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A SECOND LOOK AT NEUTRON RESONANCE TRANSMISSION ANALYSIS AS A SPENT FUEL NDA TECHNIQUE

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ABSTRACT

Neutron Resonance Transmission Analysis (NRTA) is measurement technique capable of quantifying plutonium in spent fuel. Having first been explored in the mid-1970s for the analysis of individual spentfuel pins, a second look is now underway to investigate the suitability of the NRTA technique for assaying complete spent nuclear fuel assemblies using advanced simulation and modeling methods. The technique is similar to neutron time-of-flight methods used for neutron cross-section measurements but operates over only the narrow 0.1-20 eV range where strong, 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 exists for six important fission products (⁹⁹Tc, ¹⁰³Rh, ¹³¹Xe, ¹³³Cs, ¹⁴⁵Nd, and ¹⁵²Sm) which provide additional information to support spent fuel plutonium assay determinations. Initial modeling shows excellent agreement with previously published experimental data for measurements of individual spent-fuel pins where plutonium assays were demonstrated to have a precision of 2-4%. Within the simulation and modeling analyses of this project scoping studies have explored fourteen different aspects of the technique including the neutron source, drift tube configurations, and gross neutron transmission as well as the impacts of fuel burn up, cooling time, and fission-product interferences. These results show that NRTA may be a very capable experimental technique for spentfuel assay measurements. The results suggest sufficient transmission strength and signal differentiability is possible for assays through up to 8 pins. For an 8-pin assay (looking at an assembly diagonally), 64% of the pins in a typical 17×17 array of a pressurized water reactor fuel assembly can be part of a complete transmission assay measurement with high precision. Analysis of rows with up to 12 pins may also be feasible but with diminished precision. Preliminary analysis of an NRTA simulation has demonstrated the simplicity of the technique. This work is part of a larger effort sponsored by the Next Generation Safeguards Initiative to develop an integrated instrument, comprised of individual nondestructive assay techniques with complementary features, that is fully capable of determining Pu mass in spent fuel assemblies.

Keywords: spent fuel, neutron time of flight, plutonium assay, nuclear safeguards, non-destructive assay

INTRODUCTION

In 2009, the Next Generation Safeguards Initiative (NGSI) of the U.S. Department of Energy's National Nuclear Security Administration (NNSA) began a five-year effort to develop an integrated instrument fully capable of determining Pu mass in, and detecting diversion of pins from, commercial spent nuclear fuel (SNF) assemblies.[1,2] Following a rigorous modeling evaluation of 14 individual measurement techniques against a library of 64 spent fuel assemblies, efforts in 2011 are now turning to the construction of one or more integrated systems comprised of the most promising techniques.[3] This NGSI effort brings together measurement and modeling experts from multiple U.S. national laboratories, several universities, and international partners.

BACKGROUND

One focus of the NGSI research program is to examine assay methods suitable as portable techniques, where a set of equipment can be shipped to a facility to perform on-site inspections of SNF assemblies in a storage pool. An application for this category of instrument would be to recover from a loss of continuity of knowledge at a nuclear power plant subject to international safeguards monitoring by the

International Atomic Energy Agency (IAEA). Due to the limitations of size and weight for this class of instruments the precision for plutonium assay measurements is not likely to be better than 5-10%.

A second focus of the research program is to study larger-scale measurement approaches where the goal is to be able to perform analyses with precision approaching 1%. This level of measurement precision is hard to reach and represents a grand challenge in the field of safeguards nondestructive analysis. These high-precision systems will likely be too large, costly, and complicated for routine field use at site locations. An application for these systems will be at spent fuel reprocessing facilities, where they may be used to verify shipper-declared plutonium inventories prior to disassembly. These systems may be of particular value for future facilities using pyroprocessing, where there aren't expected to be input accountability tanks suitable for safeguards destructive assay measurements. A second application for these systems may be at long-term and permanent SNF disposal facilities where regulations may require a final accounting to a) remove items from a safeguards balance sheet or b) quantify the total plutonium and transuranic inventory being interred in order to meet repository legal regulations.[4]



