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Spent Nuclear Fuel Measurements

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Wood

July 2014



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Pacific Northwest National Laboratory
Richland, Washington 99352

Executive Summary

An unsolved longstanding challenge to expand the Safeguards regime is to assay spent nuclear fuel (SNF) for its plutonium content. Regardless of the process used to assay the fissile content, there must be a gamma-ray measurement to determine the plutonium mass. Fissile material isotopic measurements in the presence of the intense gamma-ray background from ^{137}Cs and other fission products are difficult. Plutonium-mass measurements using high-resolution passive gamma-ray spectroscopy are practically impossible due to the $\sim 10^7$ higher activity of isotopes such as ^{137}Cs . Assay using measurements of gamma rays with energies above 3 MeV that are emitted from the short-lived decay products of neutron irradiated SNF is more promising but still requires gamma-ray spectroscopy at rates on the order of 1 million counts per second (Mcps) or higher in the gamma-ray spectrometers due to the passive gamma ray background.

PNNL has developed new digital filtering and analysis techniques to produce an ultra high-rate gamma-ray spectrometer that can operate at these count rates. A standard coaxial $\sim 40\%$ efficient high-purity germanium (HPGe) crystal has made passive gamma-ray measurements of SNF at a throughput of about 400 kcps at an input rate of 1.3 Mcps. Optimized filtering algorithms have been developed to preserve the spectroscopic capability of the system even in the very short processing times between events at these high rates. This Ultra-High Rate HPGe (UHRGe) spectroscopy system also operates at real time using a field-programmable gate array to perform spectral operations.

The system operation was demonstrated in a measurement with an aged SNF rod segment in a hot cell at PNNL's Radiochemical Processing Laboratory. This piece of fuel originates from the Quad Cities reactor complex, has a well-known burnup and adjacent sections have been destructively assayed using radiochemical techniques. The UHRGe system was calibrated using known sources and the direct measurements of the SNF matched the predicted rates from the known mass and isotopic composition. This test confirms the promise of the UHRGe system's approach for high-rate gamma-ray spectroscopy of SNF.

Acronyms and Abbreviations

ADC	Analog-to-digital converter
BWR	Boiling Water Reactor
DAQ	Data acquisition system
DGA	Delayed gamma assay
FPGA	Field-programmable gate array
FWHM	Full-width at half-maximum
HPGe	High-purity germanium
NDA	Non-destructive assay
PNNL	Pacific Northwest National Laboratory
PWR	Pressurized Water Reactor
SNF	Spent nuclear fuel
UHRGe	Ultra-High-Rate Germanium

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1.0 Introduction

Each spent nuclear fuel (SNF) assembly contains a fraction of an IAEA significant quantity of plutonium. In the absence of a viable direct measurement technique, the current process can only rely upon operator declarations and modeling to estimate the plutonium content in the fuel. A goal of improved safeguards [1] is to have nondestructive assay (NDA) methods to independently determine plutonium mass in spent nuclear fuel, enabling better techniques to verify declarations, detect pin diversion, reestablish continuity of knowledge, recover after the loss of containment, and determine the mass of material sent to long-term disposition or reprocessing. Multiple measurement techniques will be required to achieve this goal [2-3]. While several neutron measurement techniques are being developed to assay SNF to determine total fissile material mass, assaying the particular special nuclear material (SNM) isotopes will require high-resolution gamma ray spectroscopy [4-7].

The difficulty in making this measurement is that the non-destructive assay (NDA) of a SNF assembly requires a high-resolution measurement in the presence of much larger gamma-ray emissions from the long-lived fission products such as Cs-137. Unlike fresh fuel or separated SNM, spent nuclear fuel has a high fission-product content that makes a direct measurement very difficult, if not impossible. The table below illustrates the problem that the activity of Cs-137 in particular is many orders of magnitude greater than that of the individual fissile isotopes. Given the long half-lives of these isotopes, the values will be relatively stable over decades of cooling time; the only non-negligible impacts will be on Pu-241 ($t_{1/2}=14.35$ years) and Cs-137 ($t_{1/2}=30.07$ years). All of the actinide signatures are many orders of magnitude lower in gamma intensity than the Cs-137 and are at energies where there is significant background from the down-scattering of the dominant 662 keV gamma ray from the decay of Cs-137. The U-235, Pu-240 and Pu-241 gammas are at sufficiently low energy that they will be heavily absorbed in the fuel itself and are unlikely to penetrate shielding (e.g. pool water, lead glass viewport windows, or lead filters). While the Pu-239 gamma-rays are energetic enough to penetrate through the fuel and shielding, they are unlikely to be detectable as they will be buried under the Compton down-scatter of the Cs-137 gammas in the shielding materials and the Compton continuum of the Cs-137 line in the detector itself.

Table 1: Gamma emissions of actinides compared to main fission product emission from Cs-137 for a typical PWR assembly with 3% initial enrichment and 30 GWd/tU burn-up.

Isotope	Mass per Assembly (kg)	Specific Activity (Bq/kg)	Signature Gamma (keV)	Branching Ratio (%)	γ /s for Assembly
Cs-137	0.33	3.2×10^{15}	661.7	85.1	9.0×10^{14}
U-235	3.6	8.0×10^6	185.7	57.2	1.6×10^7
Pu-239	1.9	2.3×10^{12}	413.7	0.001466	6.4×10^7
Pu-240	1.1	8.5×10^{12}	160.3	0.000402	3.8×10^7
Pu-241	1.8	3.7×10^{15}	148.6	0.0001855	1.2×10^{10}

Given that the direct passive assay is practically impossible, an alternative approach is using active interrogation techniques [8] to produce short-lived fission products whose delayed gamma spectrum has

characteristic gamma rays at energies well above the dominant Cs-137 background, as shown in Figure 1. Delayed gamma assay [4] uses thermal-neutron-induced fission to generate fresh, short-lived (few-second to few-minute half life) fission products and relies on detecting gamma-rays with energies in the 3-6 MeV energy range (orange curve in Figure 1), above the passive gamma-ray background from the long-lived fission products (blue curve in Figure 1). Although the actively induced signal is separated in energy from the passive background, the gamma-ray spectrometer must be able to count at extremely high rates with excellent energy resolution due to the lack of an effective filter at these energies.

