The Use of Self-Induced XRF to Quantify the Pu Content in PWR Spent Nuclear Fuel

William S. Charlton, Daniel Strohmeyer, Alissa Stafford
Texas A&M University, College Station, TX 77843-3133 USA

Steve Saavedra
Oak Ridge National Laboratory, Oak Ridge, TN 37831 USA

Andrew S. Hoover and Clifford Rudy
Los Alamos National Laboratory, Los Alamos, NM 87545 USA

Abstract:

The development of techniques for the accurate quantification of the plutonium content in spent nuclear fuel would provide significant advances for shipper/receiver differences and for input accountability at reprocessing facilities. Several techniques have been studied previously for achieving this goal but these have met with limited success. Due to the radioactive nature of spent fuel, decay energy is being deposited in the fuel at a relatively constant rate. That decay energy leads to self-induced x-ray fluorescence of the uranium and plutonium atoms in the fuel. These resulting x-rays are then emitted by the fuel rod and can be measured in an appropriately designed and implemented instrument. The presence of uranium x-rays has been observed on numerous occasions; however, due to its dilute nature in the spent fuel and the presence of a large background, the plutonium x-rays have only been observed in a small number of experiments and generally with fuel containing very large loadings of plutonium. In this work, a feasibility study was conducted using both Monte Carlo simulations and measurements of PWR spent fuel rods at Oak Ridge National Laboratory as part of the Coupled End-to-End (CETE) demonstration. This feasibility study demonstrated the measurability of the plutonium x-rays for PWR spent fuel with burnups ranging from 35 to 70 GWd/MTU and the potential application of this technique as a quantitative assay tool.

Keywords: spent fuel; nondestructive analysis; x-ray fluorescence; plutonium quantification

1. Introduction

One of the most difficult non-destructive assay (NDA) problems in safeguards is quantitative measurements of plutonium (Pu) in spent nuclear fuel. Light water reactor spent fuel contains approximately 1% Pu and 3% fission products with the remainder uranium (U). The fission products produce an intense gamma-ray field that obscures the Pu gamma rays. The radioactive decay of the spent fuel however will induce fluorescence in the U and Pu and produce K x-rays. These x-rays might prove to be a useful NDA signature of the Pu to U ratio in the spent fuel.

The U and Pu K x-rays range from about 94 to 120 keV. The energy and relative intensities of these x-rays is shown in Table 1. Since the bulk of the spent fuel is U, the Pu x-rays with similar energy to the U x-rays will likely be obscured. However, the 103.7 keV $K_{\alpha 1}$ x-ray of Pu is relatively well separated from the U x-rays and may be measureable in spent fuel.

Table 1. Uranium and plutonium x-ray data [1].

X Ray	Energy (keV)		Relative Intensity	
	Uranium	Plutonium	Uranium	Plutonium
$K_{\alpha 1}$	98.44	103.76	100	100
$K_{\alpha 2}$	94.67	99.55	61.9	62.5
$K_{\beta 1}$	111.30	117.26	22.0	22.2
$K_{\beta 2}$	114.50	120.60	12.3	12.5
$K_{\beta 3}$	110.41	116.27	11.6	11.7

Measurement of U and Pu x-rays has been performed previously. This signature is routinely measured in small aliquots of spent fuel dissolutions [2]. However, the plutonium K x-rays have only rarely been measured in solid spent fuel. Observations of the 103.7 keV Pu x-ray from spent nuclear fuel previously have been reported by A. Bushuev et al. in 1982 [3] and C. Rudy et al. in 2005 [4]. In the latter paper, x-ray measurements and determinations of Pu/U ratios of BN-350 fast breeder reactor fuel that had been cooling for 5-10 years were reported. The ratio of the 103.7 keV peak area to the continuum was low and the peak areas had large errors, but the detector being used for this measurement was not specifically designed to measure these x-rays. Thus, it is expected that an optimized and collimated detector system might be capable of measuring this signal for spent fuel in a dry hot cell.

In this work, a study was performed to determine the feasibility of using self-induced x-ray fluorescence (XRF) as a direct measure of the Pu content of solid spent fuel pins in a dry hot cell. Experiments were performed on spent fuel rods at Oak Ridge National Laboratory (ORNL) to determine if a well designed measurement system could measure the 103.7 keV x-ray from Pu and if it could be correlated to Pu content in the fuel. Simulations were also performed to aid in detector design, data analysis, and fundamental understanding of this technique.

