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Abstract
Neutron resonance transmission analysis (NRTA) is an active-interrogation nondestructive assay (NDA) technique capable of assaying spent nuclear fuel to determine plutonium content. Prior experimental work has definitively shown the technique capable of assaying plutonium isotope composition in spent-fuel pins to a precision of approximately 3%, with a spatial resolution of a few millimeters. As a Grand Challenge to investigate NDA options for assaying spent fuel assemblies (SFAs) in the commercial fuel cycle, Idaho National Laboratory has explored the feasibility of using NRTA to assay plutonium in a whole SFA. The goal is to achieve a Pu assay precision of 1%. The NRTA technique uses low-energy neutrons from 0.1-40 eV, at the bottom end of the actinide-resonance range, in a time-of-flight arrangement. Isotopic composition is determined by relating absorption of the incident neutrons to the macroscopic cross-section of the actinides of interest in the material, and then using this information to determine the areal density of the isotopes in the SFA. The neutrons used for NRTA are produced using a pulsed, accelerator-based neutron source. Distinguishable resonances exist for both the plutonium (^{239,240,241,242}Pu) and uranium (^{235,236,238}U) isotopes of interest in spent fuel. Additionally, in this energy range resonances exist for six important fission products (^{99}Tc, ^{103}Rh, ^{131}Xe, ^{133}Cs, ^{145}Nd, and ^{152}Sm) which provide additional information to support spent fuel plutonium assay determinations. Based on extensive modeling of the problem using Monte Carlo-based simulation codes, our preliminary results suggest that by rotating an SFA to acquire two orthogonal views, sufficient neutron transmission can be achieved to assay a SFA. In this approach multiple scan information for the same pins may also be unfolded to potentially allow the determination of plutonium for sub-regions of the assembly. For a 17 $\times$ 17 pressurized water reactor SFA, a simplified preliminary analysis indicates the mass of ^{239}Pu may be determined with a precision on the order of 5%, without the need for operator-supplied fuel information or operational histories.

Keywords: spent nuclear fuel, plutonium, assay, neutron, resonance

Introduction

Neutron resonance transmission analysis (NRTA) is a quantitative analytical technique capable of determining the mass of uranium, plutonium, higher-order actinides, and several fission products in spent fuel. In ad-hoc testing the approach was experimentally demonstrated for commercial light-water reactor spent fuel pins over 30 years ago; the measurement assay precision of the technique for quantifying plutonium was demonstrated in the range of 2 - 4%.[1-3] Recognizing the capabilities of this technique, in 2009 Idaho National Laboratory (INL) initiated a research program to explore the feasibility of using NRTA as a method for assaying a whole spent fuel assembly (SFA).[4-7] The aim of this project was to understand the capabilities and limitations of NRTA for dealing with more massive objects (assemblies versus single fuel pins) and, in particular, to estimate the measurement precision that can be achieved. To make this estimate a system-level approach was used to consider the many different variables that impact mass estimation in NRTA measurements. Constraints were placed on this analysis to consider only

currently-available technology. The ultimate goal was to see how close a conceptual design could come towards achieving the 'grand challenge' goal for assaying Pu in a SFA of having a Pu assay precision of 1% or better. This analysis was entirely done based on simulation and modelling. Recognizing this limitation, effort was taken to model the earlier NRTA experimental work assaying fuel pins, to serve as a benchmark comparison for building confidence in the modelling.

INL's work to examine NRTA as a method for assaying Pu in spent fuel has been part of a larger program of research sponsored by the U.S. Department of Energy's Next Generation Safeguards Initiative (NGSI). The NRTA technique is just one of many technologies studied in the NGSI to investigate measurement techniques for determining Pu mass in commercial SFAs and for detecting the diversion of pins from commercial SFAs.[8,9]

Background Information

Neutron time-of-flight (TOF) measurements represent a well known set of tools for determining neutron-reaction cross sections of nuclei.[10,11] NRTA, in the simplest sense, is a cross-section measurement in reverse where the cross-section is the 'known' and the transmission-target mass is the 'unknown.' It uses low-energy neutrons in the 0.1-40 eV energy range for quantifying plutonium (see Figure 1); this energy range is at the bottom end of the actinide-resonance range, where most actinides have one or more resonances. For most actinides in spent fuel these resonances are typically large in magnitude, narrow in breadth, and well-separated, resulting in distinctive resonance transmission spectra.

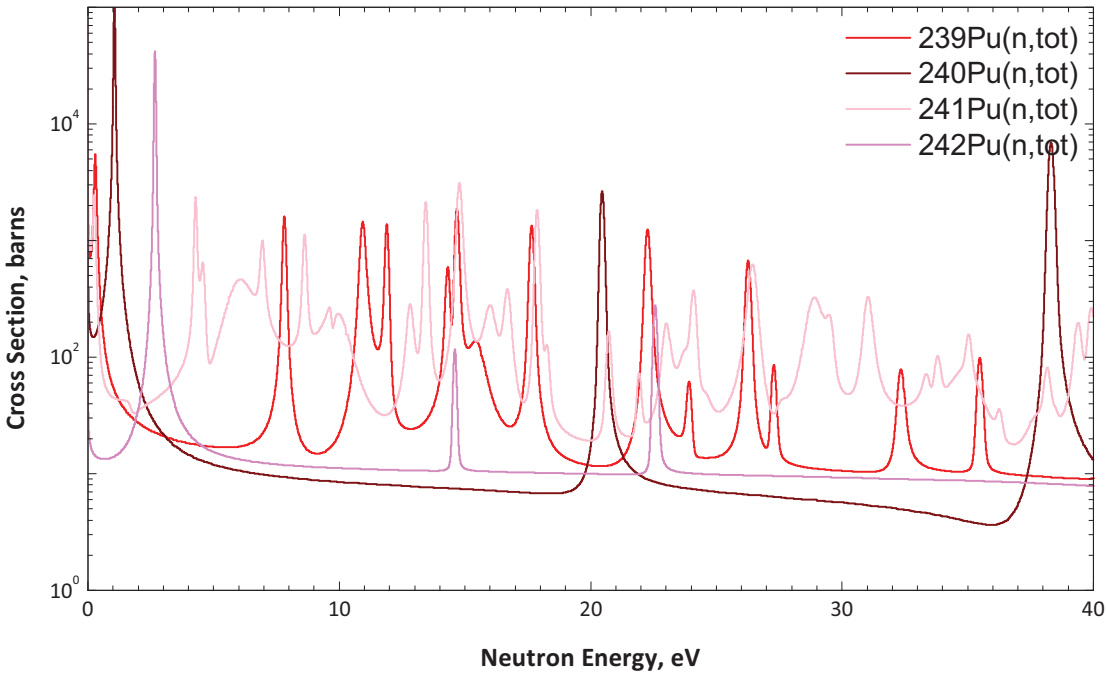

Figure 1 The total neutron interaction cross section for four plutonium isotopes.