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

One measurement concept that may be capable of meeting the high-precision assay goals of the NGSI spent fuel assay program is neutron resonance transmission analysis (NRTA). NRTA is a time-of-flight (TOF) based measurement that exploits the unique neutron-interaction resonance structure of plutonium isotopes in the 0.1 – 20 eV energy range (Figure 1) to quantify the mass of these isotopes in an inspection assembly. Neutron TOF measurements are well understood in the field of nuclear data analysis and have been used for decades as a reliable and accurate method for characterizing the neutron-interaction resonance structure of isotopes. While not as well-known, TOF methods as employed in NRTA have also been studied and characterized to a high-level of understanding. For SNF analysis the first NRTA measurements began in the mid 1970's and were then followed by a considerable research effort for about a decade.[5] More recently, as a part of the NGSI program, Idaho National Laboratory (INL) has revisited the NRTA concept to investigate its suitability for assaying complete SNF assemblies for total-elemental Pu determination.[6] This paper describes our work in this area providing a second look at NRTA for nuclear safeguards.

NRTA CONCEPTUAL OVERVIEW

The conceptual instrumentation layout, as originally proposed by Shrack et al. and shown in Figure 2, includes the following key components: 1) pulsed accelerator-based neutron source; 2) neutron moderator; 3) neutron filter; 4) neutron collimator and drift tube, and 5) neutron detector.[7-9] The

accelerator may be either a light-ion accelerator producing neutrons from a reaction such as the ⁹Be(p,n) reaction or an electron accelerator that produces high-energy x rays which then produce neutrons through photonuclear reactions in materials such as deuterium or beryllium. The neutron moderator and filter may be separate or integrated into a single assembly. Moderation is needed to downscatter fast neutrons from the accelerator system to the 0.1 - 20 eV energy range. Filtering is needed to eliminate very low energy neutrons below the 0.1 eV threshold from entering the system; the filter may be placed next to the detector as well. Neutron collimation is needed to limit off-axis neutron scattering, and to limit the amount of the SNF assembly exposed to the detector. The quantity of SNF exposed to the detector must be restricted in order to limit the background neutron count rate in the detector from SNF-originating neutrons (i.e., (α ,n) neutrons and spontaneous fission neutrons from isotopes of curium and plutonium in the spent fuel.)

Accelerator-generated neutrons travel in all directions; only a small fraction of the neutrons travel the entire length of the collimator to reach the target. Faster neutrons arrive at the detector very shortly after each pulse while slow neutrons, created as fast neutrons that scatter and lose energy in the moderator, arrive at later times. The collimation also serves as a drift region to allow sufficient spread in neutron energy to occur for the detector system (with an inherent resolving time) to resolve the ~0.5- to 1- eV wide resonances of the plutonium isotopes in the 0.1 - 20 eV energy range. The neutron detector must have sufficient timing resolution to resolve the neutron energy to about 0.1 eV or better over the NRTA 0.1 - 20 eV energy range.



Figure 2 Schematic representation of the NRTA measurement approach.

Table 1	Neutron	energies and	velocities,	and transit	times t	through a t	5-m drift tube.
		0	,			0	

Neutron Energy [eV]	Velocity [cm/ms]	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

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 μ s 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. 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.