Detailed model calculations of the delayed gamma assay technique show that even with a very intense neutron generator the specific gamma rays of interest will be produced at rates 10^6 - 10^8 below the passive background rates. Thus, a 3% measurement of a signature isotope will require collection of 10^9 - 10^{11} gamma-rays, and this must be done in a reasonable period of time ($<10^4$ seconds). For example, the Clink facility in Sweden requires assay of one complete canister per day and each canister contains 12 BWR or 4 PWR assemblies [9]. Assuming that each assay views $\sim 1/10$ of the length of the canister and 24-hour per day operation, each measurement must take ~ 2 hours including repositioning of either the fuel or the detection system between measurements.

Calculations for this scenario show that with the measured passive background and the modeled delayed gamma spectrum as the signal, the gamma-ray spectrometer must still operate at counting rates of order 10^6 - 10^7 cps in order to collect sufficient statistics for the signals of interest in acceptable times (~ 1 day per assembly). Note that this is the rate of events processed into an analyzable spectrum and not merely the input rate to the system. Very good energy resolution is still required since the gamma-ray spectrum must be analyzed for a large number of spectral features in which only a relatively small number provide high degrees of discrimination for Pu isotopes and U-235. In addition, the fast neutrons from the fission of U-235, Pu-239 and Pu-241 will induce fission in the dominant U-238 in the fuel at a rate of roughly 10% of the thermal neutron-induced fissions (see Appendix). While the energy resolution of typical laboratory high-purity germanium (HPGe) detectors is not necessary, resolution of better than 10 keV FWHM is still required in the 3-6 MeV energy range, ruling out use of scintillation detectors. The high processing rate and associated fast charge collection requirements in the detector element rule out room temperature semiconductors such as CZT that have poor mobility and thus slow charge collection. The most viable technique is to use germanium, the only material available with both the spectroscopic performance and fast charge collection time, as the detector element, and focus on the engineering of high-rate electronics and processing algorithms that can collect and spectroscopically process about 1 Mcps of gamma-rays.

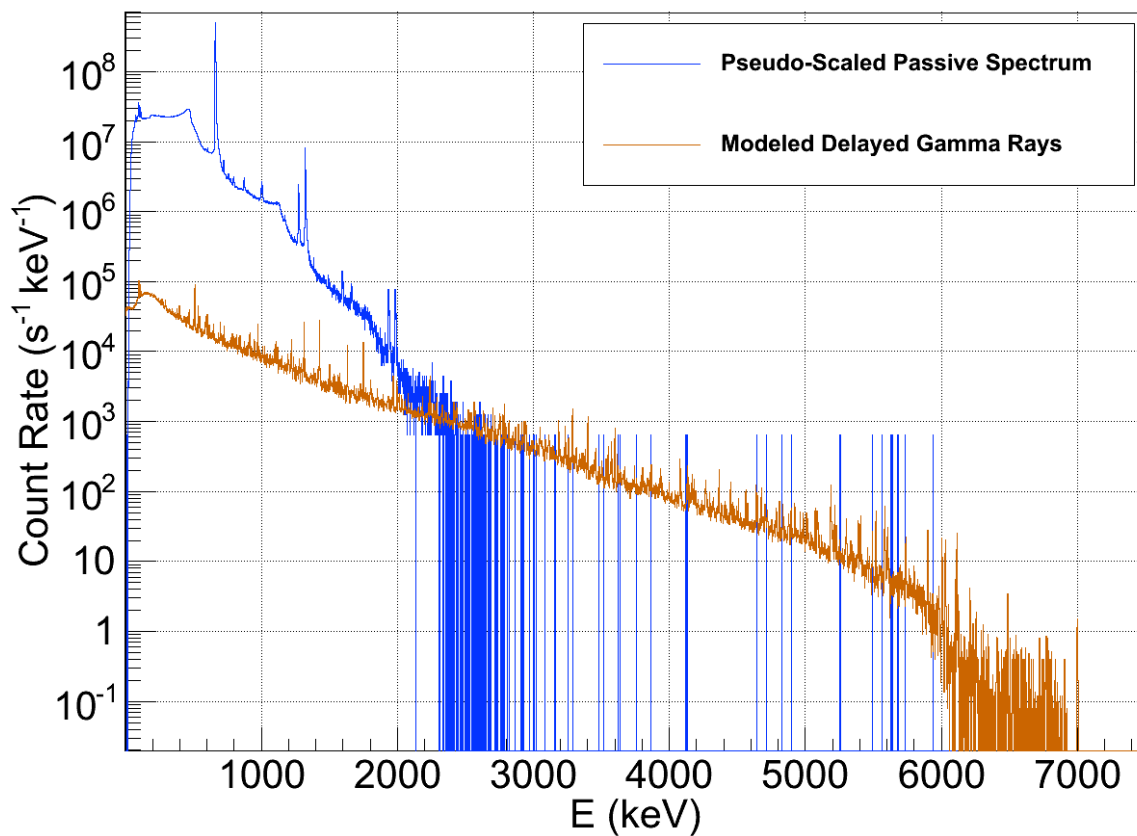


Figure 1: Measured passive background of ~65 GWd/tU burnup, 22.5 year cool down BWR fuel scaled to match equivalent exposure time for simulated delayed gamma assay scenario with 10^{10} n/s (thermal) neutron irradiation with 10 second irradiation/10 sec measurement cycles.

