides are very similar, whereas the yields for the low mass (<120) fission products vary significantly. The ratio of suitable low mass to high mass fission product yields, determined using gamma spectrometry, can then be used to uniquely flag the presence of an odd mass-numbered actinide (e.g., ^{239}Pu) in a binary mixture of ^{235}U and ^{239}Pu.[3] The primary objective of the DNDG method is to flag the presence of ^{239}Pu in the sample. Quantification of ^{239}Pu and ^{235}U by combining DN and DG results is an added bonus.

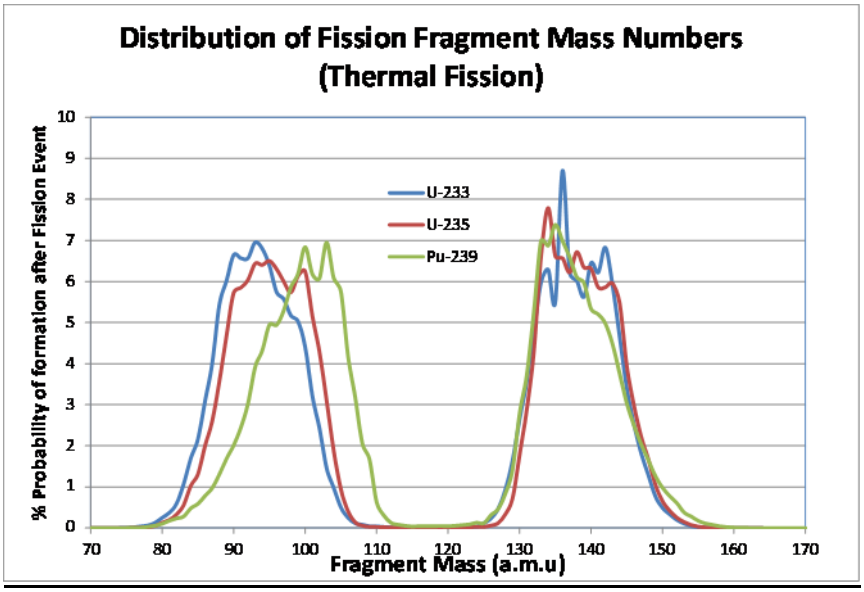

Figure 1. Distribution of fission product yields for various odd mass-numbered actinides.

Delayed Neutron Measurement Method

Cellulose samples contained in polyethylene rabbits were irradiated in HFIR's pneumatic transfer (PT) facility PT-2, for a time period t_{irr}. After the irradiation stops, precursor production ceases, and the activity produced decays with decay constant λ_i. The irradiated sample from PT-2 is transferred to the center of a ^{3}He-based neutron counter. After a period of delay following irradiation, t_{dly}, a count of duration, t_{cnt}, is begun using a neutron counter with an efficiency of ε_i counts per DNs emitted. The net number of events recorded, assuming negligible counter deadtime losses, is therefore the sum of all DN groups.

$$C = m\frac{N_A\sigma_f}{A}\varphi\sum_{flux}\sum_{i=1}^{n_{grp}}\varepsilon_i\frac{\beta_i}{\lambda_i}\left(1-e^{-\lambda_i t_{irr}}\right)e^{-\lambda_i t_{dly}}\left(1-e^{-\lambda_i t_{cnt}}\right), \quad (1)$$