PRIOR WORK AND BENCHMARK MODELING

INL's NRTA review program is using numerical simulations with the MCNP (Monte Carlo N-Particle) transport code as the basis for initial evaluations of the technique for SNF assembly assays.[10,11] In order to develop a level of confidence in the MCNP codes, models, and cross section data, it was prudent to first compare a calculated transmission spectrum against an actual measured transmission spectrum. Fortunately, two measured spectra were available in the published literature for ad-hoc experiments using a single spent fuel pin.[8] In this reference, from the U.S. National Bureau of Standards (NBS), there are two NRTA-measured spectra based on SNF samples cut from two regions of a single spent fuel pin, one from the center of the pin and the other from the end of the pin. Both cylindrical samples were approximately 1.0 cm in diameter and 2.5 cm in length (probably a PWR fuel pin). The fuel pin had an estimated burnup of approximately 25 GWD/MTU but no initial enrichment or cooling time was given. For the MCNP modeling, the fuel was assumed to be from a PWR assembly with 30 GWD/MTU with an initial enrichment of 3.2 wt% ²³⁵U (a reasonable estimate for SNF in the 1970s timeframe). The burnup characteristics between the actual measured spent-fuel composition and the assumed composition are not exact but are likely reasonably close (as illustrated below).

The two measured transmission spectra from the reference paper are shown in Figure 3. Spectrum A, the lower spectrum, 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 are clearly marked in the figure. It is interesting to note the deeper depression in spectrum A for the ²⁴²Pu resonance at 2.65 eV. This is due presumably to the higher burnup at the pin center and the correspondingly higher ²⁴²Pu concentration. Also, the ²³⁵U depression at 8.8 eV is deeper for spectrum B relative to A due to the higher ²³⁵U concentration remaining at the end of the pin relative to the center.

The corresponding MCNP-calculated transmission spectrum is shown in Figure 4. The spent fuel composition used in the model is from a compilation of spent fuel libraries developed at Los Alamos National Laboratory as a part of the NGSI spent fuel assay project.[3] It represents an average fuel-pin burn-up at 30 GWD/MTU, 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. For comparison purposes, the MCNP-calculated spectrum was arbitrarily normalized to the potential scattering levels of spectrum A and B which appear to be approximately equal.

In addition to the difference in the burnup between the samples and the assumed material composition in the calculations, two other differences may exist: (1) the measured transmission neutrons were from a slowing-down neutron energy spectrum (12.5-cm slab of polyethylene) versus MCNP neutrons sampled uniformly over the 0-40 eV energy range and (2) possible differences in the neutron

beam geometry (the calculation assumed a cylindrical beam of diameter slightly less than 1.0 cm, whereas the measurement beam geometry is unknown).

It is important to note that the calculated spectrum over the approximately 0.5-40 eV energy range looks very much like the measured spectra. The same isotopic depressions are observed with a similar depression magnitude, which means that the calculated spent-fuel compositions contain the right resonant isotopes and the concentrations are reasonable. The good agreement between the measured and calculated transmission spectra for the spent fuels lends some level of confidence and credibility to the MCNP-calculated transmission spectrum, MCNP code models, LANL spent-fuel compositions, ENDF-7 neutron cross section data and, finally, the INL feasibility studies in the 2010 report.[6]



Figure 3 Measured resonance transmission spectra as a function of neutron energy for the NBS spent nuclear 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 4 MCNP-calculated transmission spectrum as a function of neutron energy through a single spent fuel pin; important isotopes have been highlighted in red.

CONCEPTUAL STUDIES

Fourteen conceptual modeling studies (Table 2) have been performed to investigate different aspects of NRTA, to determine assay capabilities and technology limits.[6] The calculated results are all based on the MCNP code, models, and neutron cross-section data used for the validation study shown above. The goal of these studies was to better understand the basic physics behind the NRTA technique and to preliminarily assess its ability to assay a PWR spent fuel assembly. Parametric variables considered in these studies included spent-fuel isotopics, burnup, cooling time, background neutron emissions, and characteristics of the collimated neutron beam. The results of the conceptual studies have

also provided a preliminary basis upon which a conceptual NRTA system design has begun to emerge. It must be stated that the conceptual studies are not complete. Additional conceptual studies are expected to continue into the future, with the possibility that some of the conceptual studies here will be expanded or re-worked as needed.

