NEUTRON RESONANCE TRANSMISSION ANALYSIS (NRTA) FOR NUCLEAR FUEL CHARACTERIZATION USING A PORTABLE DT NEUTRON GENERATOR

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ABSTRACT

Neutron resonance transmission analysis (NRTA) is an isotopically sensitive, nondestructive assay technique that has demonstrated value for safeguards applications. Previous research utilized MCNP simulations to assess the technique's feasibility for performing plutonium (Pu) assay in spent fuel assemblies and detecting potential pin diversion. To date, NRTA has been limited to fixed experimental facilities and no on-site NRTA capability exists for safeguards measurements. This work will discuss the potential of using a portable, 5E+08 n/s D-T neutron generator for the characterization of fresh and spent nuclear fuel. Experimental results for analysis of ~1-cm thick depleted uranium targets will be presented to demonstrate the capability to determine presence of uranium (U) and quantify enrichment by analyzing neutron resonances in the 1-50 eV range. MCNP simulations will be presented that characterize the ability of the proposed DT generator NRTA setup to assay U and Pu in fresh fuel and spent fuel, respectively.

INTRODUCTION

Neutron resonance transmission analysis (NRTA) is a non-destructive analysis technique for assaying a target's isotopic composition. NRTA was first used to assay the isotopic composition of spent nuclear fuel in 1975[1] and fresh nuclear fuel in the early 1980s.[2] A decade ago, the feasibility of using NRTA to analyze spent fuel was studied as part of the Next Generation Safeguards Initiative (NGSI) to accurately assay plutonium concentration.[3] Key potential advantages of an NRTA-based fuel assay system include unique identification of fission product isotopes, measurement precision of 5% uncertainty or less, and insensitivity to spent-fuel gamma radiation. Previous work considered large accelerator systems for assay of up to 12 pins at a time, which would require neutron fluxes of the order 1E12-1E13 n/s, but recognized that detecting a diversion of a fuel pin from the assembly could be accomplished using lesser fluxes.[3]

The portable DT neutron generator represents a less intense neutron source which has the benefit of mobility. Potential applications for a more portable NRTA spent fuel assay system include those cited by Chichester and Sterbentz—verification of shipper inventory declarations at large-scale fuel reprocessing facilities, inspection at a long-term storage facility, as well as non-destructive assay of debris materials at the Fukushima-Daaichi complex[3]—and fuel testing as cited by *Vogel et al*.[4] As said in the latter report, "It is possible that, in the next decade, a "small-scale" accelerator source with sufficient neutron production performance might become available to provide "pool side" implementation of the techniques described in this report."[4]