where m is the mass of the fissile isotope, ϕ is the neutron flux, σ_f is the induced fission reaction cross section, A is the atomic weight of the fissile isotope, N_A is the Avogadro number, and β_i is the fraction of fissions that results in DN production with the decay constant λ_i in units of seconds^{-1}. Equation (1) is the fundamental predictive (or causal relation) physical model equation for neutron counting in the case of a single fissioning species subjected to a single irradiation-delay-count cycle. The model can be used in various ways: in an absolute sense to explore nuclear data parameters, inverted (or solved) for the flux, solved for the mass of the fissile material in unknown samples relative to known calibration measurements, or used to formulate uncertainty quantification. For the present work, the research team adopted the eight-group (n_{grp} = 8) model developed by the Nuclear Energy Agency working group.[4] In practice, the DNAA measurements conducted at the HFIR Neutron Activation Analysis (HFIR-NAA) laboratory are done via a comparator method. This method uses a known DN emission quantity (C_{known}) to scale known ^{235}U mass (m_{Uknown}) to the unknown fissile sample. This calculation is described in Eq. (2). The measured counts from an unknown are represented as C_u, and the unknown effective U mass is m_{Ueq}:

$$m_{Ueq} = C_{Ueq} * \frac{m_{Uknown}}{C_{known}} \ . \quad (2)$$

The result of this calculation is equivalent to the mass of U because DNs do not indicate the parent fissioning species. In the DNDG method, the gamma component of the measurement is used to determine the fraction of the fissile isotope in a binary mixture.

Delayed Gamma Measurement Method

After the DN counting has been completed and 3–10 days have elapsed in the same HFIR cycle, *the same cellulose sample* containing the fissile material is irradiated in PT-1. After irradiation is complete, the sample is retrieved from the irradiation location via a pneumatic transfer mechanism. The pneumatic transfer line from PT-1 delivers the sample inside a shielded cubicle located in the HFIR NAA laboratory. It is then manually transferred to a shielded high purity germanium (HPGe) detector for measuring the DGs emitted from the activation product. The PT-1 facility offers an order of magnitude higher neutron flux, thereby improving the sensitivity of the DG measurement. Figure 2 illustrates the PT-1 flight tube.

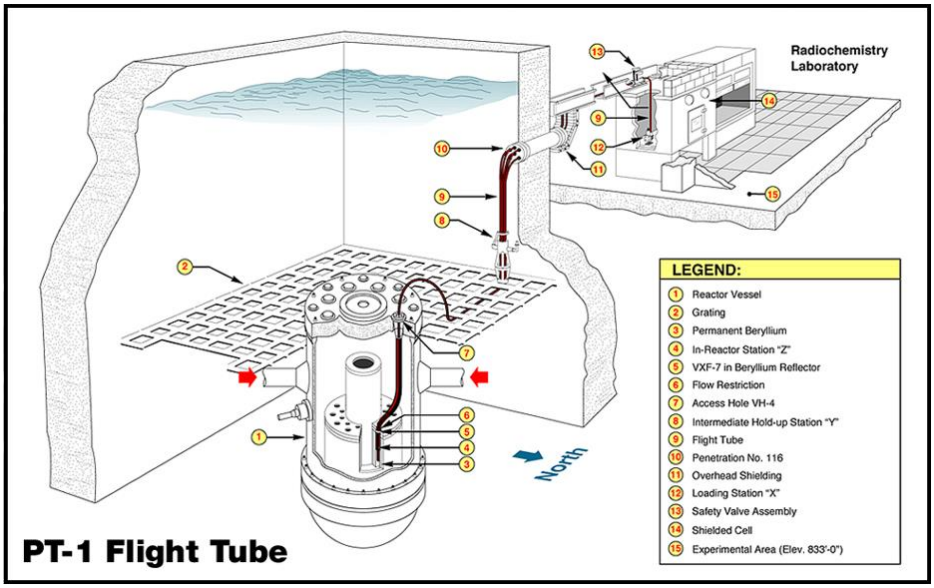

Figure 2. Flight tube diagram for PT-1 irradiation facility at HFIR.