NRTA Implementation

The neutrons may be generated using reactor neutron sources with a chopper wheel but, for implementation for safeguards measurements, accelerator-based sources are a more practical approach. Accelerator neutron production may be achieved using either electron or light-ion beams. A simplified approach using a pulsed electron accelerator, for example, would start by having a short duration pulse (lasting for a microsecond or less) of high-energy electrons impact a high-Z converter (e.g., tungsten or tantalum), producing a continuum of bremsstrahlung photons.

It is practical to consider using an accelerator having an endpoint energy in the range of ~10-12 MeV; higher energy systems are also suitable. The high-energy bremsstrahlung photons then pass through a low-Z photo-neutron converter (heavy water or beryllium) to produce high-energy neutrons. These high-energy neutrons pass through a low-Z neutron moderator (polyethylene, light or heavy water, or beryllium) where they scatter and lose energy (a process called thermalization). This scattering process yields a continuous distribution of neutrons, varying in energy from the starting energy down to approximately 0.025 eV, the thermal kinetic energy of nuclei. It is the portion of this continuum having thermal and epithermal neutrons that is of interest to the NRTA technique. The accelerator would be pulsed with a sufficiently long period to allow all of the thermal neutrons from one burst to exit the system prior to the start of the next pulse. Candidate pathways for producing neutrons using light-ions include the ^{7}Li(p,n) or ^{9}Be(p,n) reactions.

Once thermalized, irrespective of the production method, the neutrons need to be collimated to create a neutron beam directed at the SFA. The collimation process must also incorporate a minimum length of 'drift space' to allow the multi-energy neutron beam to self-separate in time and space. High-energy neutrons traverse this drift space most quickly, arriving at the SFA sooner after the end of each beam pulse, while lower-energy neutrons arrive at later times after each burst of neutrons. A fraction of the 0.1-40 eV neutrons incident on the fuel assembly are removed from the beam as they interact with fuels pins in the SFA. The removal processes include elastic scattering, neutron-capture absorption, and neutron-capture fission. The rest of the neutrons pass through the spent fuel assembly unaffected, this is the transmitted signal. It is these transmitted neutrons – the modulated beam – that is measured in the NRTA technique. The transmitted signal is an integral scan through the assembly and is analogous to how a traditional x-ray radiograph is used to detect the presence of dense objects in the human body (bones) and the location of low density regions (bone cracks). The NRTA technique does not assay individual pins in a SFA, with the exception of the four corner pins. Measurements are made using a single-pixel or pixelated detector arrays that are sensitive to thermal neutrons. The time of arrival of the neutrons is recorded as a function of time following each burst of neutrons; the results from each successive burst are added together over the duration of a measurement. Since different energy neutrons travel at different speeds, the time of arrival can be related to the energy of the neutrons when the drift length is known. At neutron energies where there are resonances in isotopes present in the fuel the count intensity is observed to decrease. The magnitude of signal decrease is directly related to the areal density of the absorbing isotopes in the SFA. A schematic representation of the NRTA measurement approach is shown in Figure 2.

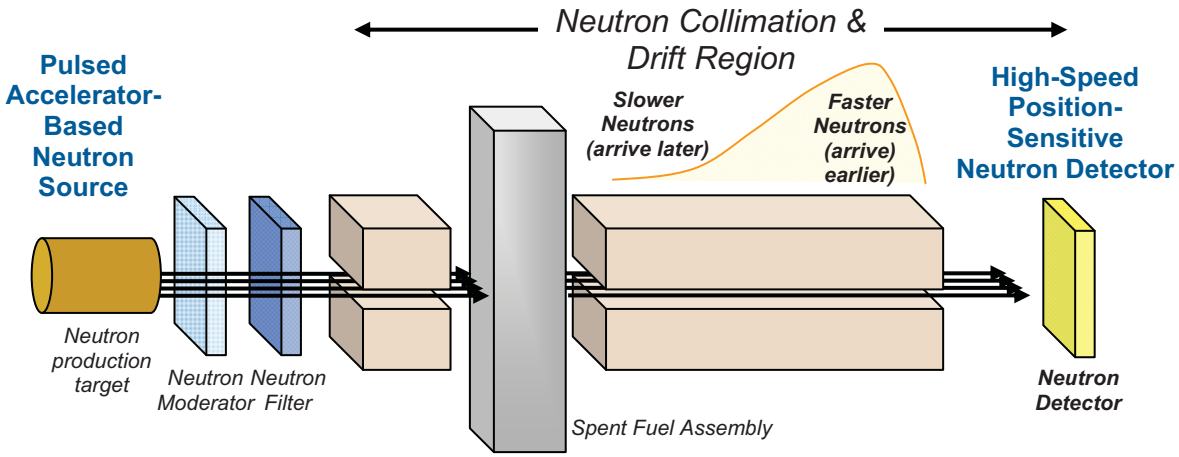

Figure 2 Schematic representation of the NRTA measurement approach.

Time and Energy Resolution – The Drift Length

For TOF measurements such as NRTA, the energy-resolution requirements for a measurement determine the drift tube length and the detector's timing-resolution requirement. Starting with a known detector resolving time, the drift tube length may be determined to ensure that the neutron energy spread (resolution goal) is less than the detector resolving time. Conversely, starting with a known drift tube length, a detector resolving time specification may be defined to achieve the desired energy resolution. A tabulation of neutron energies and velocities is shown in Table 1. If one were to assume a drift tube length of 5 m, it is clear that a detector time resolution of 0.20 μs would be needed to have an energy difference of 0.1 eV between 20 eV and 19.9 eV. At lower energies though the detector timing requirements are reduced; between 1 eV and 0.9 eV the same 0.1-eV energy resolution would only require a detector time resolution of 19.5 μs. A neutron detector time resolution of 0.1 μs is reasonable using a ^{3}He-based detector, for example, but other detector options exist as well. Recognizing this, a drift-length of 5 m has been the basis for most INL conceptual NRTA modeling to date. With a drift length of 5 m the maximum permissible accelerator pulse rate may be determined by choosing a period so that the slowest neutrons of interest, 0.1 eV for NRTA, have sufficient time to reach the detector. For a 5-m drift tube the fastest permissible pulse rate is 876 Hz.