2. Spent Fuel Measurements

Three measurement campaigns were performed at ORNL in May 2008, July 2008, and January 2009. All measurements were performed in the hot cells at Building 3525 at ORNL and all measurements were for PWR spent fuel in individual rod segments. The measurements in May 2008 failed to produce a viable Pu x-ray measurement due to the large Compton continuum from gamma-ray interactions in the fuel, detector, shielding, and other materials. The detector arrangement was simulated with MCNPX [5] and these simulations suggested possible changes to the detector shielding which would decrease the Compton continuum by as much as a factor of 10. In July 2008, modifications to the detector shielding were performed based on the results from these simulations and this resulted in a measureable Pu x-ray signal. In January 2009, additional modifications to the detector and experimental arrangement were performed to provide an increased signal-to-noise ratio. Thus, Pu/U x-ray ratios were shown to be measureable for LWR spent fuels with burnups ranging from 30-70 GWd/tU. The description in the remainder of this paper is limited to the January 2009 measurement campaign which covered a burnup range of 25-50 GWd/tU.

2.1 Spent Fuel Characteristics

All measurements described here were performed on rod D5 from assembly NJ05YU from TMI-1. This rod had an average burnup of approximately 50 GWd/tU. The characteristics of the rod and assembly are given in Table 2. The rod had been previously cut into segments of varying lengths. These segments were then packaged into stainless steel shipping tubes with a thickness of approximately 0.16 cm. The rods were measured while still in the shipping tubes.

Table 2. Characteristics of TMI-1 spent fuel rod D5 from assembly NJ05YU [6].

Parameter		Value
Assembly and reactor data		
	Design	B&W PWR
	Lattice geometry	15x15
	Rod pitch (cm)	1.4427
	Number of fuel rods	208
	Number of guide tubes	16
	Number of instrumentation tubes	1
	Assembly pitch (cm)	21.8110
Fuel rod data		
	Fuel material	UO_2
	Fuel pellet density (g/cm^3)	10.196
	Fuel pellet diameter (cm)	0.9362
	Clad material	Zircaloy-4
	Clad inner diameter (cm)	0.9576
	Clad outer diameter (cm)	1.0922
Guide/instrument tube data		
	Guide/ instrumentation tube material	Zircaloy-4
	Guide tube inner diameter (cm)	1.2649
	Guide tube outer diameter (cm)	1.3462
	Instrumentation tube inner diameter (cm)	1.1201
	Instrumentation tube outer diameter (cm)	1.2522

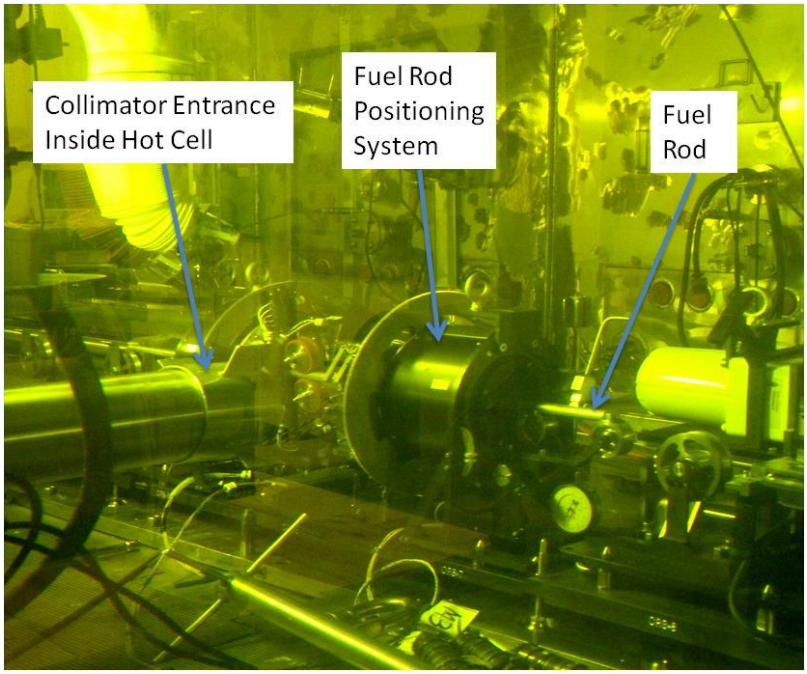

Figure 1. Collimator, fuel rod, and rod positioning system arrangement inside hot cell.