1.	Fresh Fuel Performance	8.	Transmission Through Multiple Fuel Pins vs. One Pin
2.	Differences Between Fresh Fuel & Spent Fuel	9.	Differences Between Evacuated and Air-Filled Drift Tubes
3.	Effects of Burnup	10.	Potential Interferences from ¹⁰³ Rh at 1.25 eV with ²⁴⁰ Pu at 1.07 eV
4.	Effects of Cooling Time	11.	Inherent SNF Neutron Emission & Impact on Signal Strength
5.	Effects of Neutron Beam Diameter and Scattering	12.	Inherent SNF Photon Emission & Impact on Signal Strength
6.	Performance of a Fan-Shaped Irradiation Beam	13.	Feasibility of Detecting Fuel-Pin Diversion
7.	Performance of an Ideal Pencil-Beam Irradiation	14.	Feasibility of Detecting Missing Fuel Pins

Table 2 Fourteen conceptual study topics for an initial analysis of the NRTA technique.

Studies 1 and 2 demonstrated the driving mechanisms leading to shape in the NRTA spectra. Oxygen and the fuel-clad materials do not impact resonance structure in the spectra. Studies 3 and 4 showed an expected relationship between resonant absorption and the removal of neutrons in the NRTA TOF spectra, scaling with the concentration of the actinides and fission products in the fuel. Studies 5 through 7 were carried out to understand how neutron scattering might serve to blur the resonant features of NRTA spectra. The impact is minimal.



Neutron Energy (eV)

Figure 5 MCNP-calculated transmission spectrum as a function of neutron energy through multiple pins (study 8 from Table 2).

Study 8 investigated the change in transmitted signal versus the number of pins. The results of this study are shown in Figure 5. Transmission through one pin reduces the neutron intensity to \sim 70% of the unattenuated intensity (no pins). Transmission through eight pins produces a neutron intensity roughly 6% of the value for the case with one pin.

Study 9 indicates the value of using an evacuated flight tube versus and air filled tube. Flight tubes of 1, 2, and 4 meters result in attenuation losses of 4, 8, and 16% compared with evacuated flight tubes of the same length. Study 10 examined the fission product ¹⁰³Rh, with a resonance at 1.25 eV, and whether or not its 1.25-eV resonance would pose a significant interference for measuring the 1.07-eV resonance of ²⁴⁰Pu. For concentrations of ¹⁰³Rh found in fuel ranging from 0-60 GWD/MTU no significant interference was observed. Studies 11 and 12 evaluated the background signals present with SNF and their potential impact on NRTA measurements. Problems caused by these neutron and photon emissions from SNF can be minimized by keeping the fuel three or more meters away from the detector system and through the judicious use of shielding. Studies 13 and 14 evaluated how well NRTA can detect the diversion of a single fuel pin anywhere in the bundle (replaced with an unirradiated fuel pin) and removal of a fuel pin (no replacement). The NRTA technique is particularly robust in being able to detect both of these scenarios. Both diversion cases are readily detected using fast neutron radiography alone, thermal neutron radiography, and resonant analysis of the fission products ¹³¹Xe or ¹⁵²Sm.

MASS DETERMINATION

The goal of the NGSI spent fuel assay program is to determine the elemental plutonium concentration of spent fuel; the NRTA measurement approach does not measure elemental concentrations but rather is an isotopic-specific technique. Fortunately, all of the spent-fuel plutonium isotopes appear to be determinable using NRTA. For a SNF assembly assay each isotope would be determined individually and then added together to produce the total Pu inventory declaration. This section provides a simple exercise showing the basic method for how this is done, using the simulation results shown in Figure 4 to determine the ²³⁹Pu atom density.

