U and Pu Gamma-Ray Measurements of Spent Fuel Using a Gamma-Ray Mirror Band-Pass Filter


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Abstract. We report on the use of grazing incidence gamma-ray mirrors to serve as a narrow band-pass filter for advanced non-destructive analysis (NDA) of spent nuclear fuel. The purpose of the mirrors is to limit the radiation reaching a HPGe detector to narrow spectral bands around characteristic emission lines from fissile isotopes in the fuel. This overcomes the normal rate issues when performing gamma-ray NDA measurements. In a proof-of-concept experiment, a set of simple flat gamma-ray mirrors were used to directly observe the atomic fluorescence lines from U and Pu from spent fuel pins with the detector located in a ‘shirt-sleeve’ environment. The mirrors, consisting of highly polished silicon substrates deposited with WC/SiC multilayer coatings, successfully deflected the lines of interest while the intense primary radiation beam from the fuel was blocked by a lead beam stop. The gamma-ray multilayer coatings that make the mirrors work at the gamma-ray energies used here (~ 100 keV) have been experimentally tested at energies as high as 645 keV, indicating that direct observation of nuclear emission lines from $^{239}\text{Pu}$ should be possible with an appropriately designed optic and shielding configuration.

1. Introduction

Spent nuclear fuel represents an extreme example of difficulties encountered with measuring gamma spectra in a very high background environment. Typically, gamma-ray spectroscopy is used to measure $^{137}\text{Cs}$ activity or ratios of fission product activities to verify the cooling time and burnup of the fuel. However, the gamma-emissions from spent fuel are dominated by large Compton backgrounds from scattering within the fuel, as well as interfering gamma peaks from isotopes that are not of interest. To detect the lines of interest, one must reduce the flux from the fuel so that the overall count rate at a spectrally capable detector [such as high purity germanium (HPGe)] is reasonable. While simple collimation is one approach to limit the flux, the orders-of-magnitude flux reduction required at the detector means that count times in the regions of interest would be prohibitive. To overcome this limit, we are applying gamma-ray mirror technology to act as a narrow band-pass filter, allowing only spectral windows around the lines of interest to reach the detector while rejecting other gamma-ray energies. In this way the overall flux at the detector can be reduced, while the solid angle of the detector to the fuel rod is not restricted by collimation. To make this work requires optics that can achieve high reflectivity in a tailored pass band that can also focus the gamma-rays from the fuel rod onto the detector.

1.2 Gamma Ray Optics

The optics used here rely on the latest advances in grazing-incidence, high-energy optics under development in a number of fields. Such instruments were originally developed for use at x-ray energies based on the observation that high-energy photons undergo total external reflection from a smooth surface of high-Z material at shallow angles because the index of refraction at high energies is less than one [1]. In the early 1950’s, Wolter recognized that pairs of mirrors could be used to focus the radiation to a point over a wide field of view if the mirrors were built from surfaces of revolution of conic sections (e.g. hyperboloids and ellipsoids) [2]. Although Wolter’s original designs were for use in biology, the first realization of working instruments was as X-ray telescopes for high energy astrophysics in the late 1960’s [3]. The astronomy community also advanced on Wolter’s design by recognizing that the collection efficiency of the grazing incidence optics could be increased by orders of magnitude if a series of nested confocal mirrors were used. More recently, near-field optics suitable for the applications originally envisioned by Wolter allow imaging objects at distances of order meters have been developed [4].
The progressively smaller graze angles required to achieve total external reflection at higher photon energies limited the response of the first generation x-ray telescopes to 10 keV and below. Such instruments also provided no selectivity since they reflect all energies below the cut off. As such, they were unsuitable for safeguards applications. However, the latest technological advance solves both of these problems simultaneously. In 1991, Christensen, et al. [5] proposed that coatings of alternating high and low index materials could be used to extend the useful reflectivity of grazing incidence mirrors to higher energies. The multilayers act as an artificial crystal, reflecting only radiation that satisfies Bragg’s law:

$$2d \sin \theta \approx n \lambda,$$

where $d$ is the bilayer pair spacing, $\theta$ is the graze angle, $n$ is the order (here set to one), and $\lambda$ is the wavelength. The expression is not exact in that there are material-dependent terms that modify the response. This provides a limited (narrow) band pass for the mirror. However, the band pass can be extended by taking advantage of the many bilayer pairs needed due to the penetrating power of the gamma-rays. By varying the bi-layer spacing as a function of depth in the multilayer structure, one can tailor the overall angular/energy response. An astrophysical instrument with a response to 79 keV has been successfully placed in orbit [6], and more recently, sample surfaces have shown significant reflectivity out to 625 keV [7, 8]. In this paper we report on the first use of simple optics with multilayer coatings to observe the K-shell fluorescence of uranium and plutonium from spent nuclear fuel.