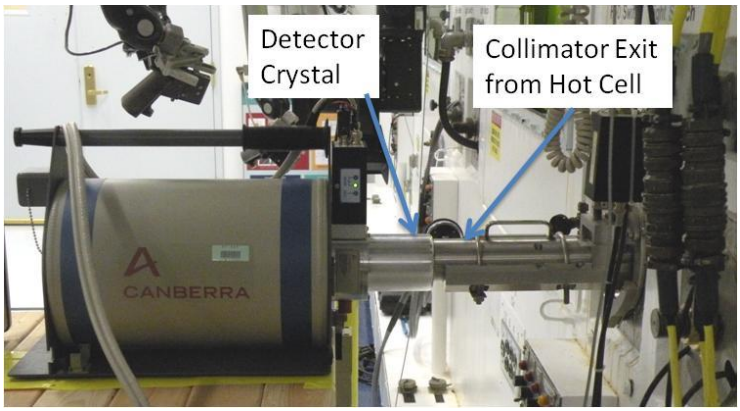

Figure 2. Detector and collimator arrangement outside hot cell.

2.2 Experimental Arrangement and Procedure

The spent fuel rod segments were placed in the hot cell in building 3525 at ORNL. This hot cell contains a rod positioning system (shown in Figure 1) that allowed the rod to be moved laterally in front of a collimator that would collimate gamma rays into a thin beam projecting through the hot cell wall. The collimator was 65.75" long and the hot cell walls were 36.0" thick. The collimator extended 27.5" into the hot cell. The fuel rod was 8" from the end of the collimator inside the hot cell. There was an extension on the collimator that extended 12.25" outside the hot cell wall. A Canberra Model GL0515R planar detector was placed directly against this extension (shown in Figure 2). The tungsten collimator on the detector was removed and no shielding was placed around the detector. Thus, the detector front face was 73.75" from the fuel rod. A similar set of measurements were also performed with a coaxial detector to acquire a broad energy spectrum for each location as well. Spectra were collected from various positions along the fuel rod with the majority of positions being near the top of the fuel rod (where the fuel burnup changed the most with change in distance from the end of the rod). Count times varied from 1-16 hours. Detector dead times ranged from 6-16% depending upon the rod position. With the detector and collimator in this arrangement and using fairly long count times, the 103.7 keV x-ray peak from Pu was clearly visible. An example spectrum is shown in Figure 3.

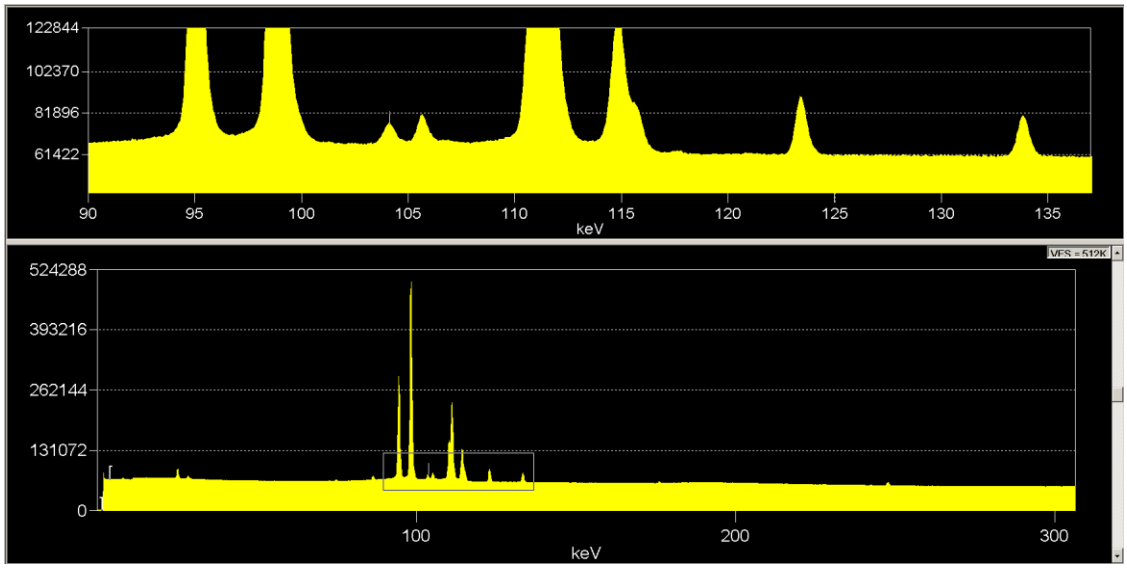

Figure 3. Measured spectra from TMI spent fuel rod D5 showing the Pu x-ray peak at 103.7 keV.