The sample is irradiated for a time period t_0. The time elapsed between the end of irradiation and the start of data acquisition using the HPGe gamma ray spectrometer is $(t_1 - t_0)$. The sample counting time is $(t_2 - t_1)$. The net count rate in the gamma ray peak from the fission product produced from the induced fission of a given fissile isotope at the end of the irradiation period is represented by A_0. So, the saturated net count rate would be A_∞ if the sample were irradiated for an infinite time. This NAA irradiation and measurement protocol is illustrated in Figure 3. The saturated count rate is given Eq. (3):

$$A_{sat} = \frac{\lambda_i C_{net}}{\left(\left(1-e^{-\lambda_i t_0}\right)e^{-\lambda_i (t_1-t_0)}\left(1-e^{-\lambda_i (t_2-t_1)}\right)\right)}, \quad (3)$$

where λ_i is the decay probability of fission product nuclide *i* produced in the induced fission of a given fissile isotope. Saturated net count rate A_{sat} is determined for gamma ray peaks emitted by a low mass and a high mass fission product nuclide.

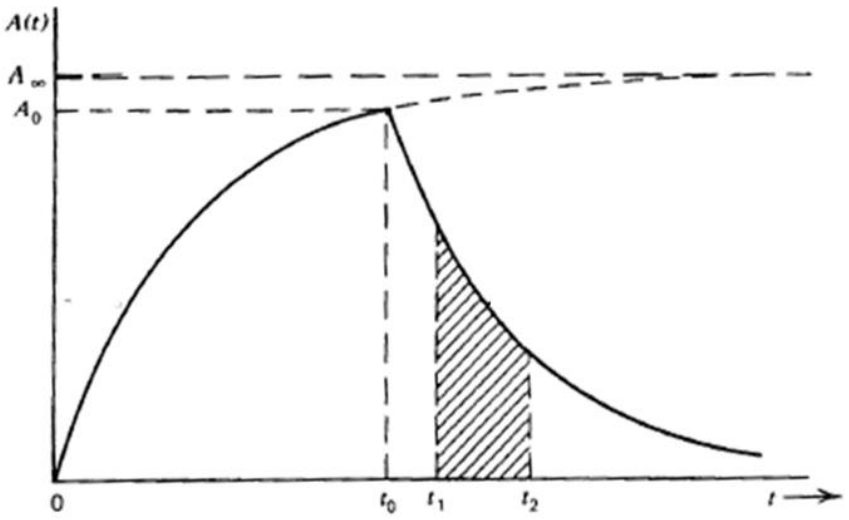

Figure 3. NAA analysis protocol.

A total of 56 potential FP ratios with half-lives ranging from 23.9 s (^{95}Sr) to 18.7 min (^{94}Y) were initially considered.[5] The list was narrowed to 15 fission product ratios, based on fission product yields, half-life, and gamma ray yields. Of the 15 fission product ratios, the ratio that gave the best performance was ^{104}Tc/^{141}Ba. Technicium-104 has a half-life of 18.3 min, and barium-141 has a half-life of 18.27 min; they have very similar decay probabilities. Therefore, instead of working with saturated count rates (Eq. 3), we can directly work with the ratio of net peak areas from ^{104}Tc and ^{141}Ba gamma rays. The gamma ray peak at 358.0 keV from ^{104}Tc (gamma ray yield = 89%) and the gamma ray peak at 190.328 keV from ^{141}Ba (gamma ray yield = 45.5%) were used in the analysis. The ratio (R) of net peak areas from ^{104}Tc and ^{141}Ba from an irradiated cellulose swipe sample, which potentially contains a binary mixture of ^{235}U and ^{239}Pu can be written as

$$R = \frac{a_4 U + b_4 P}{a_1 U + b_1 P}, \tag{4}$$

where a_4 and a_1 are the net peak areas from ^{104}Tc and ^{141}Ba gamma rays from ^{235}U respectively, b_4 and b_1 are the net peak areas from ^{104}Tc and ^{141}Ba gamma rays from ^{239}Pu respectively, and U and P are masses of ^{235}U and ^{239}Pu. For a binary mixture of U and Pu, the Pu fraction (X) can be represented as

$$X = \frac{P}{U+P}. \tag{5}$$

Substituting for the quantities U and P in Eq. (4) in terms of X, rearranging, and reducing the number of model parameters from four to three, we can write

$$R = \frac{(a_4/a_1)(1-X)+(b_1/a_1)(b_4/b_1)X}{(1-X)+(b_1/a_1)X}, 0 < X < 1. \tag{6}$$