Table 1 Neutron energies and velocities, and transit times through a 5-m drift tube.

Neutron Energy [eV]	Velocity [cm/μs]	Neutron Flight Time over 5 m [μs]
0.1	0.4377	1,142
0.9	1.313	380.8
1	1.384	361.2
9.9	4.355	114.8
10	4.377	114.2
19.9	6.174	80.98
20	6.190	80.78

Competing Resonances in the 0.1 – 40 eV Range

Of the hundreds of fission-product isotopes found in spent fuel, only a half-dozen or so have a significant resonance structure in the 0.1-40 eV energy range (see Figure 3); both ^{235}U and ^{238}U have resonances in this region too (see Figure 4). However, for each of the actinide isotopes there is at least one clear resonance peak unobstructed by narrow-width interfering resonances (when the correct concentrations are accounted for). Other isotopes present in nuclear spent fuel – for example, oxygen in the UO_2 ceramic, and zirconium, tin, iron, chromium, oxygen, niobium, nickel, carbon, and silicon in the Zircaloy-4 clad – also have no resonance structure in the 0.1-40 eV range. The same is true for the hydrogen impurity (hydration) in the clad and air isotopes.

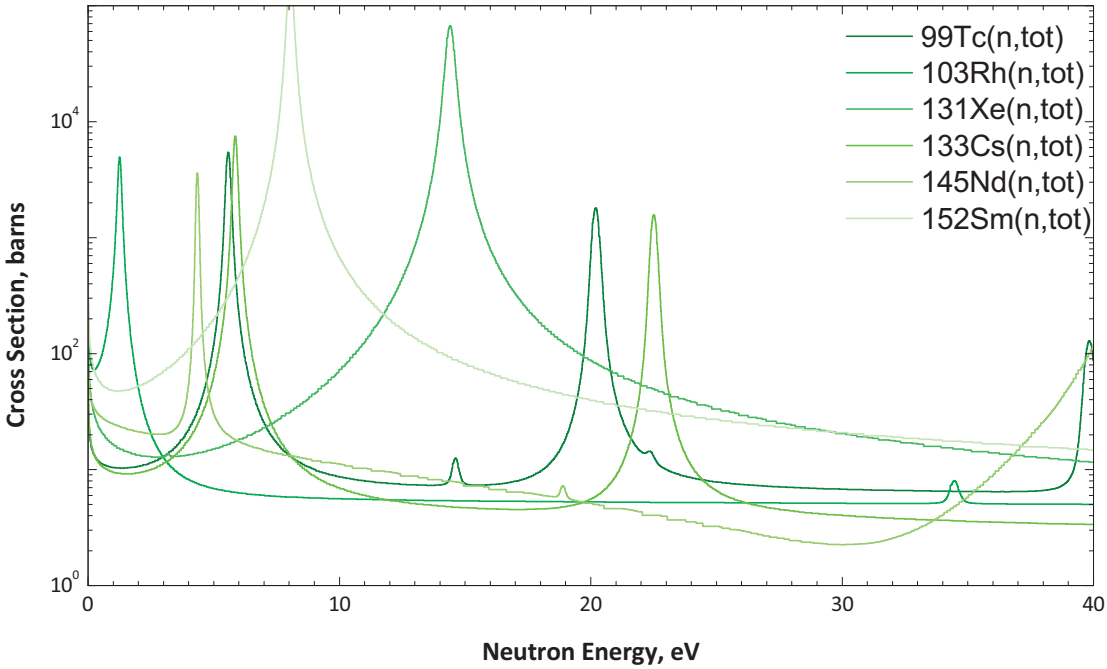

Figure 3 Total neutron interaction cross sections for relevant fission products.

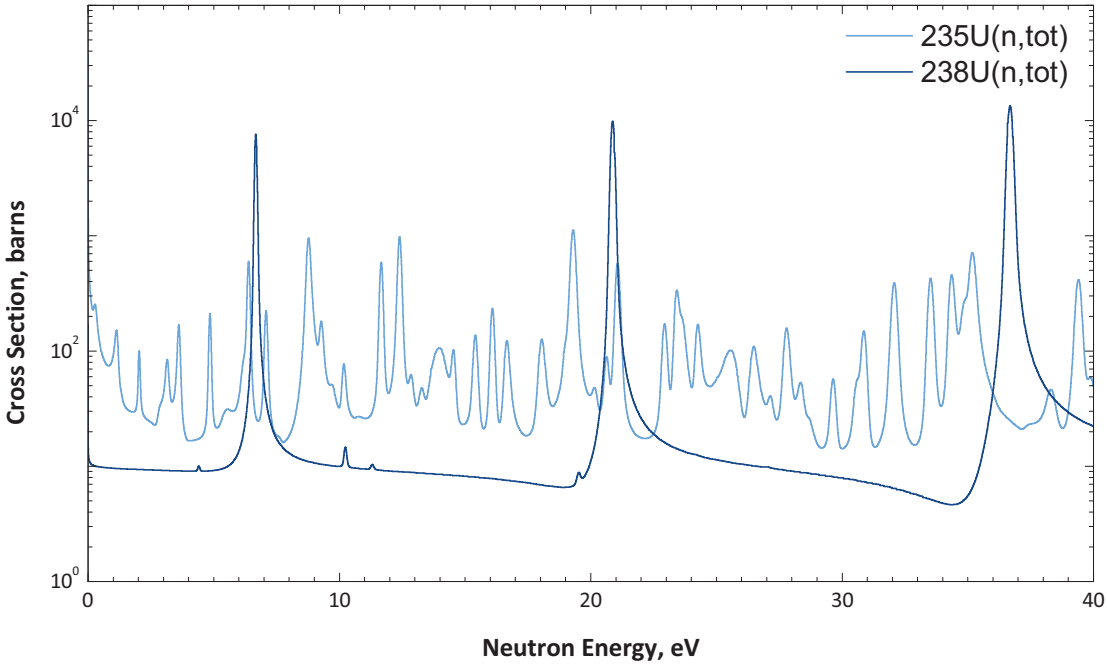

Figure 4 Total neutron interaction cross sections for uranium.