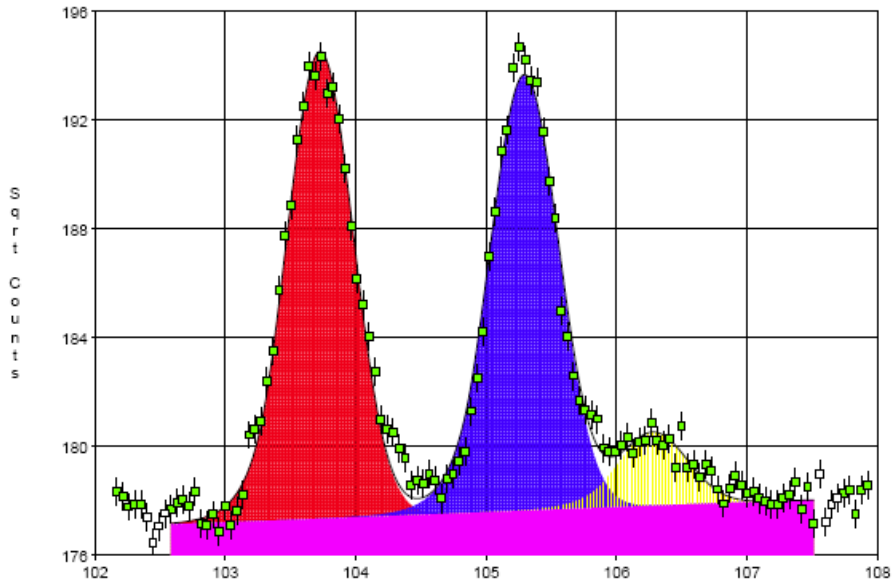

Figure 4. Example peak fit for Pu x-ray peak at 103.7 keV using the GENIE 2000 interactive peak fit.

2.3 Spectrum Analysis

The measured spectra were analyzed using Canberra's GENIE 2000 interactive peak fit. An example peak fit is shown in Figure 4. As can be seen, there is a peak at 105.3 keV from Eu-155 that is near to the Pu K x-ray peak, but with a high resolution detector, these peaks are easily separable. The spectra were analyzed to acquire count rates for the U K x-ray at 98.4 keV, the Pu K x-ray at 103.7 keV, the Cs-134 gamma ray at 604 keV, and the Cs-137 gamma ray at 661 keV. The ratio of Cs-134/Cs-137 is an indicator of fuel burnup [7]. The measured Pu/U K x-ray ratio was then plotted versus the measured Cs-134/Cs-137 gamma ray ratio (Figure 5). As can be seen, a strong correlation between Pu/U x-ray ratio and Cs-134/Cs-137 gamma ray ratio is found.

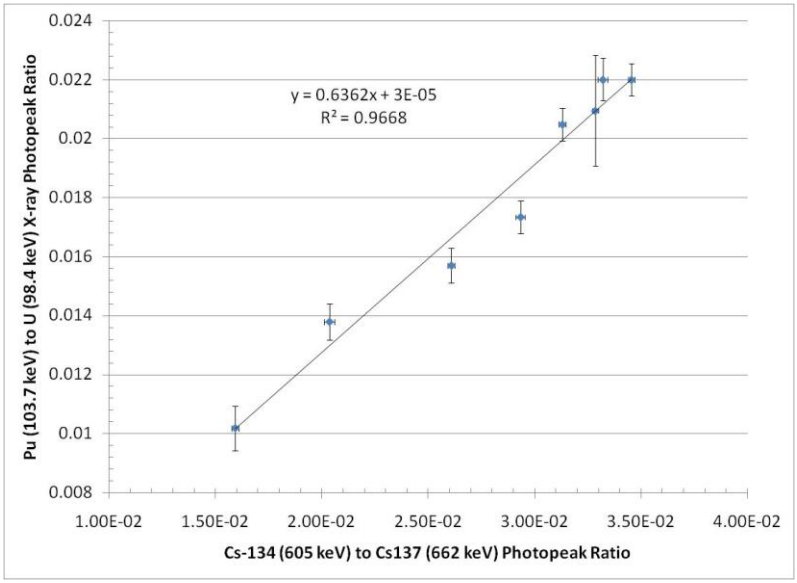

Figure 5. Measured Pu/U x-ray ratio versus measured Cs-134/Cs137 gamma ray ratio for TMI fuel rod D5.