Over the past two years, a DT-based NRTA apparatus has been constructed at the MIT Vault Laboratory. Initial results have demonstrated a capability to identify isotopic resonances for a range of mid- and high-Z elements, including depleted uranium,[5] and to measure uranium enrichment. This study will use MCNP simulations combined with experimental results to assess the feasibility of a DT-based NRTA system for characterizing both fresh and spent fuel.
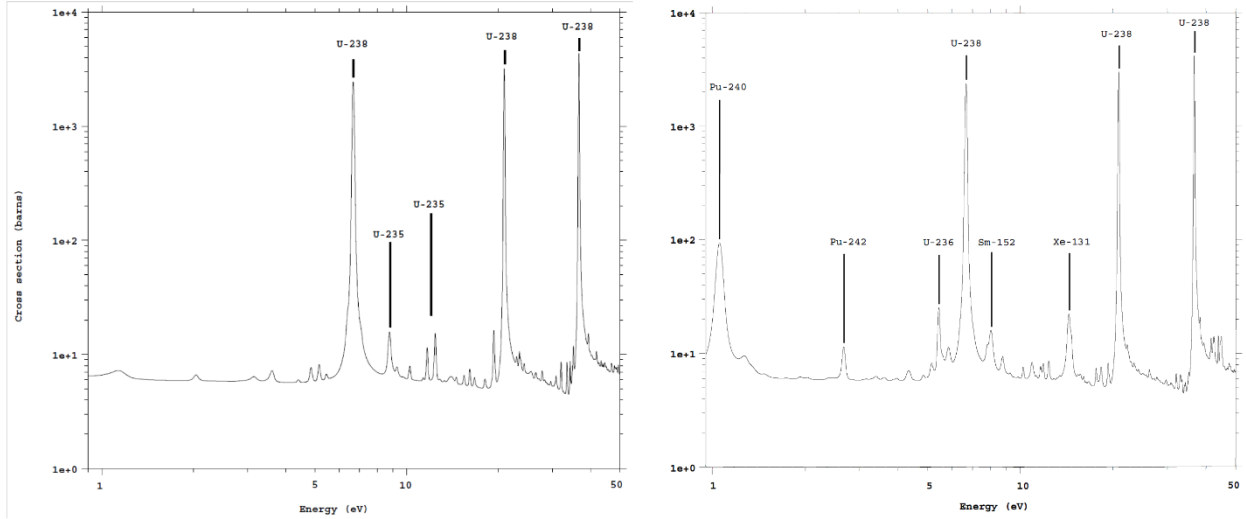

Figure 1. Cross section of (l) a fresh nuclear fuel pellet of 3% ²³⁵U enrichment and (r) a spent nuclear fuel pin of initial 3% ²³⁵U enrichment, a burnup of 30GWd/MTU, and a 20-year cooling time.

BACKGROUND

Neutron resonance transmission analysis can be used to identify the presence of isotopes that have strong neutron resonances in the epithermal energy range. Many mid- and high-Z isotopes have neutron resonances with peak cross sections above 1×10^3 b in the neutron energy range of 0.5-20 eV. The NRTA technique involves analysis of the magnitude and energy of attenuation dips in neutron transmission spectra to reconstruct the isotopic composition of a target object. In combination with evaluated neutron cross section data, the transmission spectrum can be used to estimate the linear density of the different isotopes present in the target object.[6]

Neutron transmission through a homogeneous target at a given incident energy, $T(E_n)$, will depend on the identities and areal densities of the isotopes present in the target, and can be written as

$$T(E_n) = \frac{I(E_n)}{I_0(E_n)} = \exp\left(-\sum_i n_i \Delta x_i \sum_j \sigma_{i,j}(E_n) \right)$$

where $I(E_n)$ and $I_0(E_n)$ are the neutron fluxes measured at the detector for target-in and target-out setups, respectively, n_i is the atomic number density for isotope i, Δx_i is the target thickness for isotope i, and $\sigma_{i,j}(E_n)$ is the interaction cross section for isotope i and reaction j.

Since the resonance energies and amplitudes are characteristic of the isotopes present in the target, the neutron transmission spectrum will be unique for a given target isotopic composition that includes isotopes with epithermal resonances. Although some isotopes have individual resonances sufficiently close to those of another isotope such that the two cannot be distinguished, in most cases the combination of their multiple resonances can enable isotopic differentiation. Figure 1 shows the total neutron cross sections of fresh and spent nuclear pellets with an initial enrichment of 3% ^{235}U. Numerous epithermal neutron resonances are resolvable in this range for both uranium isotopes and fission products.

