1 Direct Measurement of ^{235}U in Spent Fuel Rods with 2 Gamma-ray Mirrors

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8 Abstract

Direct measurement of plutonium and uranium X-rays and gamma-rays is a highly desirable non-destructive analysis method for use in reprocessing fuel environments. The high background and intense radiation from spent fuel make direct measurements difficult to implement since the relatively low activity of uranium and plutonium are masked by the high activity from fission products. To overcome this problem, we make use of a grazing incidence optic to selectively reflect K_α and K_β fluorescence of Special Nuclear Materials (SNM) into a high-purity position-sensitive germanium detector and obtain their relative ratios.

9 *Keywords:* Fission; Spent fuel; Non-destructive analysis; X-ray fluorescence;
10 Uranium quantification; Gamma-ray mirrors; Optics

11 1. Introduction

12 A recent study [1] has suggested that one of the gaps to be addressed for
13 safeguards in reprocessing plants is the direct measurement of the fissile con-
14 tent of spent fuel being introduced to the facility. The safeguards approach for
15 reprocessing facilities generally uses a “shipper-receiver difference” evaluation
16 that compares the quantity input in the spent fuel assemblies to that mea-
17 sured in the input accountability vessel and the head-end waste streams. The
18 fuel assembly input quantity has traditionally been based on reactor calculated
19 quantities (power history profile). A number of measurement technologies are

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20 being investigated to improve on the accuracy of these estimates and among
21 those, direct gamma-ray measurements show significant promise. However, this
22 measurement technique is time-consuming because the overall count rate at the
23 detector must typically be kept below 1 MHz and only a small fraction of the
24 counts originate from the species of interest. This poses a significant challenge
25 for transition to a production environment where dwell times must be kept short.
26 A grazing incidence optic, the subject of this paper, can dramatically improve
27 the performance of the direct gamma-ray measurement technique by allowing
28 only radiation in narrow energy bands of interest to reach the detector.

29 **2. Approach**

30 The optic is based on a grazing incidence mirror technology developed for as-
31 trophysical observations. The last fifteen years have seen the rapid development
32 of focusing hard X-ray optics for a variety of applications, including astronomy
33 [2, 3], medical imaging [4, 5] and as beamline components [6–9]. The astro-
34 physics community has spurred much of the development, and the maturity of
35 the field is evident by current space missions such as NuSTAR and ASTRO-H
36 [10, 11]. Advances in coating technologies have resulted in the demonstration
37 of efficient, narrow band (~ 2 keV FWHM) reflectors at energies above 100 keV
38 [12–14]. To advance the technology for these optics to the point where they will
39 be effective at energies between 95 and 300 keV, relevant for measuring ^{235}U
40 and ^{239}Pu contents, a series of basic science experiments have been performed,
41 including the verification that optical constants are consistent with those tabu-
42 lated using photon-interaction cross-sections for multilayer materials up to 300
43 keV and the experimental validation of the performance (peak energy reflectivity
44 and width of energy band-pass) of the multilayer coated mirrors.

45 The relevance of this system for proliferation detection is that it enables the
46 quick and accurate assaying of SNM in each fuel rod at the entry point of a
47 reprocessing facility. The envisioned concept of operation for this technology
48 would have a single fuel rod pass by a series of multilayer-coated mirrors viewed

Figure 1: Illustration of multilayer design: in a constant d -spacing design, the thickness of the alternating layers of high- and low-density materials are the same throughout the coating. This maximizes reflectivity for a unique combination of wavelength (λ) and grazing incidence angle (θ).

by a commercially available position-sensitive High Purity Germanium (HPGe) detector.

The optical element is a stack of bi-layers, where each bi-layer is made of a low- and high-density material with different indexes of refraction. The interfaces at the different material systems result in constructive interference that follows Bragg's law, that for very shallow grazing angles can be written as

$$m\lambda = 2d \sin \theta \simeq 2d\theta, \quad (1)$$

where λ is the wavelength of the photon, θ the scattering angle, d the bi-layer period and m is an integer determined by the order of reflection.

An illustration of a multilayer design with constant- d spacing tuned to work for a specific angle and energy ($1/\lambda$) is shown in figure 1. As seen from equation 1, fabricating coatings with a thin bi-layer period of 1 – 2 nm, makes it possible to work at shallow, but practical, grazing angles of a few to several mrad.

3. Experimental setup

3.1. Optics

For the current experiment, we built an optic consisting of a stack of five flat, $100 \times 100 \text{ mm}^2$ reflective mirrors. Each mirror consists of a 0.5 mm thick silicon substrate, cleaved from a single standard 200 mm diameter silicon wafer,

Figure 2: Picture of the X-ray mirror stack.