3. Simulations and Results

Destructive analysis of the samples measured for rod D5 had not yet been performed at the time of writing this paper. Thus, simulations were used to relate the measured Cs-134/Cs-137 gamma-ray ratio to burnup and expected Pu/U content in the spent fuel to prove the feasibility of this technique. A two-dimensional TransLAT simulation [8] was used to determine the expected spent fuel isotopics as a function of fuel burnup and radial position in the fuel pin. This simulation included 20 radial fuel regions in the fuel pin. The fuel pin was burned from 0 to 70 GWd/tU and included the declared decay time for the TMI-1 D5 rod from time of discharge to time of measurement. The calculated average Pu/U atom ratio in the fuel pin versus Cs-134/Cs-137 atom ratio in the pin is shown in Figure 6.

When correlated to the measured Cs-134/Cs-137 ratio from Figure 5, we find that the Pu/U atom ratio predicted using Figure 6 is very low. For example a measured Cs-134/Cs-137 ratio of 0.025 would correspond to a measured Pu/U ratio of approximately 0.016 using Figure 5. However, the same Cs-134/Cs-137 ratio of 0.025 would correspond to a Pu/U ratio of 0.003 from Figure 6. It was discovered that this is because the measured Pu/U x-ray ratio is heavily influenced by the concentration of Pu on the outer surface of the fuel pin due to the strong attenuation of the 103.7 keV x-rays in the fuel.

Figure 7 shows the Pu concentration as a function of radial position in the fuel for several burnups. The U concentration only changes slightly as a function of position within the pin, but the Pu concentration is almost a factor of 5 higher on the outer surface of the pin than in the center of the pin. The Cs-134/Cs-137 atom ratio change only slightly (about 5-10%) as a function of radial position in the pin as well, and there is only minimum attenuation of the 605 and 661 keV gamma rays from Cs-134 and Cs-137 regardless of where in the pin they are produced in the pin. Thus, the measured Cs-134/Cs-137 gamma-ray ratio is a good indicator of the average Cs-134/Cs-137 atom ratio in the entire pin.

The TransLAT calculated Pu/U atom ratio on the surface of the fuel pin was correlated to the average Cs-134/Cs-137 atom ratio for the pin. This correlation was then used to infer the Pu/U atom ratio at the surface of the pin from the measured Cs-134/Cs-137 gamma-ray ratio. A plot of the measured Pu/U x-ray ratio versus the inferred Pu/U atom ratio at the surface of the pin is shown in Figure 8. This plot shows that the measured x-ray ratio is a good measure of the Pu/U concentration at the surface of the pin. The Pu/U concentration at the surface of the pin can be directly related to the average Pu/U ratio for the entire pin given knowledge of the pin dimensions (which we would expect would be known). Thus, it is feasible to measure the Pu/U x-ray ratio and Cs-134/Cs-137 gamma ray ratio for LWR spent fuel pins with burnups from 30-70 GWd/tU and use this data to determine the Pu/U atom ratio in the pin.

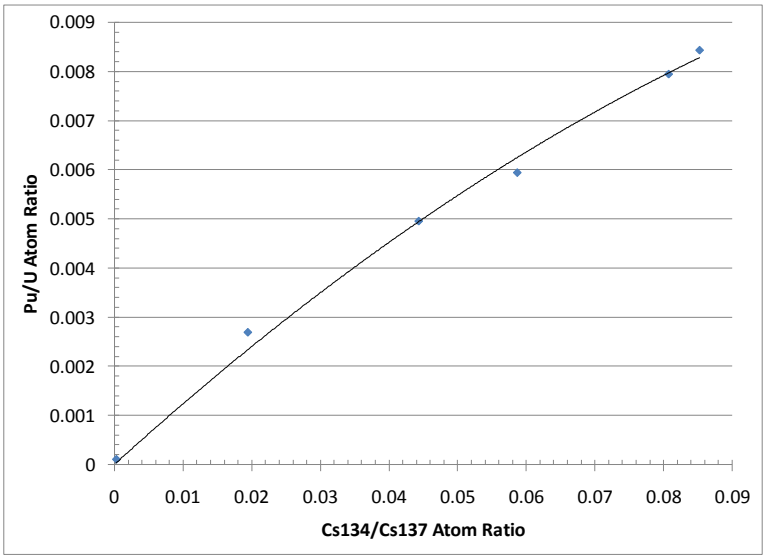

Figure 6. Calculated volumetrically averaged Pu/U atom ratio versus Cs-134/Cs-137 atom ratio for TMI fuel rod D5.