Atom density may be determined from an NRTA measurement most directly by graphical inspection of a transmission spectrum to determine the transmission loss of the neutron signal at a particular resonance. The transmitted signal is related to the atomic number density and the reaction cross section as shown in Eq. 1.

$$T(E_r) = \frac{b}{a+b} = \exp(-N\sigma x)$$
Eq. 1

In this equation **b** is the signal intensity at the center of the ²³⁹Pu resonance at energy E_r , **a** is the baseline attenuation at E_r , **N** is the ²³⁹Pu atomic number density in the fuel pin, σ is the total neutron interaction cross section at E_r , and **x** is the thickness of the fuel pin corresponding to the measurement location. For the fuel pin simulated in Figure 4 a hand measurement shows a = 0.18 and b = 1.37, as shown in Figure 6. This leads to a value for T of 0.88; the cross-section, σ , for ²³⁹Pu at 10.93 eV is 1,443 b, **x** is approximated as $\pi D / 4 = 0.643$ cm, the mean chord length of a fuel pin with a diameter, **D**, of 1 cm. Solving for N we find the mean atomic density of ²³⁹Pu for this fuel pin was 1.29×10^{-4} atoms•b⁻¹•cm⁻¹ (1.29 x 10²¹ atoms•cm⁻³). The actual ²³⁹Pu atom density used in the MCNP input file used to create the simulation was 1.1992×10^{-4} atoms•b⁻¹•cm⁻¹. It is remarkable, and a testament to the simplicity of the NRTA measurement, that such a simple, straightforward hand calculation can produce a ²³⁹Pu mass estimate within 8% of the 'true' value. Recognizing that much more sophisticated time-spectra fitting algorithms already exist for analyzing TOF data it seems likely that a more detailed analysis, correctly fitting the baseline intensity and using whole area peak fitting versus centroid fitting, could be expected to reach a precision level of just 1-2%.

For comparison with Figure 3, the authors of the NBS study reported determining a ²³⁹Pu atomic density, using the 10.93 eV resonance, of $(2.60 \pm 0.11) \times 10^{-4}$ atoms•b⁻¹, which is equivalent to $(1.672 \pm 0.071) \times 10^{-4}$ atoms•b⁻¹•cm⁻¹. The NBS team reported a measurement precision of 4% for ²³⁹Pu, 2% for ²⁴⁰Pu, and 3% for ²⁴²Pu. As described above many simplifying assumptions exist between the real NBS experiment and the model used for our validation simulations. Despite many potential sources of error between the NBS data and the model it is interesting to also note that in this exercise with our validation

test case, using a graphical-based interpretation of the simulation 'data' results, a ²³⁹Pu determination within 75% of the experimental results was achieved.



Figure 6 This figure shows an expanded view of the simulation results in Figure 4, with the 239 Pu resonance at 10.93 eV highlighted along with the position for determining *a* and *b* in Eq. 1.

SUMMARY

A set of fourteen conceptual studies have been carried out as part of an initial evaluation of the Neutron Resonance Transmission Assay technique for assaying spent nuclear fuel.[6] Results from the simulations suggest sufficient transmission strength and signal differentiability to assay a minimum of 8 pins in a row, more pins may be possible depending upon the intensity of the neutron source used for the measurements. For an 8-pin maximum, 64% of the fuel pins in an assembly can be part of an integral transmission assay measurement. Collimated neutron beams or neutron detector designs that can spatially resolve a fuel-pin diameter have the potential to assay up to 12-pin rows. For a 12-pin maximum, 100% of the fuel pins in an assembly can be part of one or more integral transmission assay measurements. Prior work has shown that a measurement precision of 3% or better can be achieved for assaying a single fuel pin using an ad-hoc experimental arrangement. Continued work is underway at INL to quantify the neurons source strength needed to be able to reach a minimum assay precision of 1% for transmission through 8 pins.

A summary of some of the strengths of the NRTA technique, as identified from these scoping studies, is presented below.