NRTA provides several benefits compared to gamma-based measurements for spent fuel assay. The high rate of gamma emissions from spent nuclear fuel make passive gamma spectrometry and gamma emission tomography suitable techniques for measuring key parameters of the assembly, such as cooling time, burnup, and initial enrichment.[7] However, the significant measurable gamma emissions are generated by fission products such as ^{134}Cs, ^{137}Cs, and ^{154}Eu, whereas gammas from U and Pu isotopes for the most part will be attenuated within the fuel itself due to their lower energy (~100-200 keV).[8] As can be seen in Figure 1, the most significant neutron resonances for both fresh and spent fuel come from Pu and U isotopes. Therefore, NRTA can provide a direct, rather than proxy, characterization of the uranium and plutonium composition of irradiated nuclear fuel.
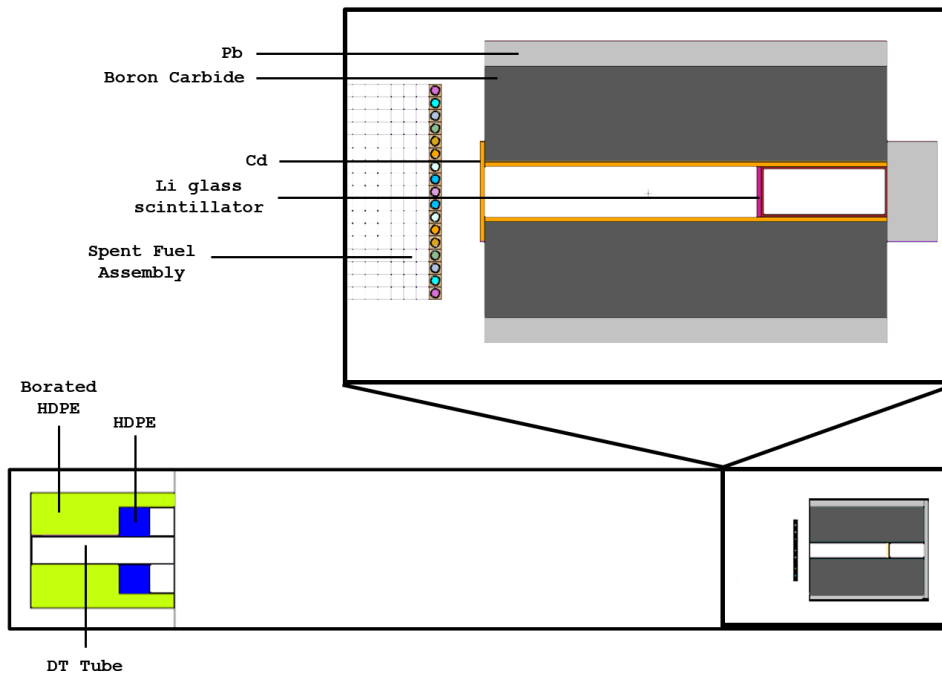

Figure 2. Experimental Setup. The D-T neutron generator is shielded by a box of borated plastic with a radial moderator composed of high-density polyethylene. The moderated neutron beam is incident on a target and detected in a 5-mm-thick, enriched-^{6}Li GS20 glass scintillator coupled to a photo-multiplier tube. The detector is shielded from neutrons by boron carbide and from photons by lead. Thermal wraparound neutrons are filtered by 3.0 mm of cadmium foil placed along the axis and along the collimator tube. The spent fuel assembly row was placed in the plane orthogonal to the TOF axis.

One major disadvantage of a portable NRTA setup for spent fuel assay is that the capability of NRTA to perform isotopic identification and quantification is limited by the thickness of the target and the presence of any coolant, such as the water in a spent fuel pool. Epithermal neutrons have very short mean free paths in hydrogenous material, ruling out the potential for performing an assay "in-pool," without using an evacuated tube or other means of accessing the assembly.

EXPERIMENTAL METHODS

Simulations of neutron transmission through fresh and spent nuclear fuel were performed in MCNP6.2.[9] The simulated experimental setup was modeled after that shown in Figure 2, for which previous NRTA experiments were conducted at the MIT Vault Laboratory. The setup consists of a D-T portable neutron generator surrounded by a high-density polyethylene (HDPE) moderator and borated HDPE shielding. The detector used in the simulation is a 2"-diameter enriched-^{6}Li GS20 glass scintillator coupled to a PMT. The detector is shielded by cadmium and boron carbide to remove thermal neutron background and lead to shield from gamma background. 3 mm of cadmium is also placed along the beam axis to serve as a filter for wraparound neutrons.