2.0 The UHRGe detector system

The above constraints are unlikely to be met in a standard HPGe detector system that has a coaxial detector diode whose signal is processed using long (approximately 4-16 μ s) filtering times. Pacific Northwest National Laboratory (PNNL) has developed a new approach to HPGe signal processing that offers an order of magnitude improvement in pulse throughput even with a conventional co-axial germanium diode and promise for an additional order of magnitude or more when these techniques are coupled to a HPGe strip detector [10]. The diode used in these measurements is a 62 mm diameter, 45 mm tall *p*-type semi-coax from Princeton Gamma Tech (PGT) with a nominal relative efficiency of 37%. For convenience the symmetry axis of the detector was vertical. Even with the side of the detector facing the source, the geometrical acceptance in this configuration is 90% of that of normal-to-the-end-face area that is used for the standard efficiency calculation. The PGT front-end electronics and RG-11 preamplifier have been modified to allow for the high steady-state currents when operating at $\sim$ Mcps rates. Modifications include lower feedback resistance (1 G Ω), increased feedback voltage rail (-100 V), and shorter tail time in the second stage of the amplifier (36.5 μ s). The intrinsic charge collection time for this HPGe diode - and in general for semi-coax detectors - is rather long (700-800 ns). This is a limiting factor in the throughput of the current system and will be addressed in the next evolution of the system (described below) that will employ planar strip diode detectors that will have charge-collection times of 200-250 ns. An additional system-level cost is that more readout channels will be required and the thin detector elements will have less absolute efficiency for full energy deposition of MeV gamma rays. The final system is expected to be a stack of several $\sim$ 1-2 cm thick planar detectors to recover the stopping power and efficiency for the gamma rays of interest.

The data acquisition system was built around capturing the preamplifier waveforms for subsequent filtering in either an offline computer analysis or a real time online FPGA processor with firmware filters. The waveform digitizer must be high speed, low noise, and have an onboard field programmable gate array (FPGA) for real-time spectral filtering. A Signatec PX14400D (14-bit, 400 MHz sampling) and an AlazarTech ATS9626 (16-bit, 250 MHz sampling) waveform digitizer were evaluated [11]. The Signatec unit demonstrated an unacceptable 350 kHz sinusoidal “noise” generated internally in a DC offset circuit and was subsequently rejected. The AlazarTech ATS9626 has an Altera Stratix III EP3SE260 FPGA dedicated for user programming. At these input rates the data-acquisition system requires continuous (dead-time free) recording of waveforms. A Signatec DR-800 system with 8-lane PCI-e data transfer to a 96 TB RAID array (2.8 MSPS maximum transfer rate) was selected; this system can stream raw digitizer data continuously for $\sim$ 16 hours, though post-analysis of this volume of data is impractical. For the work presented in this paper, waveforms were recorded to disk and analyzed offline, but the analysis algorithms were required to be implementable in FPGA firmware so that they can be moved to real-time in the future.

As seen in Figure 2, at Mcps rates the waveform has a heavy and continuous pileup where every new pulse is riding on the sum of the exponential tails of 100's of previous pulses that have occurred in the prior several RC time constants of the preamplifier output. The quiescent baseline is at 0. The signals are upward going impulses with a 55.7 μ s RC decay constant for this data, equivalent to the width of the waveform section shown in the lower half of the figure which contains $\sim$ 40 individual gamma ray events. The DC level is shifted by 4000-5000 channels in the filter output value (the gain is roughly 4.9 keV per channel) with significant fluctuations due to the Poisson fluctuations of the radioactive decay of the Cs-137 source.

The signal processing technique used is based on conventional trapezoidal filtering techniques [12-13]. A fast triangular filter (80 ns rise time) is used as a trigger filter to identify pulses and to determine event times. In addition, the time over threshold for this filter can be used to identify accidental coincidences (trigger pileup). Energy filters are trapezoidal with gap times fixed to 800 ns (slightly longer than the longest charge collection time in the HPGe diode) and rise times from 32 ns to 4 μ s. The ideal trapezoidal filter rise time at low rates is found to be 2 μ s; this should be considerably longer for this HPGe diode and preamplifier but the digitizer used is introducing some additional low frequency noise that degrades resolution for long filter lengths. This is not a concern for high rate operation where one typically gets maximum usable filter rise time around 512 ns. We employ a variable length filter where the longest filter that can be evaluated between the previous and next events is used to evaluate the current event. Our method, developed independently, is similar to that presented in [14]. The resulting energy spectrum for the Cs-137 source is shown in Figure 3. One can readily see the large increase in throughput achieved by using a variable length filter as opposed to the fixed length (512 ns) that represents the optimum at this count rate (1.1 Mcps input). One can see the single 661.7 keV line as well as the 2X and 3X 661.7 keV lines from accidental coincidences. The right-hand panel in Figure 3 shows how one can suppress accidental coincidences (trigger pile-up) using a cut on the time-over-threshold of the trigger filter output.

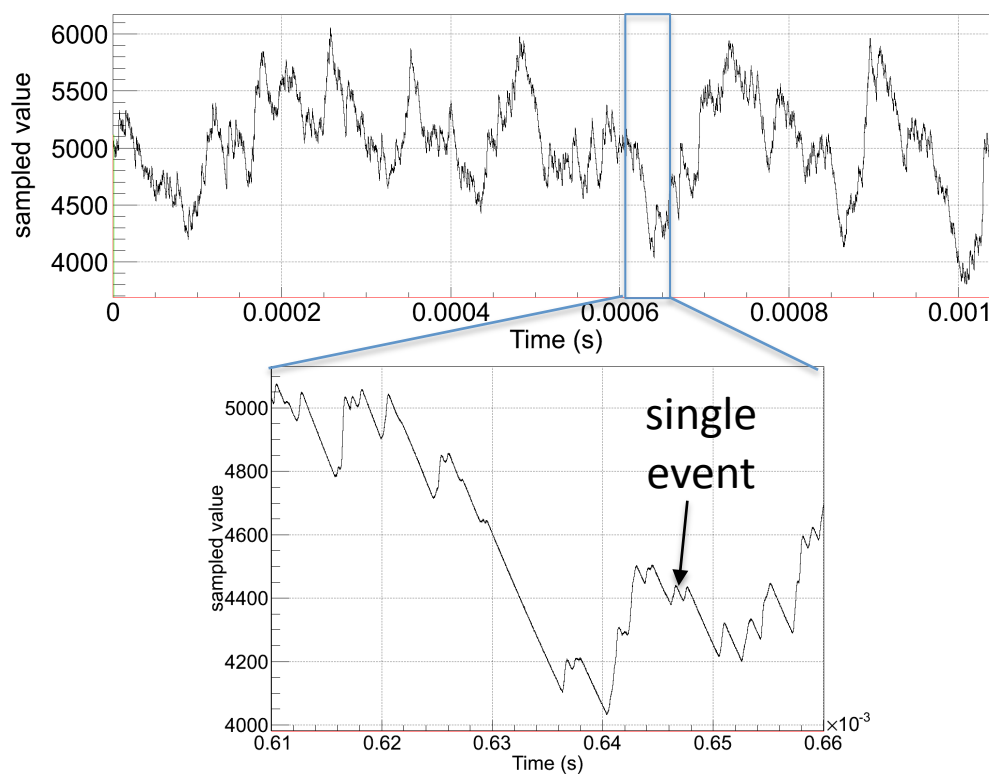


Figure 2: Waveform collected at input count rate of ~ 1 Mcps using a 722 uCi Cs-137 source. Above: 1 ms trace. Below: 50 μ s section of waveform.