Using Transmission to Determine Pu

Many advances have been made between the early NRTA work done in the 1980s and today. These advances help enable practical modeling and, potentially, the implementation of the NRTA technique. The most significant enabling advances have been improvements in the nuclear data

needed to analyze NRTA data; neutron cross sections and resonance parameters have been re-measured and evaluated and are now known with a much high degree of accuracy.[12,13]

The NRTA technique grew out of cross-section measurement techniques; it is basically the inverse to the problem of measuring neutron cross sections. In the case of neutron cross-section measurements the two known variables are (1) the sample isotopic content, or sample areal density N_i and (2) the transmission flux T_i through isotope *i* determined by the TOF measurement. A third variable, the total cross section σ_{ti} for an isotope *i*, is the unknown variable and the desired quantity to be measured. The total cross section is then calculable as a function of neutron energy, *E*, from Eq. 1.

$$T_i(E) = e^{-N_i \sigma_{ti}(E)} \qquad \text{Eq. 1}$$

For NRTA, the unknown variable is the isotopic areal density N_i, and the two known variables are the total cross section σ_{ti} and the measured neutron transmission flux T_i. Other known factors include the fuel pin and assembly geometry and dimensions, cladding material, and the fuel form (UO_2). Knowledge of these "other" factors helps to reduce systematic error and improve the overall NRTA measurement accuracy.

Calibrated reference standards, if available, can also play an important role in assessing the accuracy and precision of spent fuel assay measurements; the same would be true for an NRTA system. Unirradiated and irradiated fuel pins and assemblies with known dimensions and material compositions can be used as NRTA calibration standards. For example, pins containing (1) no fuel, (2) fresh UO_2 with 100% ^{238}U and various ^{235}U enrichments, (3) and spent fuel surrogate compositions (uranium and rare-earth elements) can all be fabricated with accurately known isotopic compositions.

High-fidelity burnup calculations can also play an important role to further support NRTA spent-fuel transmission measurements. NRTA transmission measurements can be compared to simulated transmission measurements. Agreement between measured and simulated spectra, and then comparison of the corresponding isotopic concentrations, can be used to confirm pin/assembly isotopic concentrations, burnup, and cooling times. This technique is uniquely applicable for NRTA spent fuel Pu assay due to the techniques *proven* capability (over thirty years ago) to perform *absolute* Pu measurements with better than 3% uncertainty for single pins, which would be possible for the four corner pins of a SFA.[1-3]

Understanding NRTA

Several MCNP (Monte Carle n-Particle) transport code models have been developed for NRTA feasibility studies, they were all relatively simple in geometry.[14,15] The simplest geometry models included a directed neutron source, a single fuel pin or line of pins, an evacuated flight tube, and flux-tally detector cells. More sophisticated models included a complete pressurized water reactor (PWR) 17 $\times$ 17 pin SFA, air-filled drift tubes, and a wide-area neutron beam. Some SFA models used for this project were based on models and libraries of irradiated UO_2 fuel developed at Los Alamos National Laboratory as a part of the U.S. Next Generation Safeguards Initiative.[16] The PWR spent-fuel pin geometry included a 0.82 cm UO_2 pellet diameter, 0.95 cm diameter clad (no gap), and a fuel pin pitch of 1.26 cm.

In all of the models the neutron source was uniformly sampled over the desired transmission neutron-energy range. In an actual physical NRTA system a slowing-down neutron source would be expected to exhibit some non-uniformity, in particular, slightly higher flux at higher energies. In the 0-40 eV energy range, however, with the use of a low-Z neutron moderator with low absorption, a relatively flat and uniform energy-flux distribution would be expected. The MCNP models used two different beam geometries: cylindrical beam and directed point-source beam. The cylindrical beam typically had a diameter less than the UO_2 pellet diameter (<0.82 cm). The

cylindrical-beam radius was also varied for some studies. The directed point source is essentially a cylindrical beam with zero radius: a point source directed in a particular direction creating a line of neutrons. The directed point source is useful in order to assess the effect of pellet curvature on the transmission signal. Evaluated Nuclear Data File VII (ENDF-7) was used for most of the numerical simulations.[12] ENDF-5 and ENDF-6 data were used initially and results calculated with these data compare well with the ENDF-7 data.

Benchmark Study

Without the ability to perform a new set of experiments it was important to validate the performance of the MCNP modeling approach. To do this, a model was developed to simulate the first NRTA experiments with spent fuel pins performed in the 1980s.[2] These early experiments used two segments of a single UO_2 fuel pin taken from a commercial SFA. The samples were approximately 1.0 cm in diameter and 2.5 cm in length (probably a PWR fuel pin); one sample was from the vertical center of the SFA, the other sample was from near an end. The fuel had an estimated burnup of approximately 25 GWD/MTU. No initial enrichment or cooling time was given, but prior experience indicates that a PWR assembly with approximately 25 GWD/MTU back in the 1970s timeframe was probably around 3.2 wt% ^{235}U. For the model an external pin from a SFA in the NGSI library with an initial enrichment of ^{235}U 3.0 wt%, a burnup of 30 GWD/MTU, and a 5-year cooling time was used.

NRTA-measured transmission spectra measured for the two samples are shown in Figure 5. The spectrum at the bottom of the figure is for the sample cut from the center of the fuel pin, and spectrum B, the upper spectrum, is for the sample taken from the end of the fuel pin. Note the ordinate axis discontinuity in this figure. The resonance depressions correspond to specific actinide and fission product isotopes and have been marked in the figure. It is interesting to note the deeper depression in spectrum A for the ^{242}Pu resonance at 2.65 eV. This is due presumably to the higher burnup in the middle of the spent fuel pin, due to the typical PWR-reactor cosine-like vertical power distribution, and thus correspondingly higher ^{242}Pu concentration. Also, the ^{235}U depression at 8.8 eV is deeper for spectrum B relative to A, due to the higher ^{235}U concentration remaining in the fuel at the ends of the pin relative to the center (again, due to the lower burnup at the ends of the fuel pin). A key observation from this simple set of measurements is the natural axial variation in burnup in a SFA having some average 'declared' burnup. For Pu analysis of a whole SFA this must be accounted for if a measurement precision on the order of 1% is desired.