The fresh fuel isotopic composition was modeled as UO_2 with an isotopic composition of 96.9595% ^{238}U, 3.0% ^{235}U, 0.0267% ^{234}U, and 0.0138% ^{236}U.[10] The fresh fuel target consisted of five fresh fuel pellets of 1.00-cm diameter and 5.0-cm length placed adjacent to each other with 0.5 mm spacing between. The fresh fuel pellets were surrounded by 1" thick B_4C shielding to limit detection of off-axis thermal neutrons.

The isotopic compositions of spent fuel pins were selected from the Los Alamos National Laboratory (LANL) spent fuel library #2.[11] The assembly consists of a 17x17 matrix of fuel and control rods with a standard gap and Zircaloy cladding with water surrounding the rods within the assembly. The isotopic composition of each individual fuel rod is assumed to be uniform and the composition of individual rods is symmetric about the center rod. For the simulations conducted here, only individual rows of the spent fuel assembly were assayed perpendicular to the axis of the neutron beam. Furthermore, only the central 40 cm of the spent fuel assembly was simulated to reduce computational time. The spent nuclear fuel pins used in these simulations had an initial enrichment of 5% ^{235}U, a burnup of 30GWd/MTU, and a 20-year cooling time.

As previously mentioned, NDA of spent nuclear fuel must contend with a strong gamma background. The use of the GS20 glass scintillator has excellent neutron-gamma discrimination and the ^{6}Li(n,α)^{3}He detection mechanism deposits 4.8 MeV or approximately 1.6 MeVee.[12] Gamma background can therefore come from gamma rays with energies greater than 1.6 MeV contributing to the neutron cut of the pulse area integral, or through pile-up in the detector of multiple lower-energy gamma rays. For the MCNP6.2 simulations presented here, only neutrons interacting through the ^{6}Li(n,α)^{3}He reaction were considered and the strong gamma background from the spent fuel was not measured. In addition to gammas emitted from the spent fuel, gamma background contributions due to thermal neutron capture in HDPE shielding will contribute a background of moderate magnitude in the form of an exponential. Future work will need to more thoroughly explore the sensitivity of the DT-based NRTA system to gamma emissions generated in the spent fuel pellets.