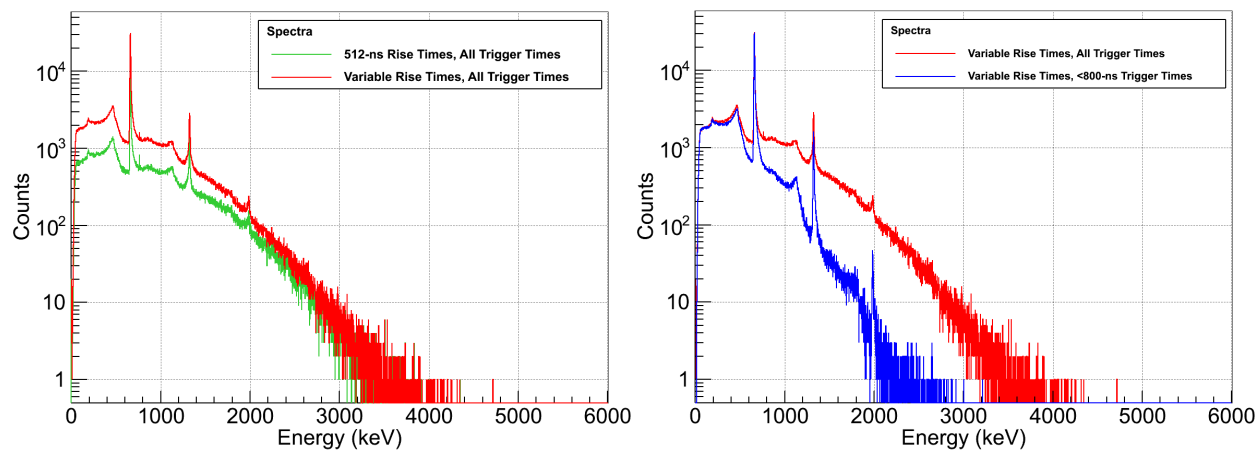


Figure 3: Energy spectrum from 722 uCi Cs-137 source at ~ 1.1 Mcps input rate. Left: energy spectra for variable length and fixed length filters. Right: spectrum for variable length filter with and without cut on time over threshold of trigger filter.

3.0 The Spent Fuel Measurement

Passive gamma-ray measurements were made on a 58.2 g sample of ATM-109 fuel shown in Figure 4. ATM-109 is high-burnup-BWR fuel with an initial enrichment of 3% U-235 that originates from an assembly irradiated in the Quad Cities I reactor from Feb 1979 through September 1987 to a burnup of 43 GWd/tU and again from November 1989 through September 1992 at an average exposure of ~70 GWd/tU, though the burnup estimates for this fuel range from 60-75 GWd/tU [15]. The cool down time from last irradiation to the measurements described in this paper is 21.5 years. The sample used in the measurements is 58.1993 ± 0.0006 g, including the cladding, and measures about 5.5 cm in length. Measurements were made with the detector viewing the fuel through a large diameter (~6") open port in a hot cell at the Radiological Processing Laboratory at PNNL. The detector was located in a waste storage area behind the hot cells at distances of ~403 cm and ~803 cm from the fuel. Measurements were taken with no shielding and with up to 3/4" lead shielding to study the impact of "light" shielding on the detector rate and performance.

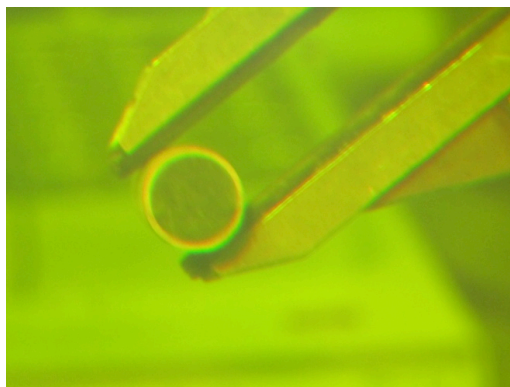


Figure 4: ATM-109 spent fuel rod segment.

The gamma-ray energy spectrum shown in Figure 5 was obtained at ~803 cm standoff with no shielding. A triangular filter with rise time $t_r=80$ ns was used as a "trigger" to identify individual pulses in the recorded waveforms. The distribution of time between triggers was fit for an exponential to estimate the rate of triggered events; the rate was 413 kHz for this running condition. A trapezoidal filter with $L=512$ ns (rise time) and $G=800$ ns (flat top) was used to determine the pulse height (energy). Several event quality cuts are made based on the trigger filter output to select the events that are displayed in the energy histogram. Most important of these is a cut on the time over threshold of the trigger filter, which was restricted to the range 300-700 ns; the lower bound removes spurious events from baseline fluctuations while the upper bound eliminates "trigger pileup" events (accidental coincidence) where the trigger filter does not return below threshold between gamma ray interactions. The resulting rate of events recorded in the energy histogram is 167 kcps, or 40% of the triggered events.