These measured transmission spectra are compared with the simulated NRTA response shown in Figure 6. This simulated spectrum was calculated with the MCNP5 code and models described above. In the NGSI spent fuel library the spent fuel composition is assumed to be axially uniform, hence the calculated spectrum represents an average over the length of the fuel pin. One might expect the magnitude of the resonance depressions to lie between the two measured spectra. Comparing Figure 5 and Figure 6, it is readily visible that the simulated transmission spectrum bears the same shape and includes all of the important actinide transmission depressions as the measured spectra. The measured depressions are proportional in depth to the simulated spectrum although the ordinate axis in Figure 6 is plotted over a larger range,

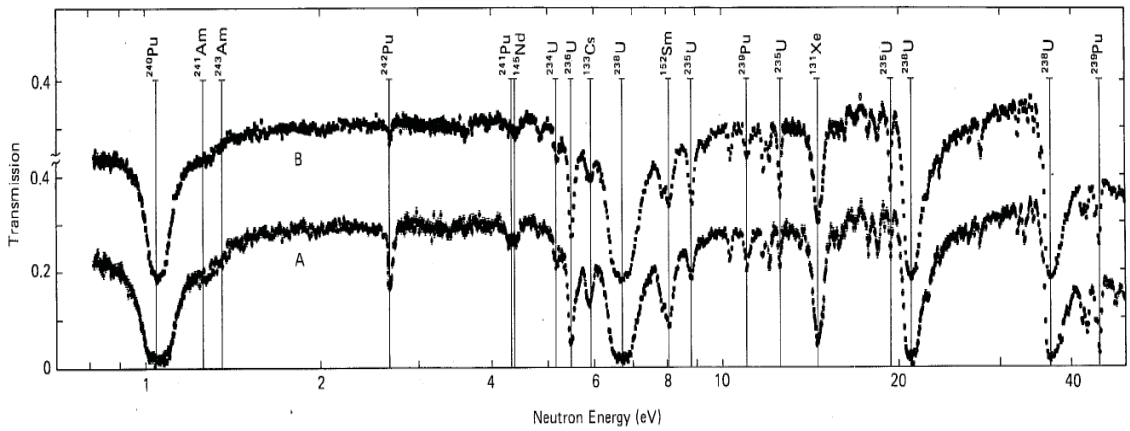

Figure 5 Measured resonance transmission spectra as a function of neutron energy for the NBS UO_2 spent fuel samples (Spectrum A is for fuel cut from the center of the fuel pin and spectrum B is cut from one end of the fuel pin).

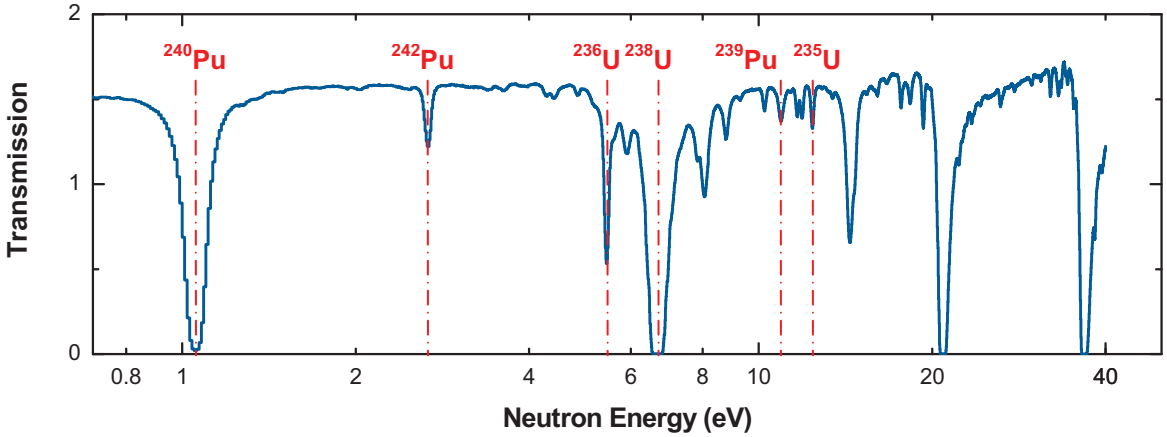

Figure 6 MCNP-calculated relative transmission spectrum as a function of neutron energy through a single UO_2 spent fuel pin; important isotopes have been highlighted in red.

Accessibility of High Yield Neutron Sources

An intense neutron source would be required to achieve reasonable count times for NRTA SFA measurements. Estimates on the order of 10^{12} n s^{-1} (4π emission) are projected due to the basic physics of the NRTA technique.[4,7] Multiple accelerator options exist to meet this need. In addition to the electron-accelerator based sources which this project has considered, light-ion accelerators may also be used. Specific examples of fielded accelerator systems that meet this neutron-intensity requirement need include the following.[17]

1. Compact pulsed hadron source, Tsinghua University, China
   E_p = 13.0 MeV, I_{ave} = 1.25 mA, rate = 50 Hz
2. Low energy neutron source, Indiana University, U.S.
   E_p = 13.0 MeV, Y ≈ 4.2×10^{13} n s^{-1}
3. KUURI-FFAG, Kyoto University, Japan
   E_p = 11.0 MeV, Y ≈ 5.0×10^{13} n s^{-1}, rate = 200 Hz, pulse width = 200 μs
4. KUURI-eLINAC, Kyoto University, Japan
   E_e = 30.0 MeV, Y ≈ 8.0×10^{12} n s^{-1}, I_{ave} = 200 μA, rate = 300 Hz

Count Time Estimates

The baseline neutron source used for modeling in this project has been a 10-MeV electron accelerator producing photoneutrons using a tungsten/beryllium converter/moderator; its neutron

source yield is 2.7×10^{11} n s^{-1} (4π emission). With this model, counting times have been estimated for a single PWR pin transmission as well as transmission through a diagonal cross section through the SFA of 8 pins.

The following assumptions are used in the count time estimates:

1. Linear electron accelerator with maximum electron energy of 10 MeV,
2. Optimal tungsten converter thickness of 1.45 cm for 10-MeV operation,
3. Neutron energy range of interest: 1.0 - 40 eV,
4. Tungsten-beryllium (W/Be) converters,
5. 5-meter flight tube distance to detectors,
6. 100-μA average beam current,
7. Detector active area of π cm^2 (1 cm radius) perpendicular to beamline axis, and
8. Neutron detector efficiency ε = 20%.

For assaying through a section of a single pin with this *baseline* neutron source the counting time needed to resolve one resonance absorption line with counting statistics of better than one percent, including accounting for background events due to spontaneous neutrons emitted from the SFA, has been calculated to be approximately 1.8 hours.[7] For an 8-pin row assay the count time goes up to approximately 13.4 hours. For a single detector this may be too long for a safeguards-relevant measurement. However, if multiple detectors are used to allow complete assay of multiple sections of an assembly simultaneously, this may be acceptable. If faster counting times are desired, e.g., to reduce the 8-pin count time from 13.4 hours to approximately 20 minutes, the neutron source strength would need to be increased by a factor of 40; as illustrated above higher yield systems are well-proven. There are several ways to get this factor of 40 increase in the neutron intensity, considering modifications to the base-line model. These include a) increasing the accelerator electron energy, b) increasing the average beam current, c) changing out the tungsten converter for a depleted uranium converter, and c) optimizing the converter materials and dimensions.