66 deposited with a WC/SiC multilayer with 300 bi-layers with a nominal period
 67 of 1.5 nm. The mirrors were coated, via magnetron sputtering, and assembled
 68 into the optic at Lawrence Livermore National Laboratory. For the safeguards
 69 application, the aim is to collect the radiation from a given length of the fuel
 70 rod and direct it towards a two dimensional position-sensitive detector. As the
 71 fuel rod essentially corresponds to a line source, the optical design of choice is
 72 a single reflection concentrator (see figure 2). In addition to the design of the
 73 optic, a new hot-cell port has been implemented. The design provides the optic
 74 with a view of the fuel rod while shielding the detector from direct exposure. It
 75 further contains elements to minimize radiation scattered off of the sides of the
 76 hot-cell port reaching the detector.

Figure 3: End view of the 30'' long collimator installed in the hot-cell wall (between the fuel
 rod and the Compton imaging HPGe detector). The opening to the fuel rod is 5 mm high and
 38 mm wide. See also figure 4.

Each mirror has been optimized for an energy band-pass associated with the emission lines from uranium and plutonium including the characteristic K_α and K_β lines from each element and isotope-specific lines from special nuclear material (SNM, U-235 and P-239). The detector is shielded from the direct line-of-sight of the fuel rod, effectively enhancing the detection of radiation reflected by the mirrors. Only a small fraction of the photons from the rod are in the energy band-pass of the mirror; therefore, a larger field-of-view can be used allowing for more photons within the energy range of interest to be detected in the same amount of integration time.

The narrow band feature of the designed optic enables it to significantly reduce gamma-ray background from fission products. The reduction occurs through suppression of out-of-band flux and limiting subsequent down-scattered photons in the detector. This greatly relaxes performance requirements for the detector setup since it will be submitted to much lower event rates.

3.2. *HPGe detector*

A position-sensitive, 10 mm thick and 90 mm diameter, mechanically cooled HPGe detector (PHDS GeGI [15]) with 160×160 pixels was mounted across the room from the hot-cell port. This detector was used because its energy resolution is better than the energy band-pass of the mirror, allowing to determine both fluorescence and background continuum contributions. Alignment between the germanium detector and the fuel rod was performed using laser sighting between the detector face and the fuel rod center.

The X-ray optic and the germanium detector were able to monitor a large field-of-view of the fuel rod and enable the direct determination of the fluorescence lines present in the spent nuclear fuel without exceeding the count-rate capabilities of the germanium detector.

3.3. *Tandem system and measurement protocols*

The two subsystems, the X-ray optic and the HPGe detector, have been successfully integrated and used for a detailed laboratory-based campaign conducted at the Irradiated Fuels Examination Laboratory (IFEL) at Oak Ridge

Figure 4: Schematic of the hot-cell port of the experiment. Implementation of the mirror assembly and collimator.

107 National Laboratory. The collimator was designed and fabricated at Oak Ridge.
 108 The mirror stack was mounted downstream from the 30'' lead collimator with an
 109 opening of $5\text{ mm} \times 38\text{ mm}$ (see figure 3) centered on the fuel rod. The collimator
 110 and mirror assembly was installed in the hot-cell port.

111 Two spent fuel rod samples, TMI AG616E and MOX 605B, were investigated
 112 by using different grazing angles between the optic and the collimator. The
 113 measurement procedure consisted of acquiring data for 14400 seconds at a fixed
 114 grazing angle. Measurements were acquired over angles ranging from 2.2 to
 115 7.9 mrad, in increments of 0.2 mrad. Figure 5 shows a sketch with the relevant
 116 parameters of our setup. The best results were seen for grazing angles that
 117 maximized the characteristic emission lines (95-120 keV), which from Equation
 118 1, corresponds to grazing angles of 3.4-4.4 mrad.

Figure 5: Cartoon of the sample-optic-detector setup for a θ grazing angle.

Figure 6: Rate in Hz/mm^2 observed on the full face of the detector with the optic inserted as described in figure 4. The counts in the x -columns from pixels 65 to 80 correspond to the image of reflected lines of the fuel rod on the HPGe detector. Notice how the overall rate at the energy range of the Uranium fluorescence is negligible on the rest of the detector compared to the image of the fuel rod created by the optic.

119 4. Performance

120 The energy bandwidth of the optic is such that at a fixed grazing angle all
 121 K_α and K_β lines of interest, are reflected onto the detector. Photons of different
 122 energies are specularly reflected at angles given by equation 1. In practice
 123 this means that the collimated beam is dispersed over $\sim 9\text{ mm}$ in the x -axis,
 124 with the lower energy photons scattering the most into low x -axis locations and
 125 oppositely so for high energy photons. The significance of the background was
 126 measured at the different angle setups of the mirror in absence of the fuel rod
 127 samples and its contribution has been normalized and subtracted from the fuel
 128 rod measurements, allowing us to accurately determine the relative intensity of
 129 individual fluorescence lines.

130 The location of the collimator image in the 2D HPGe detector it is estab-
 131 lished by the x -columns showing any fluorescence line activity above the mea-
 132 sured background and its position shifts along the x -axis for different grazing
 133 angle setups. A direct view of the collimator, in absence of the X-ray optic,
 134 appears between the x -columns 95 and 160 of our detector.