Figure 5 clearly illustrates that the passive spent fuel spectrum has a large amount of trigger pileup remaining; in fact, most of the events above the 661.7 keV Cs-137 peak are trigger pileup events with one or more Cs-137 gamma-rays (full energy peak or Compton) with the tail of this distribution extending up to the region where delayed gamma signatures of interest reside. The 700 ns bound on rejecting this pileup is determined by the (longest) charge collection times in the HPGe diode. As mentioned above, this also sets the lower limit for the gap time of the energy filter (800 ns) used in the analysis. Both of

these can be reduced to ~200-300 ns using a planar (strip) detector configuration and this is being pursued as the next evolution of the system development. This should greatly improve throughput and reduce accidental coincidence events due to trigger pileup.

3.1 Comparison of measurements with known fuel history

At long cooling times, the gamma ray emissions of spent fuel are dominated by Cs-137. The mass of Cs-137 can be estimated based on the sample mass, burnup and cool down time. Production is ~6.25 g Cs-137 per GWd/tU burnup. This equates to 22 mg Cs-137 in 50 g ATM-109 (for 70 GWd/tU). The average production time of the Cs-137 is about 25 years (see irradiation history above) so the current Cs-137 mass is estimated to be 1.2 mg with an activity of 40 GBq and 661.7 keV gamma-ray production rate of $\sim 3.6 \times 10^{10}/s$. The observed rate of 661.7 keV gammas is $2.8 \times 10^4/s$ unshielded at ~800 cm standoff. Random coincidences in the detector will shift Cs-137 gamma rays out of the photopeak (to higher energies). The rate of events in the 2×661.7 keV peak ($3.8 \times 10^2/s$) and the peak-to-total ratio for the detector (~0.2) can be used to estimate the magnitude of this effect; correcting for this ~7% loss, the 661.7 keV photopeak event rate is $3.0 \times 10^4/s$. With a $5\text{ cm} \times 1\text{ cm}$ source at this range a point source approximation is appropriate. We can scale the observed rate at 803 cm to the expected rate at the nominal 25 cm range used for the ASME standard for efficiency measurements; the result is $3.1 \times 10^7/s$ or an absolute photopeak efficiency of 0.086%. This is consistent with, but somewhat lower, than expected for this HPGe detector indicating that the assumed 70 GWd/tU burnup estimate is too high and that the burnup is closer to 65 GWd/tU. In addition to Cs-137, both Cs-134 and Eu-154 are observed in the spectrum. For Cs-134 the 563, 569, 604.3, 795.6 and 802 keV lines were observed. For Eu-154 the 247.2, 591.2, 723, 757.8, 873.1, 994.1, 1004, 1275, 1493 and 1597 keV lines were observed. The 757.8 keV feature in the spectrum includes a significant contribution of accidental coincidence summing of the 661.7 keV Cs-137 line with 96.5 keV uranium x-rays. The Cs-137/Eu-154 ratio can be used to estimate the burnup [16] with a result of 67 GWd/tU, in reasonable good agreement with the pedigree of the sample.

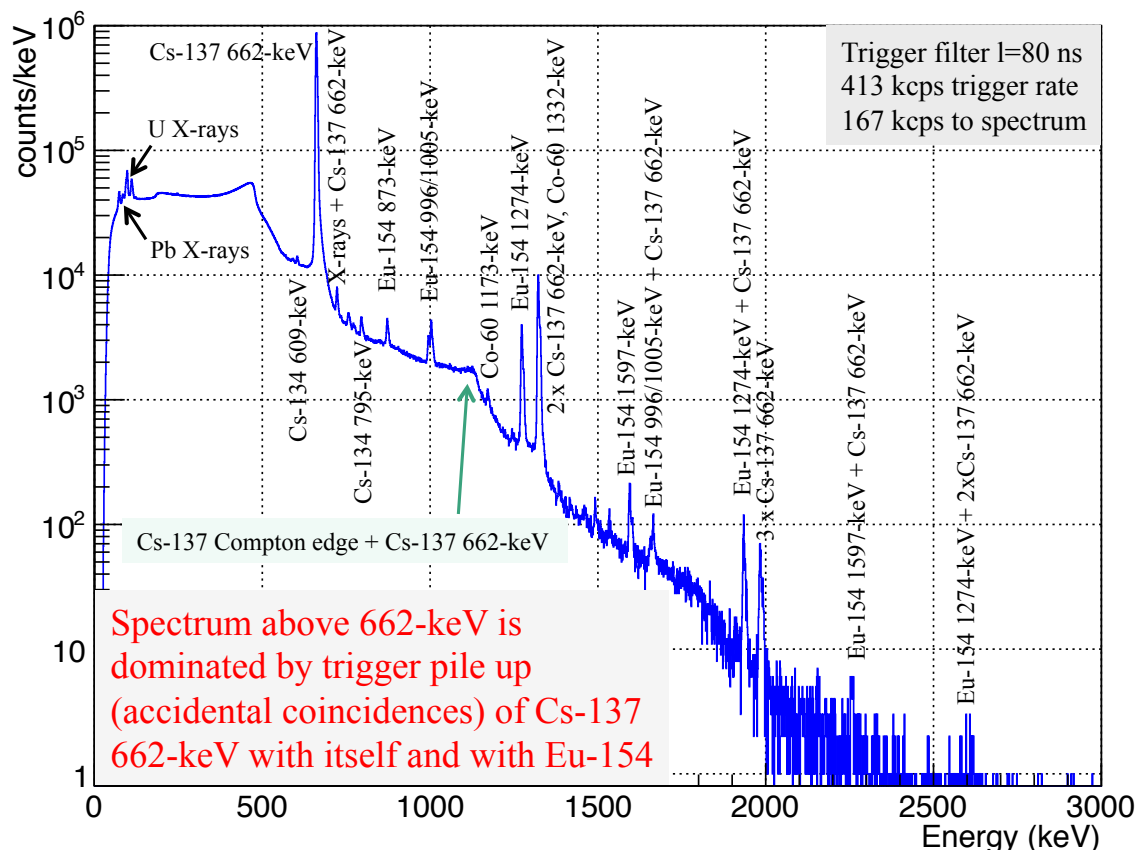


Figure 5: Energy spectrum obtained at 803 cm with no shielding.