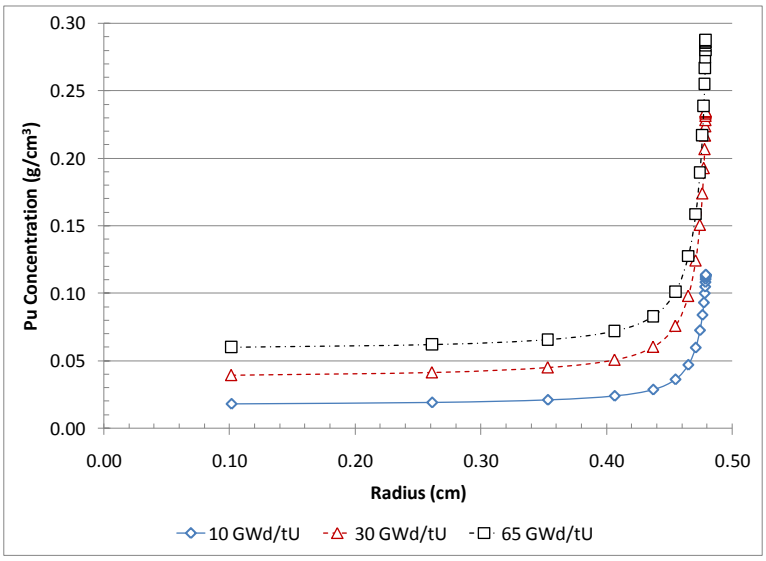

Figure 7. Calculated Pu concentration versus radius for TMI fuel rod D5.

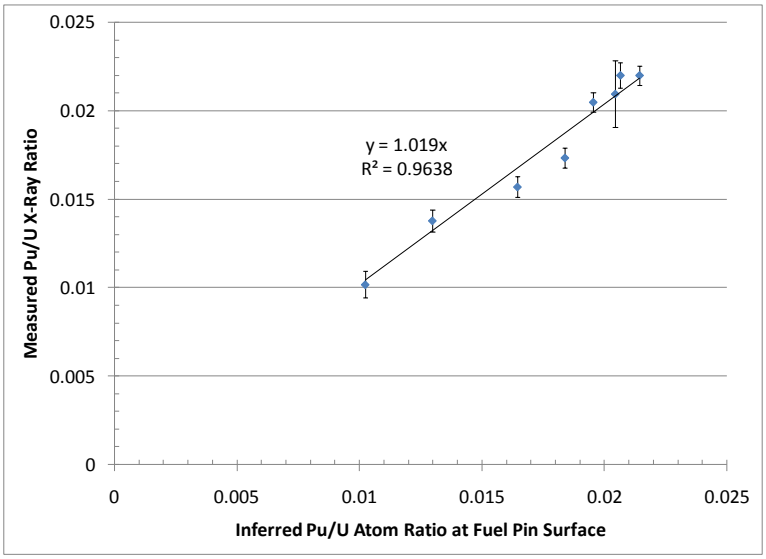

Figure 8. Measured Pu/U x-ray ratio versus Pu/U atom ratio at fuel pin surface inferred from the measured Cs-134/Cs-137 gamma-ray ratio.

4. Conclusion

Measurements and simulations were used to demonstrate that with a properly configured detector system, it is feasible to measure the Pu/U atom ratio at the surface of a fuel pin using measurements of the 103.7 keV K x-ray from Pu. Since the Pu/U atom ratio at the surface of the pin is directly related to the average Pu/U concentration for the pin, this implies that this technique will be feasible for measuring the Pu/U concentration in an LWR spent fuel pin. However, this requires knowledge of the fuel pin design and operating history to properly correlate surface Pu to average Pu for a pin. Also, the count times that were used in these measurements were longer than would be acceptable in most applications. These measurements were also restricted to individual fuel pins and it is unlikely that this technique could be

extrapolated to entire fuel assemblies. For future work, optimized detector systems and measurement approaches will be explored to develop a system that can measure the Pu/U x-ray ratio with shorter count times. Also, the sensitivity of this method to variations in system parameters will be studied to determine the expected accuracy of this technique.

5. References

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