135 Figure 6 shows the resulting collimator image at a given grazing angle. Hot

spots are visible between the x -columns 65 and 80 a 8.3 mm deflection, in accordance with our expectations from Bragg's law. The multilayer coated surface of the mirror assembly, and in turn the collimator image, is co-parallel to the y -axis of the detector.

Figure 7 shows the location of the maximum rate for a given fluorescence as a function of its energy, corroborating a linear dependence as expected from Bragg's law.

The overall spectra observed in the HPGe detector downstream of the collimator are shown in figure 8, where the red line represents the spectrum registered at the detector without shielding and without X-ray optic. Note how the rate decreases almost two orders of magnitude due to the combination of the optic response and four inches of lead shielding for the direct light (blue line). Selecting only the pixels on the detector in which the reflected beam impinges, one notes more prominent spectral features with the energy band pass of the optic (green line). Subtracting background further enhances the spectral contribution of the different fluorescence (black line).

The ^{137}Cs line (662 keV) and the resulting Compton edge at 470 keV are clearly visible, even after background subtraction, which suggests that there is still some direct light from the fuel rod arriving at the surface of the HPGe detector. Once our setup was fully assembled and the background was subtracted, the mean rates at the fluorescence energies became ~ 1 Hz.

Figure 7: Evolution of the maximum intensity position of the $K_{\alpha 1}$, $K_{\alpha 2}$, $K_{\beta 1} + K_{\beta 3}$ and $K_{\beta 2}$ emission lines of ^{235}U (squares) and best fit to the data points (dashed blue line).

Figure 8: Overall spectra on the full face of the HPGe detector (90mm diameter) with (blue) and without the X-ray band pass optic (red). Also, the integrated spectrum for the concentrated image of the collimator ($8.3 \times 90 \text{ mm}^2$) as in figure 6 before (green) and after (black) background subtraction. The shadowed region between 70 and 120 keV corresponds to the band pass energy regime of the optic.

157 Using the above data, we have been able to quantify the K_α and K_β branch-
 158 ing ratios of uranium from the TMI AG616E and MOX 605B samples (see figures
 159 9 and 10 and table 1). Effects of attenuation from a sapphire window encasing
 160 the fuel rod have been considered negligible for the calculation of the branching

Figure 9: Integrated spectrum along the y -axis for the dx -axis portion of the detector containing the most intense collimator image ($8.3 \times 90 \text{ mm}^2$). In orange, the K_α and K_β emission lines of uranium for TMI AG616E before background subtraction. In black, the spectrum remaining after background subtraction. In this figure, a gaussian fit to $K_{\alpha 1}$, $K_{\alpha 2}$, $K_{\beta 1} + K_{\beta 3}$ and $K_{\beta 2}$ emission are plotted in red, blue, violet and green respectively.

Line	Energy [keV]	Expected [%]	Measured TMI / MOX [%]	Deviation TMI / MOX [%]
$K_{\alpha 1}$	98.44	100	100 / 100	0 / 0
$K_{\alpha 2}$	94.67	61.9	62.2 / 62.7	0.3 / 0.8
$K_{\beta 1} + K_{\beta 3}$	110.99	33.6	34.4 / 34.3	0.8 / 0.7
$K_{\beta 2}$	114.50	12.3	11.3 / 11.3	1.0 / 1.0

Table 1: Expected and measured K_{α} and K_{β} branching ratios of uranium for samples TMIAG616E and MOX605B for the positional maximum intensity of the concentrated ($15 \times 160 \text{ mm}^2$) collimator image shown in figure 6.

ratios, since its contribution, $\sim 3.4\%$ of extra attenuation for the higher fluorescence lines is already taken into account by the background measurements.

5. Conclusion

The results demonstrate the feasibility of using X-ray optics as a band pass filter to enable the direct measurement of SNM. Two fuel rods, TMIAG616E and MOX605B, were investigated. The data show compelling results for the quantification of the K_{α} and K_{β} Uranium lines and will be used to improve

Figure 10: Relative intensities of the different Uranium fluorescence lines for optimum tilting of the X-ray optic. In red the $K_{\alpha 1}$ intensity, in blue the $K_{\alpha 2}$ line, in black the $K_{\beta 1} + K_{\beta 3}$ intensities and in green the $K_{\beta 2}$ contribution.

our modeling capability, and to optimize future measurement campaigns and prototypes. A model that takes into account the branching ratios for the K_α and K_β emission lines has been developed and the overall performance of the experimental setup has been presented in this article.

The analysis of the data shows that the X-ray mirrors perform as expected and validate the concept of their use to assay spent nuclear fuel. The spectral response is broad enough to cover the energy range of interest and narrow enough to suppress both high and low energy background contributions. We have further demonstrated the ability to reconstruct the branching ratios for the ^{235}U fluorescence lines in the reflected spectrum to better than 1 % indicating the quantitative performance of the technique.

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