Normalizing the ratio R with respect to U-only samples and designating $Y = \frac{R}{(a_4/a_1)}$, Eq. (6) can be rewritten as

$$Y = \frac{(1-X)+cdX}{(1-X)+cX}, \tag{7}$$

where $c = b_1/a_1$ and $d = \frac{(b_4/b_1)}{(a_4/a_1)}$.

Finally, we arrive at the quantity of interest X, the ^{239}Pu fraction:

$$X = \frac{(Y-1)}{(Y-1)+c(d-Y)}. \tag{8}$$

To determine X, the ^{239}Pu fraction using the binary mixture, we need only measure the ratio R of the net peak areas of the fission products ^{104}Tc and ^{141}Ba from the irradiated swipe samples and use a few calibration parameters that have been established based on at least one known standard each that contains U only and Pu only. The calibration parameters that need to be established are the ratio of the net peak areas a_4/a_1 based on a U-only standard, the ratio of net peak areas b_4/b_1 based on a Pu-only standard, the parameter d, and the parameter c. The method is purely dependent on the measurement of ratios of net peak areas, rather than absolute quantities such as cross sections, neutron flux, detector efficiencies, activities, or masses. This dependence is a great advantage because the systematic uncertainties arising from nuclear data and detector efficiencies can be avoided entirely. The uncertainty in the ^{239}Pu fraction X in the binary mixture can be determined from first principles by propagating the uncertainties arising from counting statistics and comparator mass values.

EXPERIMENTAL DETAILS

The newly developed DNDG method was experimentally validated by irradiating a set of ten U and Pu standards of known masses, both pure and binary mixtures of U and Pu, in HFIR and measuring the gamma ray emissions from the fission products using an energy-calibrated HPGe detector. The measured ^{239}Pu fraction was compared against the known ^{239}Pu fraction.

The concept of operations for the DNDG method is a two-step process. The first step is the irradiation of the sample in HFIR location PT-2 and the measurement of DN emissions from the irradiated sample. The second step is the irradiation of the same sample in HFIR location PT-1, 3–10 days after the first irradiation, and measurement of DG emissions from fission product decays using an HPGe detector. The irradiation in PT-1 is to take advantage of neutron flux that is an order of magnitude higher in PT-1 (4.0×10^{14} n·cm^{-2}s^{-1}) compared with PT-2 (4.0×10^{13} n·cm^{-2}s^{-1}). The higher flux in PT-1 translates into improved sensitivity for the DG analysis. The ^{235}U and ^{239}Pu masses of the standards prepared for the DNDG validation exercise are given in Table 1. Samples DNDG-8, DNDG-9, and DNDG-10 were not irradiated in PT-2 and therefore no delayed neutron data is available.

Table 1. Uranium-235 and plutonium-239 standards used in DNDG validation

Sample ID	^{235}U mass (ng)	^{239}Pu mass (ng)	Fraction ^{239}Pu	DN (counts)	Uncertainty 1σ (counts)
DNDG-1	5.071	0.000	0.000	20,360	312.5
DNDG-2	4.063	0.977	0.194	18,707	301.7
DNDG-3	3.006	2.033	0.403	18,352	299.4
DNDG-4	0.000	4.974	1.000	15,345	278.6
DNDG-5	5.125	0.000	0.000	20,663	317.2
DNDG-6	5.193	0.000	0.000	20,876	320.4
DNDG-7	0.000	5.121	1.000	16,981	290.1
DNDG-8	0.000	5.000	1.000	N/A	—
DNDG-9	0.000	5.017	1.000	N/A	—
DNDG-10	0.000	5.045	1.000	N/A	—