Measurements were made at the near (403 cm standoff) position with no shielding and lead shielding of $\frac{1}{4}$, $\frac{1}{2}$ and $\frac{3}{4}$ inch thickness. The results are shown in Figure 6. The bare case has an input rate of 1.4 Mcps. The $\frac{1}{2}$ " lead shielding case at 403 cm has a very similar count rate (~ 500 kcps input) and spectral shape as the data collected at 803 cm with no shielding. The bare fuel data include the ~ 100 keV uranium x-rays while the shielded data do not. The shielded data has roughly the same yield for the 661.7 keV Cs-137 peak but lower Compton continuum at low energy. At higher energy (above the 2X 661.7 keV accidental coincidence peak) the continuum is nearly identical while the photopeaks for the higher energy (1274.4 keV, 1596.5 keV) Eu-154 lines are slightly larger.

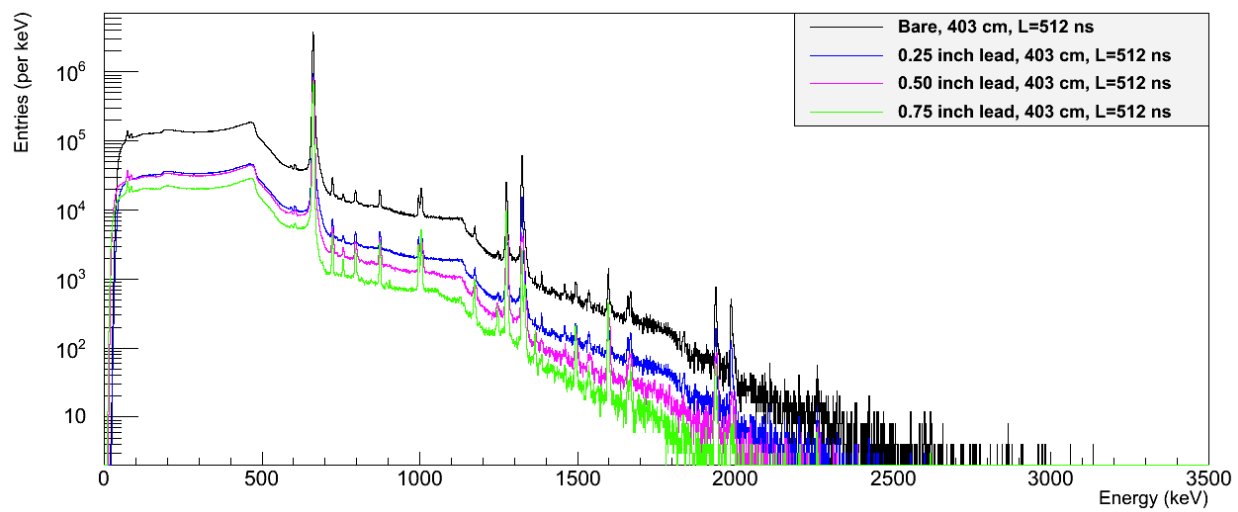


Figure 6: SNF data taken at 403 cm standoff bare and with various thicknesses of lead shielding.

4.0 Current and future development

This measurement was made using stored waveforms and offline analysis. Since this measurement, the UHRGe system has been further developed and the real time processing implemented with the offline analysis algorithms implemented in the digitizer FPGA and real-time data-collection in the host CPU. The FPGA performs the filter algebra on the data stream in real-time while the host CPU performs the final energy calculations that require floating point division. A total of (at least) 8 filters can be evaluated concurrently at full data rate. At present, the energy resolution from the real-time analysis is slightly worse than from the offline code. The filters being used have a minimal gap time (flat top) so the output is extremely sensitive to precise event timing and it is suspected that this may not yet be getting calculated correctly in the real-time processing.

To further improve the rate capability it will be necessary to speed up the charge collection time in the detector diode and go to a strip design that distributes the gamma rays over multiple readout channels. A planar strip detector is being developed by Lawrence Berkeley National Laboratory for the next iteration of the system. The planar geometry will result in much shorter drift times (~ 200 ns vs. ~ 700 ns), which will directly and dramatically increase the throughput and reduce the accidental-coincidence (trigger-pileup) rate. This new detector is segmented into 10 strips, and assuming the demonstrated UHRGe rates with the coaxial detector, the total input-rate capability for the system will be in excess of 5 Mcps and the usable spectroscopic event rate should be well over the target 1 Mcps.

As part of the larger SNF assay project, gamma-ray mirrors are being developed by the Lawrence Livermore National Laboratory. Those multilayer mirrors have high reflectivity even at energies up to 600 keV [17-19]. It may be possible to use a mirror assembly to deflect the dominant 661.7 keV gammas out of the line of sight to the detector. Four consecutive 80% reflectivity elements would be required to suppress the dominant gamma line by a factor of 625. Since these mirrors have very shallow reflection angles it is necessary to place them as close as possible to the source (fuel assembly). The manufacturing limitation for these mirrors might dictate different collimation and scanning geometry such as a longer slit collimation rather than viewing a ~ 22 cm $\times$ 22 cm square length of an assembly or similar section of a cask.

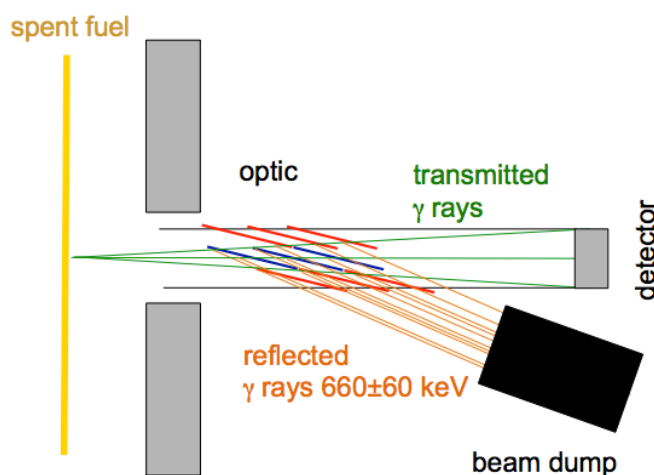


Figure 7: Proposed layout for use of gamma-ray mirrors to deflect dominant Cs-137 gamma rays away from the detector to reduce "background" rate and hence assay time for given detector rate limit.