RESULTS AND DISCUSSION

As a means of experimental validation of the MCNP simulations shown for the DT-based NRTA system, neutron transmission measurements were performed at the MIT Vault on a depleted uranium (DU) target of comparable thickness to a nuclear fuel sample. Figure 3 shows the calculated neutron transmission spectrum for this sample for a measurement time of 100 minutes. Neutron resonances of ^{238}U are resolvable up to nearly 70 eV, indicating sufficient energy resolution for identifying the prominent resonances in nuclear fuel samples.
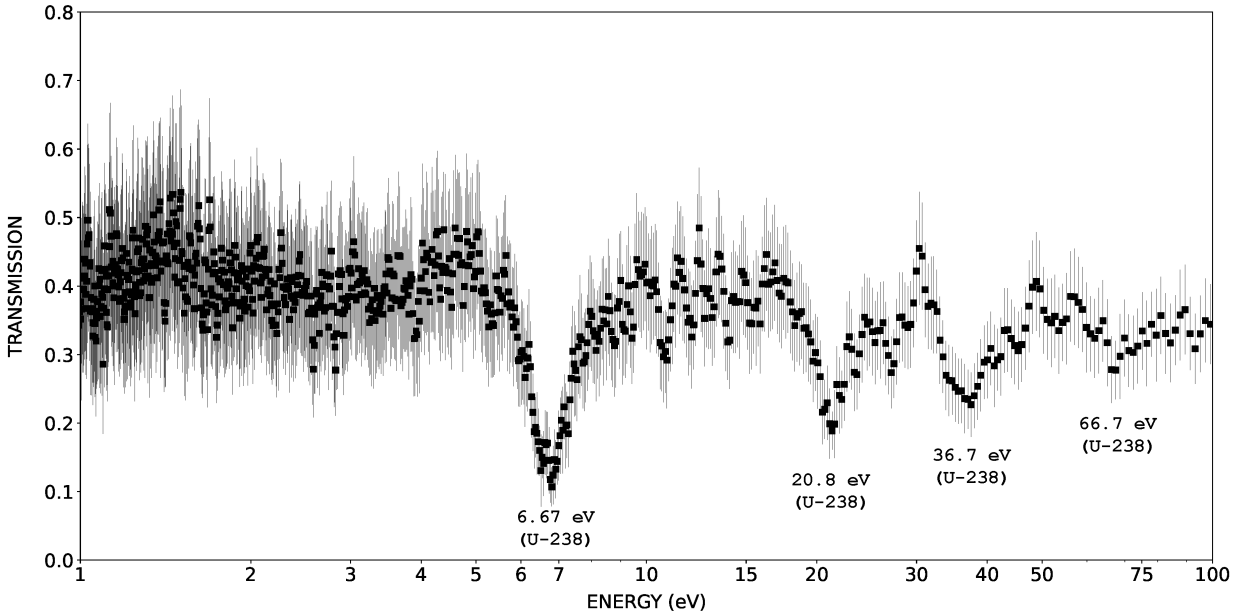

Figure 3. Experimental neutron transmission through a 1-cm thick depleted uranium target for a measurement time of 100 minutes and a bin width of 200 ns.