Assaying Pu in a SFA

The simplest approach for estimating the Pu content in an NRTA measurement is to determine the neutron removal fraction of each Pu isotope at a single resonance energy. For each resonance a straight line is drawn over the top of the depression from the high points on each side of the depression and then the attenuation from the point on the line corresponding to the resonant energy to the bottom of the depression is measured and recorded as the “a” value, in counts. The other important variable to be measured is the distance from the bottom of the depression to zero on the ordinate axis (number of detector counts). The transmission factor (T) is then estimated to be the ratio of “b” divided by “a+b.” The mass can then be derived from the transmission factor, T.

To demonstrate how the transmission approach above can be used to estimate the total ^{239}Pu mass in a SFA, the single ^{239}Pu resonance at 10.93 eV will be used; no interfering resonances (e.g. ^{235}U) have been accounted for. While this particular resonance is relatively well isolated other resonance interferences still affect the analysis. The simulations for this example involved a PWR spent fuel assembly with a burnup of 45 GWD/MTU, initial enrichment of 3 wt% ^{235}U, and a 1-year cooling time. The fuel composition is taken directly from LANL spent fuel library #1, where each of the 264 fuel pins in the assembly has been divided into four radial fuel-pin depletion regions, with a single axial value over the full length of each fuel pin. The depleted fuel assembly has 39 different depleted fuel pins and a total of 156 different fuel compositions. In LANL library #1 the SFAs have depleted fuel pin symmetry about the horizontal and vertical axes through the center of the assembly, as well as symmetry about the two diagonal axes through the corners and center of the assembly.

The total ^{239}Pu mass in the library SFA (45 GWD/MTU) is 2,610.45 grams. In an ideal situation, an estimation of the ^{239}Pu mass using this simple approach should equal this value. However, in

practice curve-fitting the transmission depression signals, and accounting for additional resonances, would be needed to best measure the transmission factor. For the example here, however, only the single time-bin value at the center of the resonance (and only the ^{239}Pu resonance cross section) is used to calculate the transmission factor. This is illustrated in Figure 7, which shows the resonance absorption method being used to determine the transmission factor T. The transmission spectrum in the figure is the 8-14 eV portion of the energy range which includes the ^{239}Pu resonance depression of interest at 10.93 eV; the red-hatched area is this transmission depression. The values “a” and “b” are shown and estimated by simply drawing a straight line (red) across the top of the depression and measuring the “a” and “b” distances. The transmission factor T is then given by the formula: T = b/(a+b).

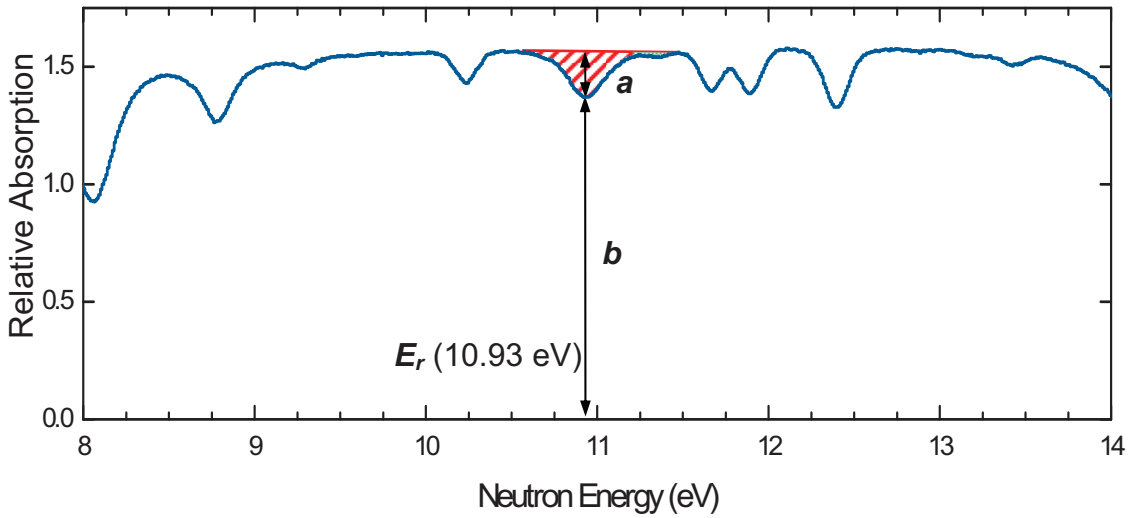

Figure 7 Estimation of the transmission factor (T) using the 10.93 eV resonance depression from ^{239}Pu. The transmission factor T is simply given by T = b/(a+b).

The transmission factor (T) is related to the average ^{239}Pu number density (N) in the transmission path by the following attenuation formula

$$T(E_r) = \frac{b}{a+b} = e^{-N\,\sigma_t(E_r)\,x} \qquad \text{Eq. 2}$$

where $T(E_r)$ is the transmission factor at the resonant energy, σ_t is the magnitude of the ^{239}Pu total neutron interaction cross section at the resonant energy (10.93 eV), and x the average chord length of the transmission path through the fuel pin(s) in an assay row. With this approach, the ^{239}Pu mass was estimated for 15 of 16 paths (d1-d15) through the SFA. These paths correspond to the labeled diagonal lines in Figure 8. The sixteenth scan (d16) was not calculated. The results of the calculations are shown in Table 2, which gives the calculated mass estimate (Calc. Mass) in grams along with the 'true' mass from the LANL library fuel compositions (Lib. Mass) for scan numbers d1 through d15. The last column is the percent difference between the calculated mass and the LANL library mass. The percent difference is always negative, meaning that the calculated mass using this approach always slightly under predicts the library values. The under prediction is believed to be due to the presence of the overlapping resonances of the other material in the fuel. It is also possible that small fission product or actinide resonances may be present inside the ^{239}Pu 10.93-eV resonance but a search for these has not yet been performed.