- 1. NRTA has the potential for accurate assay measurements with a precision in the range of 1% 4% uncertainty.
- 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 ²⁴⁰Pu concept.
- In addition to ²³⁹Pu NRTA can identify and assay several additional important fissionable isotopes and spent-fuel actinides directly including ^{235,236,238}U and ^{240,241,242}Pu.
 NRTA can identify the presence of ²³⁴U and ^{241, 243}Am. Americium-241 is of particular relevance
- NRTA can identify the presence of ²³⁴U and ^{241, 243}Am. Americium-241 is of particular relevance in higher burn-up fuels (45-60 GWD/MTU) with large cooling times (>5 years).
 NRTA does not suffer resonance interference effects for ^{235,236,238}U and ^{239,240,241,242}Pu; the
- 5. NRTA does not suffer resonance interference effects for ^{235,236,238}U and ^{239,240,241,242}Pu; the resonances are strong, narrow, and well-separated.
- 6. NRTA can identify 6 resonant fission-product isotopes: ⁹⁹Tc; ¹⁰³Rh; ¹³¹Xe; ¹³³Cs; ¹⁴⁵Nd, and ¹⁵²Sm, which can potentially be used to estimate assembly burnup, cooling time, and diversion, and to verify operator-reported burn-up values.

- 7. The neutron resonance transmission analysis technique is a mature technology with a solid foundation in theoretical physics.
- 8. Nuclear data for spent fuel actinide and fission product isotopes are known to high accuracy, especially the total cross section and resonance parameter data needed for assay determinations.
- 9. NRTA system calibrations with pin/assembly standards can be straightforwardly used to reduce NRTA systematic errors.
- 10. An NRTA system can be designed to be insensitive to spent-fuel gamma radiation.
- 11. If a photoneutron source is used as the NRTA radiation source, the accelerator's gamma flash precedes the NRTA measurement without interference and may be useful for radiography.
- 12. NRTA does not suffer resonance interferences from the presence of oxides in fuel. (Oxides do, however, attenuate the transmitted signal to a small extent.)
- 13. NRTA does not suffer resonance interferences from zircaloy cladding; only minor signal attenuation occurs due to the cladding because of the relatively small zirconium total-neutron-attenuation cross section.
- 14. NRTA does not suffer resonance interference from hydrogen impurities in the cladding.
- 15. The NRTA signal loss from the presence of hydrogen in the cladding is negligible.
- 16. NRTA does not suffer resonance interferences from the multitude of spent-fuel fission products. The single resonances from each of the six resonant fission products identified so far that are in the energy range of interest for NRTA: ⁹⁹Tc; ¹⁰³Rh; ¹³¹Xe; ¹³³Cs; ¹⁴⁵Nd, and ¹⁵²Sm, do not interfere with the actinide-isotope resonances.
- 17. The multiplication factor for a spent fuel assembly in vacuum or air is small; multiplication neutrons are therefore relatively minor and do not significantly affect the transmitted signal for NRTA measurements.
- 18. Background neutrons from spent fuel (spontaneous fission and alpha-n) should be a minor impact through the judicious use of neutron shielding and since there will be a small solid angle to detectors. Background signal subtraction may also be used.
- 19. No exotic-isotopic, high-purity threshold detectors are required; NRTA neutron detectors are total count detectors, and detection can be performed using standard technology.
- 20. Vacuum in the NRTA flight tube may not be required, especially for system designs employing relatively short flight tubes (<5-m); in these cases, flight tubes could be air-filled without serious signal attenuation.
- 21. The NRTA approach may be accurately modeled using conventional neutronics tools, such as MCNP. Relatively simple MCNP models are adequate for NRTA simulations.

A summary of some of measurement requirements for the NRTA technique, as identified from these scoping studies, is presented below.