Given the validation of the experimental setup's ability to resolve epithermal neutron resonances in the range of interest, the next step is to assess the system's ability to identify the ^{235}U resonances present in the fresh fuel. MCNP simulation of neutron transmission through five fresh fuel pellets of 1.00-cm diameter and 5.0-cm length, including the experimentally measured DT neutron pulse width, is shown in Figure 4. The ^{238}U resonances at 6.67 eV, 20.8 eV, and 36.7 eV are readily apparent, as are higher energy >60 eV resonances. The ^{235}U resonances at 8.76 eV and 12.38 eV are also present, though of lesser magnitude. In order to quantify enrichment, a least-squares fit to the transmission data was performed using the REFIT-2009 code,[13] initially assuming a target composed of uranium dioxide with natural uranium composition (0.72% ^{235}U). The REFIT-2009 code settled on a best-fit of uranium enrichment of between 7% and 9% (depending on the parameters of the initial fit), compared to the true value of 3% enrichment. These results indicate that the low enriched nature of the fresh fuel can be identified by NRTA, but greater resolution or longer measurement times may be required to more accurately quantify the enrichment of the fuel.
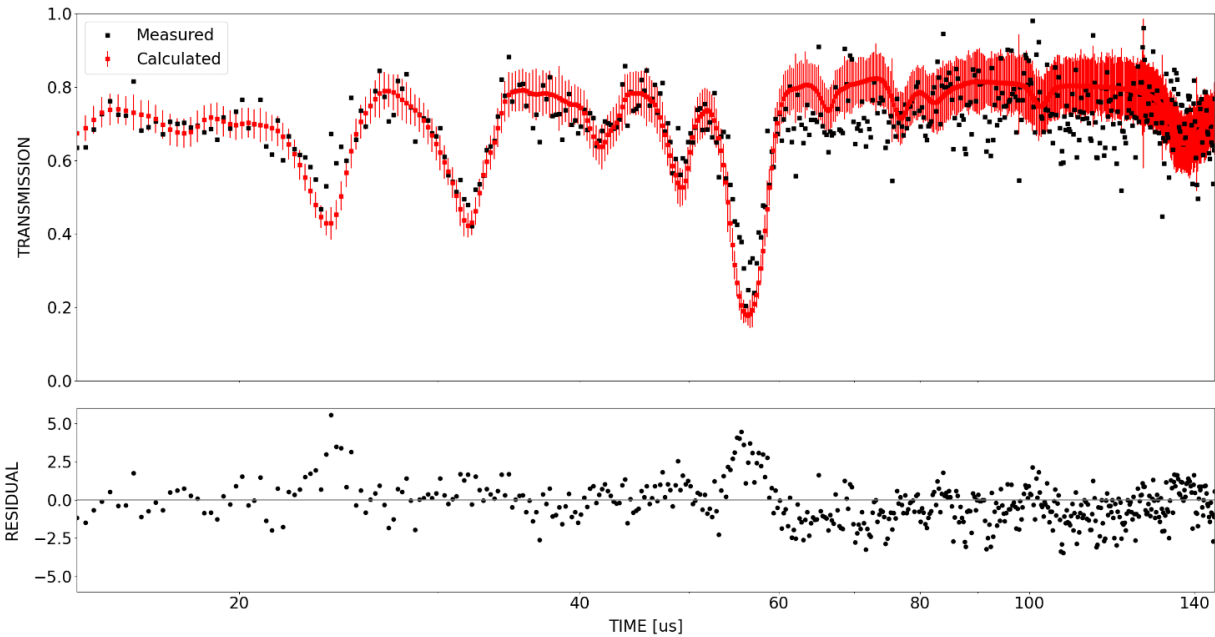
Figure 4. REFIT least-squares fit for MCNP simulations of neutron transmission through fresh fuel (UO_2, 3% ^{235}U). Selected resonances of ^{235}U and ^{238}U are labeled.
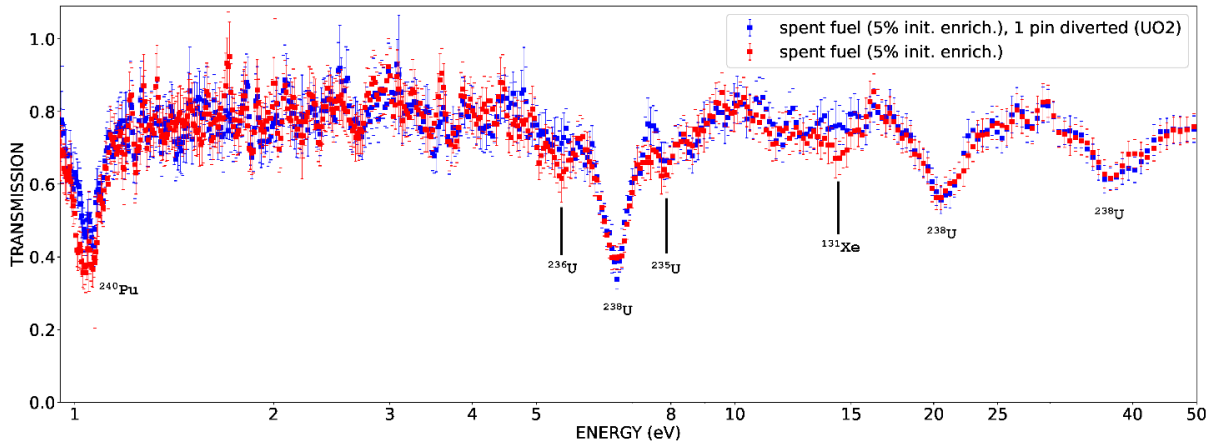

Figure 5. MCNP simulations of neutron transmission through a single row of spent nuclear fuel (blue) and a spent fuel row with the center pin replaced by fresh UO_2 fuel. Selected resonances are labeled. For the measurement of the diverted pin, a decrease in the magnitude of the ^{240}Pu resonance and an increase in the magnitude of the ^{238}U 6.67 eV resonance is observed.