Irradiation in PT-2 and Delayed Neutron Counting

First, the samples were irradiated for 180 s at HFIR location PT-2. The irradiations were performed during HFIR cycle 490. Decay time before neutron count and measurement period are 5 and 60 s, respectively. The samples were retrieved and automatically located at the center of a ^{3}He-based neutron counter consisting of 18 ^{3}He proportional tubes embedded in polyethylene, as shown in Figure 4. Each tube has a diameter of 5 cm, an active length of 30 cm, and fill pressure of 5 atm. A polyethylene moderator surrounds the detectors and flight tube and forms a cube approximately 45.72 cm (18 in.) in each dimension. Lead shielding, 5.08 cm (2 in.) thick, is molded around the flight tube and air exit line at the counting position to provide personnel shielding.

Figure 4. DN counter at the HFIR NAA laboratory

The signal processing electronics in the DN counting chain have been upgraded in the recent years. The new ORNL-engineered board consists of a charge-sensitive preamplifier, a five-pole shaper, and a fast discriminator. The output of the preamplifier board is converted into a 50 ns wide TTL signal by the processor board, shown in Figure 5. The NIM-based analog signal train is now 100% digital-processing electronics.

Because the new preamplifiers, shown in Figure 5, are cylindrical and coaxial with respect to the ^{3}He tubes, long cables, which were contributing to noise and loss of sensitivity, are no longer required. The upgrades to the DN counting signal processing chain have increased the reliability of the DN counter and have lowered DN counting uncertainty by a factor of three. The DN emissions from the irradiated samples were measured using the upgraded DN counter, and these results are listed in Table 1.

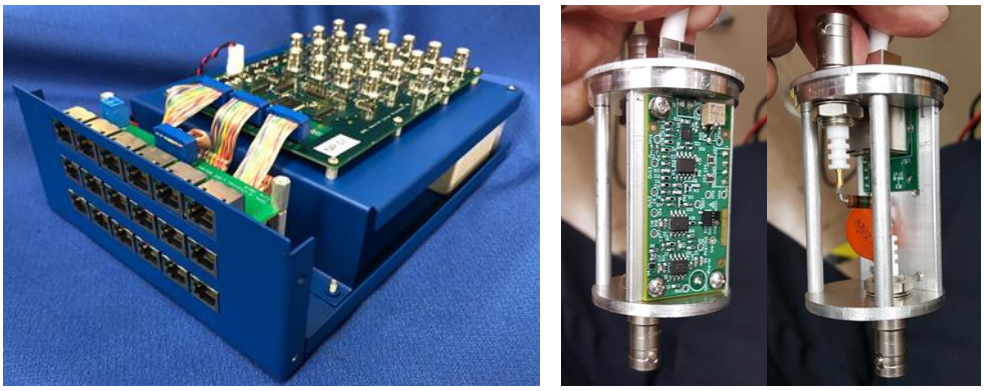

Figure 5. Processor board (*left*) and preamplifiers (*right*) in the DN electronics chain

Irradiation in PT-1 and Delayed Gamma Measurement

Each of the ten standards listed in Table 1 was irradiated at HFIR location PT-1 for 300 s. At the end of irradiation, each sample was allowed to cool for 1,200 s. The sample was then manually transferred to a calibrated HPGe detector measurement station at a distance of 20 cm from the detector. Each sample was counted for a live time of 1,800 s. The HPGe detector was configured with a Canberra LYNX digital signal processor (DSP) employing loss-free counting.[6] Peak analysis was performed using the Peak Easy[7] software package by setting up left and right continuum regions in the gamma ray spectrum on either side of the full energy peak of interest. This approach was selected so that the statistical uncertainty in the net peak area could be determined from first principles. The net peak areas at 358 keV (^{104}Tc) and 190 keV (^{141}Ba) and their uncertainties were determined. The calibration parameters *c* and *d* were established based on irradiation of three U-only samples and five Pu-only samples. The mean and standard deviation of the calibration parameters are $c = 3.00 \pm 0.15$ and $d = 1.61 \pm 0.13$. The measured DN counts, $C_{fissile}$, are expressed by Eq. (9)