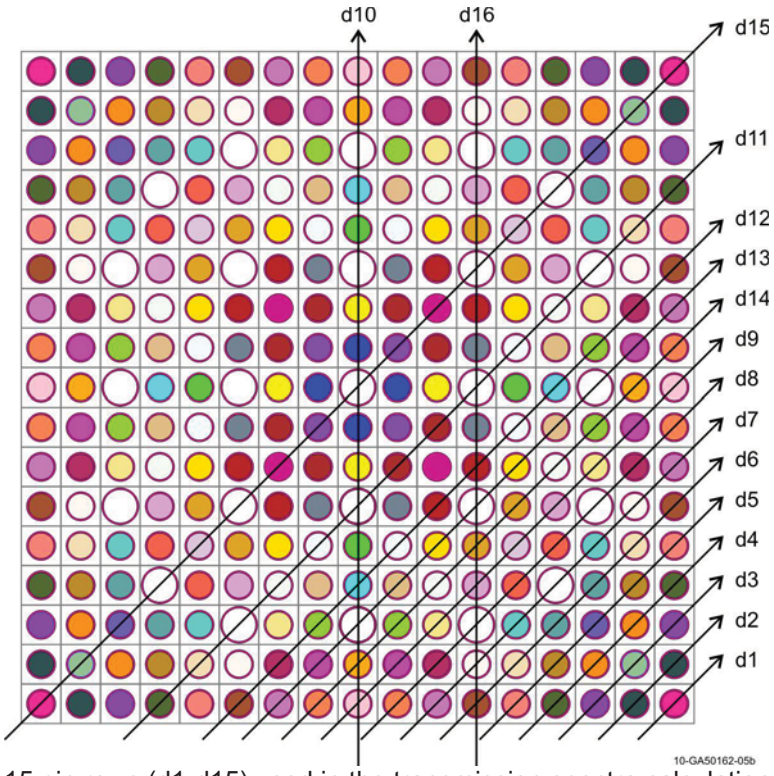

Figure 8 The 15 pin rows (d1-d15) used in the transmission spectra calculations.

Table 2 Results of the ^{239}Pu estimate by scan row.

Scan No.	No. of Pins	Lib. Mass [g]	Calc. Mass [g]	Diff. [%]
d1	1	10.86	10.49	-3.4
d2	2	21.62	20.82	-3.7
d3	3	31.96	30.51	-4.5
d4	4	41.92	41.04	-2.1
d5	5	51.27	48.61	-5.2
d6	6	60.20	57.40	-4.7
d7	6	59.52	57.41	-3.5
d8	6	59.48	57.29	-3.7
d9	9	87.82	83.02	-5.5
d10	12	115.49	103.55	-10.3
d11	10	98.37	89.04	-9.5
d12	12	116.95	105.75	-9.6
d13	8	78.45	73.75	-6.0
d14	10	97.51	89.42	-8.3
d15	12	120.46	110.51	-8.3
d16	12	---	---	---

These values may be used to develop estimates of the total ^{239}Pu mass in the assembly. First, limiting the discussion to a set of scans only reaching a total thickness of 8-pins in a row (the d1-d8 and d13 scans, which may be the practical limit for attenuation assays), then 156 of 264 (59%) of all the fuel pins are assayed directly. For an estimate one uses the directly measured fuel masses from scans d1-d8 and d13 and then, for the remaining internal pins which are not included, an average ^{239}Pu mass was derived from the average value of the d13 scan. The final

calculated SFA ^{239}Pu mass is then 2,511 grams, which should be compared with the MCNP library mass of 2,610.45 g. The -3.80% difference is directly attributable to the simplified peak fitting approach which omits known resonance interferences from uranium and fission products. This is a systematic error, it could likely be improved upon by using more sophisticated assay analysis techniques beyond the 'peak height' analysis used here. If a counting statistics error of 2.5% is acceptable then a measurement would take 13.4 × 2 = 26.8 hours using a 2.7×10^{11} n s^{-1} neutron source or 20 × 2 = 40 minutes using a 1.1×10^{13} n s^{-1}. The total uncertainty would be approximately 4.3%.

If 12 pins per row can be assayed (e.g., if more time is allocated for measurements) then all 15 scans (d1-d15) can be used in the total ^{239}Pu assembly estimate. The total mass of ^{239}Pu in the assembly is then estimated to be 2,488 grams. Compared to the 'true' total ^{239}Pu assembly mass, this is now a -4.68% difference. The difference has grown slightly due to resonance interferences but it is also impacted by more overall signal degradation due to the notably larger UO_2 distance traversed in the scanning of 9 to 12 pins in a row. Note the percent difference in Table 2 tends to increase with the number of pins in a row. The lower total inventory estimate of 2,488 grams is also due to the generally lower average ^{239}Pu pin content of the interior pins relative to peripheral pins, as shown in Table 3 (which indicates a more sophisticated averaging approach must be used to estimate Pu in the inner region of the SFA).

Table 3 Estimated average ^{239}Pu pin content by transmission scan.

Scan No.	Average ^{239}Pu mass [g]
d1	10.5
d2	10.4
d3	10.2
d4	10.2
d5	9.72
d6	9.57
d7	9.57
d8	9.55
d9	9.22
d10	8.63
d11	8.90
d12	8.81
d13	9.22
d14	8.94
d15	9.21

An NRTA measurement with 12-pin scan capability would allow every pin in the assembly to be part of one or more scans. Because of this it may prove possible that the measurement precision can be improved on by developing an approach to optimize the assignment of average ^{239}Pu pin mass determinations using data from the 16 integral transmission scans (rather than the single average value assigned to all pins as used in this discussion). Starting with the corner pins and moving inward, for example, it might be possible to determine the ^{239}Pu mass content of each pin in the assembly to a higher degree of accuracy using an iterative approach. Ultimately some approximations may be required for the interior pins but, as recently described elsewhere, the interior pins have a fairly uniform and predictable ^{239}Pu distribution that may support this extrapolation.[18]

Pin Diversion

Although a large neutron source strength and perhaps long count times would be needed to perform a quantitative NRTA 12-pin scan, lower system requirements would be needed to quickly detect diversion of a pin in the assembly. This would be true even if the neutron intensity only

allowed sufficient transmission to support quantitative measurements for scans through a depth of 8 pins. This analysis would involve examination of the uranium isotopes as well as the fission products ^{99}Tc, ^{103}Rh, ^{131}Xe, ^{133}Cs, ^{145}Nd, and ^{152}Sm in the assembly. Use of ^{131}Xe is a particularly valuable indicator in this case.

Discussion

As a measurement technique for performing high-precision plutonium assays the NRTA technique has a number of strengths in comparison with other nondestructive assay approaches.