The objectives of spent fuel characterization were twofold: (i) identification of spent fuel composition and (ii) determination of potential fuel pin diversion. Figure 5 shows transmission spectra for a row of spent fuel pellets (5% initial enrichment) and a row of spent fuel pellets (5% initial enrichment) with the central pin diverted (*i.e.* replaced with UO_2), and. Compared with the fresh fuel spectrum in Figure 4, additional resonances are readily apparent in the spent fuel sample due to the modified elemental composition of the fuel following irradiation. Notable resonances include the 1.06 eV resonance of ^{240}Pu, the 5.45 eV resonance of ^{236}U, and the 14.41 eV resonance from ^{131}Xe. By integrating the number of counts in these regions, a measure of the areal density of

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Table 1. Transmitted Neutron Counts in Selected Resonance Regions

Isotope	Resonance Region (eV)	Target	Integrated Counts ($n^{-1} \cdot cm^{-2}$)
^{240}Pu	0.9–1.1 eV	spent fuel (divert.)	$(9.0 \pm 0.1) \times 10^{-10}$
		spent fuel	$(8.2 \pm 0.1) \times 10^{-10}$
^{236}U	5.25–5.75 eV	spent fuel (divert.)	$(4.52 \pm 0.07) \times 10^{-10}$
		spent fuel	$(4.21 \pm 0.07) \times 10^{-10}$
^{131}Xe	13.0–15.5 eV	spent fuel (divert.)	$(7.15 \pm 0.09) \times 10^{-10}$
		spent fuel	$(6.68 \pm 0.08) \times 10^{-10}$
Total	-	spent fuel (divert.)	$(2.07 \pm 0.02) \times 10^{-9}$
		spent fuel	$(1.91 \pm 0.02) \times 10^{-9}$

spent fuel can be calculated. Table 1 shows the MCNP tally results for detected neutron counts for the selected regions.

The ability to differentiate between the full row of spent fuel pellets and the row with central pin diverted will be dependent on both detector area and measurement time. The MCNP tally shown yields the number of neutrons captured in the ^{6}Li per source neutron generated per cm^2 of detector area. The transmitted neutron counts per minute for each sample can be calculated for a 5E+08 n/s neutron source (with an assumed reduction in flux of 60% due to operation at a 5% duty cycle to reduce pulse width) and a 5.08 cm-diameter detector. By resampling from this distribution for each bin and assuming an uncertainty in the counts of $\sqrt{n_i}$, where n_i is the number of counts in bin i, a measure of deviation between the two distributions can be calculated by performing a chi-square two-sample test at a significance level of 0.01 for the three resonance regions indicated in Table 1. Based on the results of the chi-square test, a measurement time of at least 25 minutes would be needed to identify a deviation in the spent fuel neutron transmission spectrum due to the diversion of the central fuel pin. The true measurement time required would likely be considerably longer due to the presence of the gamma background not accounted for here.

CONCLUSIONS

Neutron resonance transmission analysis has been considered for nuclear fuel assay for decades due to its isotopic sensitivity and gamma insensitivity. Extending the range of instruments capable of this technique to portable neutron generators could enable greater use of NRTA for nuclear safeguards and other fuel assay applications. Experimental measurements using a DT-based NRTA system demonstrate the ability to resolve ^{238}U isotopic resonances and MCNP simulations indicate the potential for quantifying uranium enrichment in fresh nuclear fuel pellets, though additional work is needed to reduce systematic error. Although the DT-based NRTA system would not be capable of assaying an entire spent fuel assembly, in situations where portions of the assembly could be assayed, MCNP simulations indicate the ability to resolve neutron resonances of uranium and plutonium isotopes and indicate the presence of a measurable neutron transmission signal as a potential means of detecting spent fuel pin diversion. Additional work is needed to explore the sensitivity of the DT-based NRTA system to gamma emissions generated in the spent fuel.

ACKNOWLEDGEMENTS

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