- 1. The temperature of the spent fuel assembly (spent fuel assembly decay heat) may need to be measured/controlled to eliminate the impact of Doppler broadening on the transmitted spectra.
- 2. The fuel to be assayed may not be immersed in water.
- 3. The range of penetrability of low-energy neutrons into a PWR assembly is fundamentally limited; multiple orthogonal views must be used to reconstruct an assembly's complete burn-up and Pu-inventory profile.
- 4. An intense pulsed neutron source is needed. A raw accelerator neutron spectrum with an intensity prior to moderation of $>10^{12}$ n·s⁻¹ is the estimated requirement at this time.
- 5. A large physical infrastructure is needed to accommodate the accelerator, time-of-flight tube, detectors, and data analysis system. (However, these requirements will likely be small in comparison with other physical infrastructure requirements needed to allow for the handling of spent fuel.)
- 6. New data analysis tools are needed.

An example analysis illustrating how the time-spectrum data obtained from an NRTA measurement may be used to determine isotopic atom densities in a test object has also been shown. Using only a rudimentary graphical interpretation from a validation-simulation dataset we have shown that it is relatively simple to calculate the density of ²³⁹Pu in a sample object to within 8% agreement of a 'known/true' value. Future work will be needed to develop more sophisticated analytical tools for quantifying transmission factors in NRTA data, to quantify the measurement precision achievable for NRTA analyses of whole SNF assemblies, and to identify methods for calibrating an NRTA measurement system. Also, work is planned to explore in more detail the neutron source and detector requirements needed for assaying SNF.

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REFERENCES

- 1. Veal, K. D., et al., "NGSI Program to Investigate Technques for the Direct Measurement of Plutonium in Spent Fuel LWR Fuels by Non-Destructive Assay," Institute of Nuclear Materials Management 51st Annual Meeting, Baltimore, Md., July 11-16 (2010).
- Tobin, S. J., et al., "Next Generation Safeguards Initiative Research to Determine the Pu Mass in Spent Fuel Assemblies: Purpose, Approach, Constraints, Implementation, and Calibration," Nucl. Inst. Meth. Phys. Res. A, doi:10.1016/j.nima.2010.09.064 (in press).
- 3. Conlin, J. L. and Tobin, S. J., "Spent Fuel Library Report," Report LA-UR-10-07894, Los Alamos National Laboratory, Los Alamos, N.M. (2010).
- 4. See for example, "Annual Transuranic Waste Inventory Report 2010," Report DOE/TRU-10-3425, U.S. Department of Energy Carlsbad Field Office, Carlsbad, N.M. (2010).
- 5. Priesmeyer, H. G. and Harz, U., "Isotopic Assay in Irradiated Fuel by Neutron Resonance Analysis," Atomkernenergie (ATKE) 25 (1975) 109-113.
- 6. Sterbentz, J. W. and Chichester, D. L., "Neutron Resonance Transmission Analysis (NRTA): A Nondestructive Assay Technique for the Next Generation Safeguards Initiative's Plutonium Assay Challenge," Report INL/EXT-10-20620, Idaho National Laboratory, Idaho Falls, Idaho (2010).
- Schrack, R.A., et al., "Resonance Neutron Radiography Using an Electron Linac," IEEE Trans. Nucl. Sci. 28 (1981) 1640-1643.
- Bowman, C.D., et al., "Neutron Resonance Transmission Analysis of Reactor Spent Fuel Assemblies," in <u>Neutron Radiography</u>, Barton, J. P. and von der Hardt, P., eds., ECSC, EEC, EAEC, Brussels, Belgium and Luxembourg (1983) 503-511.
- 9. Behrens, J. W., Johnson, R. G., and Schrack, R. A., "Neutron Resonance Transmission Analysis of Reactor Fuel Samples," Nucl. Tech. 67 (1984) 162-168.
- 10. "MCNP—A General Monte Carlo N-Particle Transport Code, Version 5," Report LA-UR-03-1987, Los Alamos National Laboratory, Los Alamos, N.M. (2003).
- 11. "MCNPX—A General Purpose Monte Carlo Radiation Transport Code, Version 2.5.0, MCNPX User's Manual," Report LA-UR-05-0369, Los Alamos National Laboratory, Los Alamos, N.M. (2005).