5.0 Summary

A HPGe spectrometer has been developed and evaluated for high rate operation using Cs-137. A 50 g sample of high burnup BWR spent fuel has been assayed bare and with light shielding up to $\frac{3}{4}$ " of lead. Data have been collected using high burnup BWR spent nuclear fuel at input count rates as high as 1.4 Mcps with throughput as high as 420 kcps and 247 kcps after rejecting trigger pileup events. Analysis of the Cs and Eu observed is consistent with the known fuel history. Evaluation of potential delayed gamma signal rates and the measurement speed requirements at nuclear facilities confirm that the required rate of usable events in the energy spectrum must be in excess of 1 Mcps. Achieving rates of a few Mcps will require use of segmented planar HPGe detectors, currently under development. We anticipate integration of the electronics and data acquisition techniques presented in this paper with the new strip detector into a robust laboratory system in the coming 2 years. In addition, a concept for deflecting the primary spent fuel gamma ray emission at 661.7 keV using a multi-layer gamma ray mirror will be tested this year.

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7.0 Appendix 1: Neutron Accounting in SNF

U-238, the primary actinide present in the fuel at >95% by mass, has an extremely low neutron induced fission cross section for thermal and epithermal neutrons. However, U-238 has a significant fission cross section for neutrons above ~1 MeV as seen in Figure 8. This motivates the choice of thermal neutrons as the proposed interrogation modality for delayed gamma assay, as well as other active neutron assay techniques. However, one must carefully consider the impacts of neutron-induced fission neutrons as well as spontaneous fission and (α ,n) neutrons which have energy distributions that will fission U-238 as well as the other actinides.

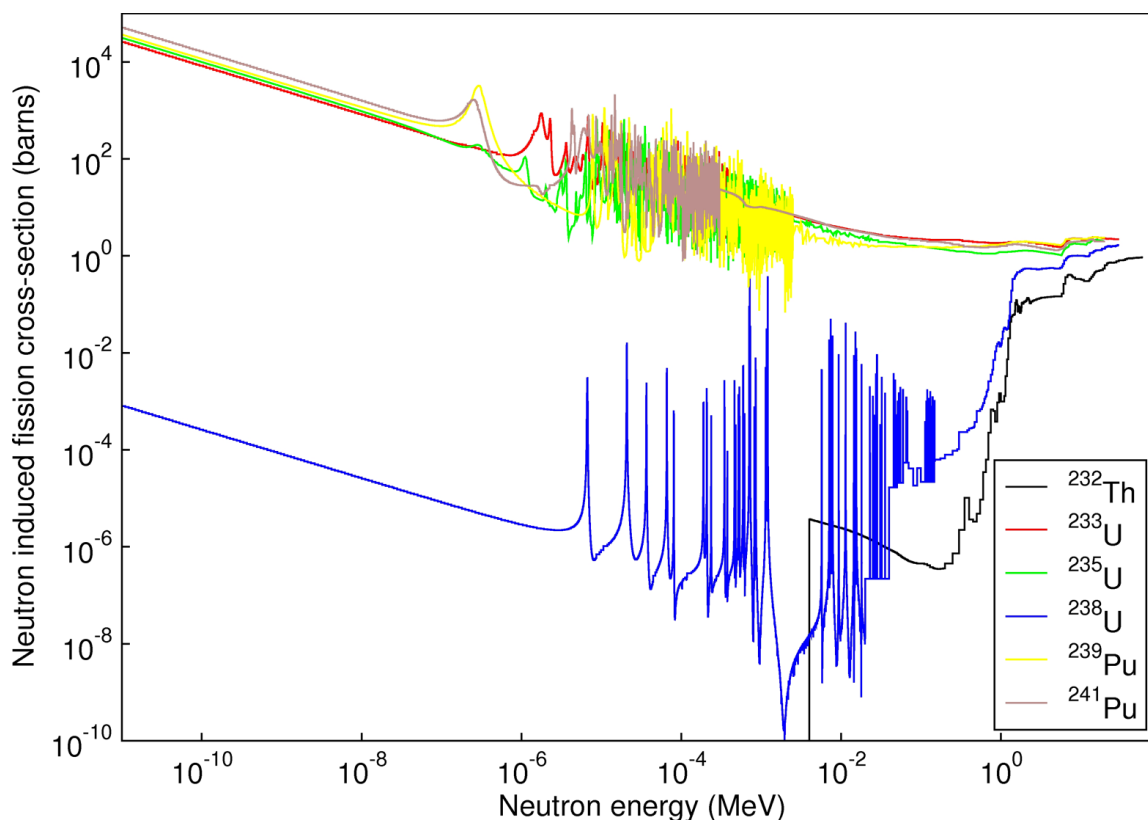


Figure 8: Neutron induced fission cross sections for major actinides

The thermal neutron cross-sections for the actinides are tabulated at <https://www-nds.iaea.org/sgnucdat/a5.htm>. For the actinides of most interest here those figures are provided in Table 2. Note that U-238, Pu-240, Np-237 are all suppressed by many orders of magnitude.

Table 2: Thermal neutron cross sections for actinides of interest (from <https://www-nds.iaea.org/sgnucdat/a5.htm>)

Nuclide	Fission cross section (barns)
U-235	582.6±1.1
Pu-239	748.1±2.0
Pu-241	1011.1±6.2
Am-241	3.20±0.09
Cm-242	<5
Cm-244	1.04±0.20

7.1 Spontaneous fission neutrons

Most of the actinides present in the spent fuel will undergo spontaneous fission, but the dominant nuclides in spent fuel are Cm-242 and Cm-244. Cm-242 has a half-life of 162.8 days so it is only relevant at short cooling times. In this paper we are focusing on cooling times >2 years where the Cm-244 will dominate the spontaneous fission neutron production. Cm-244 has a half-life of 18.2 years and activity of 4.44×10^{12} Bq/gram Cm-244 with a spontaneous fission branching ratio of 1.371×10^{-6} leading to a spontaneous fission rate of 6.09×10^6 s/gram Cm-244. The spontaneous neutron fission yield for Cm-244 is 2.691 ± 0.012 n/fission so the spontaneous neutron production from Cm-244 is 1.64×10^7 n/s/gram Cm-244. Typical PWR spent fuel contains 20-50 g of Cm per 500 kgU (one assembly) resulting in a total neutron rate from spontaneous fission of $3\text{-}8 \times 10^8$ n/s/assembly. Note that the Pu-240 spontaneous fission rate is of order 1000 times lower than the Cm-244.