$$C_{fissile} = C_{U235} + C_{Pu239}\,, \tag{9}$$

where C_{U235} and C_{Pu239} are DN counts from ^{235}U and ^{239}Pu (both unknown). As shown in Eq. (10), C_{U235} and C_{Pu239} can be determined by multiplying the measured DN counts, $C_{fissile}$, by the fractions of ^{235}U and ^{239}Pu, respectively:

$$C_{Pu239} = C_{fissile} * \mathrm{X} \text{ and} \tag{10}$$

$$C_{U235} = C_{fissile}(1 - X). \tag{11}$$

In Eqs. (10) and (11), *X* is the fraction of ^{239}Pu in the binary mixture, as determined by the DG method; $(1 - X)$ is the ^{235}U fraction. The final step is to convert the apportioned counts from ^{235}U

and ^{239}Pu to their respective mass values by using pure ^{235}U and ^{239}Pu standards with known values of delayed neutron counts per second per unit mass of the fissile material.

Results and Discussion

The ^{239}Pu fraction results for seven out of ten standards, listed in Table 2, are within 1σ of the measurement uncertainties, and the results for three of the standards are within 2σ. The results for the two binary mixtures, namely DNDG-2 and DNDG-3, are within 1σ of the measurement uncertainties. For U-only standards, the measured ^{239}Pu fractions are either small positive or small negative values and overlap zero to within the uncertainty limits. Thus, the experimental results based on the known standards validate the DG method for reliably determining the ^{239}Pu fraction.

Because the masses of the standards that were irradiated are on the order of a few nanograms, it is possible to quantify ^{239}Pu masses within 1-σ or 2-σ confidence When the method is applied to sample masses that are close to the PIC sample range (e.g., 50 pg), the presence of Pu can be flagged, but reliable quantification of ^{239}Pu mass may not be possible because of poor measurement precision. Table 2 gives the mass values of ^{239}Pu and ^{235}U in each standard that was irradiated in PT-2 and PT-1.

Table 2. Results for ^{239}Pu and ^{235}U quantification

Sample ID	^{239}Pu fraction (DG)	^{239}Pu fraction (known)	^{239}Pu fraction Devn./σ	^{235}U mass (DNDG) (ng)	Devn./σ (^{235}U) mass, ng	^{239}Pu mass (DNDG) (ng)	Devn./σ (^{239}Pu) mass, ng
DNDG-1	0.017 ± 0.019	0.000	0.894	4.980 ± 0.121	-0.756	0.100 ± 0.111	0.894
DNDG-2	0.216 ± 0.032	0.194	0.695	3.646 ± 0.170	−2.451	1.195 ± 0.191	1.140
DNDG-3	0.407 ± 0.036	0.403	0.105	2.705 ± 0.170	−1.762	2.207 ± 0.199	0.874
DNDG-4	1.043 ± 0.028	1.000	1.544	−0.163 ± 0.106	−1.543	4.724 ± 0.152	−1.643
DNDG-5	−0.011 ± 0.017	0.000	−0.654	5.196 ± 0.118	0.599	−0.068 ± 0.104	−0.654
DNDG-6	−0.005 ± 0.017	0.000	−0.294	5.218 ± 0.121	0.212	−0.032 ± 0.108	−0.294
DNDG-7	0.981 ± 0.029	1.000	−0.670	0.081 ± 0.121	0.670	4.918 ± 0.166	−1.223
DNDG-8	0.963 ± 0.029	1.000	−1.243	—	—	—	—
DNDG-9	0.975 ± 0.029	1.000	−0.865	—	—	—	—
DNDG-10	1.041 ± 0.028	1.000	1.466	—	—	—	—