1. NRTA has the potential for accurate assay measurements with a precision in the range of 5% uncertainty or better.
2. NRTA produces distinctive resonance-transmission spectra that can uniquely identify specific actinide and fission product isotopes. The method detects and measures plutonium isotopes directly; it does not rely on correlations or the "effective ^{240}Pu" concept.
3. In addition to ^{239}Pu, NRTA can identify and assay several important fissionable isotopes and spent-fuel actinides directly, including ^{235}U, ^{236}U, ^{238}U, ^{240}Pu, ^{241}Pu, and ^{242}Pu.
4. NRTA can identify the presence of ^{234}U, ^{241}Am, and ^{243}Am. Americium-241 is of particular relevance in higher burn-up fuels (45-60 GWD/MTU) with large cooling times (>5 years).
5. NRTA can identify 6 resonant fission-product isotopes (^{99}Tc, ^{103}Rh, ^{131}Xe, ^{133}Cs, ^{145}Nd, and ^{152}Sm) which can potentially be used to estimate assembly burnup, cooling time, and diversion and to verify operator-reported burn-up values.
6. The neutron resonance transmission analysis technique is a mature technology with a solid foundation in theoretical physics.
7. NRTA system calibrations with pin/assembly standards can be straightforwardly used to reduce NRTA systematic errors.
8. An NRTA system can be designed to be insensitive to spent-fuel gamma radiation.

The NRTA technique may require temperature control equipment for the spent fuel assembly. For NRTA, the fuel assembly will necessarily be assayed in air (or vacuum) but not in water. An assembly suspended in vacuum will tend to heat up due to lack of conduction and convection heat-transfer pathways. An assembly suspended in air, however, can be air-cooled through forced convection. In the latter case of forced-air cooling a temperature measurement may not be needed but in vacuum the assembly may heat up, particularly true for assemblies with short cooling times. Heat up of the UO_2 fuel that would be expected in an evacuated system will Doppler-broaden the resonances; this must be compensated for in the cross-section data that are part of the analytical computer tools.

Low-energy neutron penetrability, or the maximum number of PWR pins that can be assayed in a given row, is limited by the physics of the problem. Accelerator systems used in neutron cross-section measurements are large, high-power systems that typically use 100-MeV or higher electron energies, a 700-800 Hz repetition rate, and flight-tube lengths up to 200-meters or longer. They operate at powers ranging from 1.5-14.0 kW.[19,20] An optimized NRTA system may operate in the same repetition rate and power range, but at much lower electron energies. The relatively low-energy electrons required when using an electron-accelerator-based neutron source may be of particular value to reduce the size of the accelerator. To optimize performance the NRTA technique will require an intense source of pulsed low-energy neutrons in the range of 10^{12}-10^{13} n/s; commercial electron accelerators are available to meet this requirement, generally operating with a relatively low electron endpoint energy in the 10-20 MeV. Charged-particle accelerators may also be suitable for use in an NRTA system, instead of using an electron accelerator. Commercial accelerators using deuteron and proton beams that can generate neutron yields up to 10^{13} neutrons per second are currently available.[21]

Determination of the flight tube length is based primarily on the energy range of interest for the transmission neutrons. For the NRTA technique and plutonium assay, neutrons of energy 0.1-40 eV should be sufficient. This relatively narrow energy range includes one or more resonances for ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, and ^{243}Am isotope, plus the six resonant

fission products ^{145}Nd, ^{133}Cs, ^{99}Tc, ^{152}Sm, ^{131}Xe, and ^{103}Rh. Energies from 20-40 eV provide redundant resonances for some of these actinides and therfore it may be possible to restrict the analysis to a maximum energy of 20 eV. (Higher energies also require longer flight tubes for good energy resolution.) The NRTA system as proposed here is for a 0.1- to 20-eV energy range would minimize the flight-tube length to approximately 5 m and reduce the overall NRTA system footprint from previous work.

Multiplexing

Use of a high-power, accelerator-based neutron source may be perceived as a significant drawback of the NRTA technique. However, it may be possible to take advantage of the isotropic nature of this type of neutron source to permit the simultaneous analysis of multiple fuel assemblies. The first concept for analyzing an assembly would be to scan one section of the fuel, and then rotate it to get the orthogonal view. This would allow an assembly to be assayed at one axial location, perhaps 5 to 10 cm in width. However, if it was required to assay the complete assembly top-to-bottom this approach would be time consuming. An alternate approach would be to simultaneously collect data at multiple elevations on the assembly at the same time. Secondly, the isotropic neutron sources proposed for NRTA would permit measuring multiple assemblies in parallel. Based on size limitations and estimates of beam-tube length, it may be possible to analyze up to one dozen assemblies in parallel using one accelerator if desired. It is worth noting that any system designed to comprehensively assay SFAs will, by necessity, be technically complicated. It is difficult to handle spent fuel. As a measurement technique for spent fuel NRTA is most relevant for large scale, national facilities expected to process large numbers of SFA in an assembly-line fashion. One application where NRTA might be used, for example, would be to verify shipper inventory declarations at a large-scale fuel reprocessing facility. Another application would be as a receipt inspection tool at a longer-term storage facility or a geological repository.

Other Applications Beyond Safeguards SFA Analysis

In addition to using NRTA for safeguards measurements, the approach might also be of relevance as a tool for basic scientific research related to the development and understanding of how nuclear fuel behaves. NRTA-type measurements of small samples or single pins have the potential to generate 2-dimensional isotopic composition images of the actinide and fission production distributions in fuels with a spatial resolution of 1-mm or better. Taking multiple images, tomography methods could further allow the reconstruction of 3-dimensional distribution maps of these isotopes in fuel. This information would be valuable for understanding the migration and diffusion of elements in nuclear fuel, and for understanding rim-effects in the radial burnup distribution of commercial light-water reactor fuels.

The NRTA measurement approach may also be applicable as a non-contact, non-destructive assay measurement approach for analyzing and categorizing debris materials that will someday be collected from the remains of the Fukushima-Daiichi complex in Japan. These materials will be non-uniform in size and will be intimately comingled with structural materials from the reactors. Traditional NDA methods which might be applied to these analysis will be limited in applicability due to high radiation fields and the inability to readily make geometric and form-factor corrections. The absolute Pu-assay approach of the NRTA technique, as originally illustrated 30 years ago, is ideally suited for addressing these challenges.

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21 See, for example, the Lansar Neutron Generator from Accsys Technologies, Inc. (www.accsys.com/products/lansar.html).