7.2 Neutrons from (α ,n) reactions

The actinides have near 100% branching fractions for alpha decay. The oxide fuel is a plentiful source of oxygen so (α ,n) reactions on the oxygen are the primary source. These are less than 10% of the passive neutrons from spent fuel as shown in Figure 9.

TABLE I. Neutron Sources in Spent Fuels^a						
Neutron Sources		Light Water Reactor (LWR)			LMFBR^c	
		UOX^b 20 GWd/tu	UOX^c 45 GWd/tu	MOX^d 50 GWd/tu	CORE +AX.BLKT 80 GWd/tu^f	RAD. BLKT 5 GWd/tu
Alpha,n	Total	6.6%	1.9%	1.3%	5.2%	57.2%
	²³⁸ Pu	2.3	0.8	0.4	2.2	4.7
	²³⁹ Pu	0.7	0.0	0.0	0.5	43.6
	²⁴⁰ Pu	0.8	0.1	0.0	0.7	7.8
	²⁴¹ Am	2.1	0.2	0.1	1.1	1.1
	²⁴² Cm	0.0	0.0	0.0	0.0	0.0
	²⁴⁴ Cm	0.7	0.8	0.8	0.7	0.0
Spontaneous Fission	Total	93.3%	98.1%	98.7%	94.8%	42.7%
	²³⁸ U	0.0	0.0	0.0	0.0	0.4
	²³⁸ Pu	0.4	0.0	0.1	0.4	0.8
	²⁴⁰ Pu	4.1	0.5	0.2	3.7	41.3
	²⁴² Pu	0.9	0.2	0.1	1.1	0.0
	²⁴² Cm	0.2	0.0	0.0	0.2	0.0
	²⁴⁴ Cm	87.6	96.8	97.6	89.4	0.2
	²⁴⁶ Cm	0.1	0.6	0.6	0.0	0.0
Total		100	100	100	100	100

^aAfter a 5-year cooling time. Calculated by ORIGEN2.
^bInitial uranium enrichment = 3.0 wt%.
^cInitial uranium enrichment = 4.1 wt%.
^dInitial fissile enrichment = 6 wt%. Recycled plutonium from an LWR with a burnup of 30 GWd/tu is used.
^ePlutonium enrichment of core is 16 wt%. AX.BLKT=axial blanket, RAD.BLKT=radial blanket. Blankets are depleted uranium.
^fThis value is the burnup of CORE. The burnup of AX.BLKT is 5 GWd/t. A core fuel assembly consists of CORE (60%) and AX.BLKT (40%).

Figure 9: Excerpt for LANL note LA-12774-MS
(http://inis.iaea.org/search/search.aspx?orig_q=RN:25070652)

7.3 Calculating the number of fissions from the burnup of the fuel

1 fission is $\sim 3.1 \times 10^{-11}$ W-s and 1 GWd = 8.64×10^{13} W-s = 2.8×10^{24} fissions.

1 metric tonne of U-238 is 2.53×10^{27} atoms, so total available energy for full burnup of the fuel is ~ 900 GWd/tU and 30 GWd/tU is about 3% burnup, 45 GWd/tU is 5% burnup.

A PWR assembly is ~ 500 kgU (1/2 tU) so at 30 GWd/tU burnup the number of fissions is 4.2×10^{25} . There are 264 pins in the assembly, so each pin will have, on average, 1.6×10^{23} fissions. PWR rods have fuel OD of 8.19 mm, cladding OD of 9.50 mm and cladding thickness of 0.573 mm. The linear density is 6.828 g/cm with 5.774 g/cm (85%) UO₂ and the remaining mass in the Zircalloy cladding. A typical 17×17 assembly contains 264 fuel rods and 25 instrumentation/control rod positions.

BWR assemblies are not as standardized as PWR assemblies. Assemblies range from 6×6 to 10×10 arrays of rods. The rod sizes are also not standardized. One example set of parameters is for the General Electric 7×7 design. Rods for that assembly have OD 12.395 mm, cladding OD 14.30 mm and cladding thickness 0.8128 mm. The linear density is 14.39 g/cm with 13.23 g/cm (92%) UO₂ and the remaining mass in the Zircalloy cladding¹. Another BWR rod design has an OD of 9.55 mm, cladding OD 11.18 mm and cladding thickness 0.71 mm packaged as an 8x8 array. The linear density is 9.475 g/cm with 7.851 g/cm (83%) UO₂ and the remaining mass in the Zircalloy cladding. For a representative 8×8 assembly, of the 64 positions, 54 are full-length fuel pins, 8 are short pins and 2 water rods.

7.4 Fission neutron induced fission

The thermal neutrons used to interrogate the fuel will produce fission neutrons - 2.46 neutrons per thermal fission on U-235. At these energies U-238 will undergo fission with a cross section of 0.307 barn. The other dominant actinides in the spent fuel have cross sections of 2 bars or less for fission spectrum neutrons, but their abundances are <1% of that of U-238 so the U-238 fission will be the dominant contribution of fission neutron induced fission.

238	Neptunium	6.23E-11	1.58E+11	
237	Neptunium	6.20E-01	1.57E+21	
246	Curium	1.30E-04	3.19E+17	
245	Curium	1.17E-03	2.87E+18	
244	Curium	1.44E-02	3.56E+19	1.04
243	Curium	1.50E-04	3.72E+17	617
242	Curium	8.95E-07	2.23E+15	<5
243	Americium	1.34E-01	3.31E+20	0.1983
242	Americium	3.43E-04	8.52E+17	2100
241	Americium	1.10E+00	2.75E+21	3.20

¹ MOL.19971229 .0128; <http://pbdupws.nrc.gov/docs/ML0334/ML033450354.pdf>



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