The ratio of deviation/σ values listed in Table 2 assume that the standard is known perfectly. It must be noted that there are two different and challenging results shown in Table 2. First is the ^{239}Pu mass fraction. The second is the absolute mass of both ^{235}U and ^{239}Pu. In all cases, the fissile material has been unambiguously identified. For ^{235}U mass result for the sample DNDG-2 is −2.45, whereas the corresponding result for ^{239}Pu for the same standard is only 1.14. The larger deviation in the ^{235}U mass result for this standard is currently under investigation. All other results for both ^{235}U and ^{239}Pu masses are within 1σ or 2σ of the measurement uncertainties. The error structure in the mas values is expected to be correlated.

CONCLUSIONS

The DNDG method is a new analytical technique that was developed at the HFIR NAA laboratory at ORNL. It provides a new analytical capability to the IAEA for detecting undeclared nuclear activities. The new method combines the superior sensitivity of DN measurement with the isotopic specificity of the DG method to indicate the presence of Pu in a binary mixture of U and Pu swipe samples. The method requires only a limited number of calibration measurements using ^{235}U and ^{239}Pu standards. The signal processing chain of the DN counter at the HFIR NAA laboratory has been upgraded with a low-noise preamplifier, shaper, and fast discriminator board engineered at ORNL. The DG measurement of the ratio of gamma ray full energy peaks from low (^{104}Tc) and high (^{141}Ba) mass fission products eliminates the need to use nuclear data, such as gamma ray yields and fission product yields, in the analysis. By measuring the calibration standards and the samples at the same source-detector geometry, the need for establishing the peak efficiency of the HPGe detector was eliminated. The systematic uncertainties in the measurement protocol were thus minimized. The DNDG method was validated experimentally using ten U and Pu cellulose standards. The deviations between the DNDG measurement results vs. known values were consistent with the propagated measurement uncertainties. Should lower detection limits be required there are various possibilities. DN counting efficiency could be increased to 80% and multiple cycles applied. The fissile content in the swipe material itself varies and sets a practical base line. A gamma sphere detector could be used to increase the gamma efficiency several times and several cycles could be used.

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REFERENCES

1. International Atomic Energy Agency, *Department of Safeguards Long-Term R&D Plan, 2012–2023*, STR-375, 2013, International Atomic Energy Agency, Vienna, Austria.
2. D. C. Glasgow, I. J. Park, S. M. Hayes, J. S. Shin, J. A. Carter and J. M. Whitaker, "Fast, Sensitive, and Accurate Fissile Determination for Nuclear Nonproliferation Applications by Neutron Activation Analysis and Delayed Neutron Counting," in *Proceedings of the Institute of Nuclear Materials Management Annual Meeting*, Orlando, FL, 2004.
3. S. Croft, R. Venkataraman, R. McElroy, J. Knowles and D. Glasgow, *DN/DG Screening of Environmental Swipe Samples: FY2016 Report*, ORNL/TM-2016/748, Oak Ridge National Laboratory, Oak Ridge, TN, 2016.
4. G. Rudstam, P. Finck, A. Filip, A. D'Angelo and R. D. McKnight, *Delayed Neutron Data for the Major Actinides*, Proceedings of NEA/WPEC-6, 2002.
5. J. R. Knowles, A Generalized Method for Fissile Material Characterization Using Short-Lived Fission Product Gamma Spectroscopy, Knoxville, TN: University of Tennessee PhD Dissertation, 2016.
6. G. P. Westphal, "Loss Free Counting—A concept for real-time compensation for dead-time and pile-up losses in nuclear pulse spectroscopy," *Nuclear Instruments and Methods*, 146 (1977) pp. 605–606.
7. Rooney, B., Garner, S., Felsher, P., Karpius, P., (2015), PeakEasy 4.80 [Computer Program], Los Alamos National Laboratory, Release LA-CC-13-040, https://PeakEasy.lanl.gov