2. Experimental Design

In a proof-of-concept experiment, a stack of five planar gamma-ray mirrors (10 x 10 cm²) were placed at the exit of a hot-cell port to deflect the atomic K-fluorescence lines from a spent fuel pin onto a position-sensitive gamma-ray detector. A conceptual schematic of the experiment is shown in Fig. 1. Exposure of the mirror stack to radiation from the spent fuel was limited by a collimator with a cross section comparable to the height of the mirror stack and a width defined by the sapphire window sealing the end of the collimator from the hot cell interior. A position-sensitive HPGe detector was placed far enough from the mirrors so that the mirrors achieved a ~ 3 cm deflection of the radiation of interest. This was sufficient that a Pb beam stop could be used to keep direct radiation from the fuel pin from reaching the detector. The different components of the setup are described in more detail below.

2.1. Facility

The experiments were conducted at the Irradiated Fuel Examination Laboratory (IFEL) at Oak Ridge National Laboratory. Inside a large hot cell, fuel pin segments were positioned in a transport mechanism that moves the pins along their length in front of a 10-cm-diameter access port. A 109-cm-long lead collimator with a cross section of 6.98 x 0.50 cm² was placed in the port to limit the radiation reaching the mirror stack to the height of the stack. The width of the collimator was designed to match the diameter of a sapphire window used as an environmental barrier between the port and the inside of the hot cell. The collimator’s inside surface was coated with silver paint to minimize K-shell fluorescence from the Pb surface, since this surface was directly visible to the HPGe detector used for the measurements. The fuel rod was an additional 20 cm beyond the end of the collimator. The detector side of the port opens to a shirt-sleeve environment. The detector was placed near the far wall from the...
hot cell, providing a total distance of 3.5 m between the mirrors and the detector surface. This length was required to provide good separation of the reflected radiation from the straight-through beam.

2.2. Mirror Stack/Manipulator

The optics were fabricated from alternating layers of SiC and WC sputtered onto highly polished, 10 × 10 cm² Si substrates. The period of the layer pairs was nominally 1.5 nm (1.506 to 1.514 nm) with the thickness of the WC layer 45% of the overall bilayer thickness. A total of 300 bilayer pairs were deposited on each of five mirrors that were arranged in a stack. For this experiment, no deliberate variation in the layer-pair spacing was used. Although this means that the angle/energy response of each individual mirror is narrow, the overall energy response of the mirror system is determined by the complex geometry of the overall configuration. This includes the fact that multiple mirrors are viewing the rod, with each mirror seeing a range of angles on the rod. In addition, each region of the rod is a broadband gamma-ray source. Ray trace calculations are in progress to determine the overall system response. However, the results obtained from the fuel rod indicate an energy width of order 20 keV.

The mirror stack (Fig. 2) was built on a precision-lapped, thick, structural block. The mirrors themselves were separated from each other using graphite spacers placed at the edges of the mirrors. Spring loading was used to maintain a positive force on the stack. A 3-point kinematic mount was fabricated in the bottom of the mirror mount and this was used to mount the stack to a manipulator designed to provide repeatable angular rotations with fine resolution (Fig. 2). The manipulator comprises a front torsional bearing to allow rotation only perpendicular to the deflection axis. A linear actuator is used to drive a precision wedge with a 3° pitch between the bottom of the mirror holder and the bottom surface of the manipulator. The system was able to reproduce vertical angular displacements as small as 0.005°, over a range of angles from ~ 0.15° to 0.45°.

For the 1.5 nm bilayer pair spacing the Bragg condition is satisfied at 100 keV at ~ 0.24°. Over the 3.5 m distance to the detector this amounts to a deflection of ~ 3 cm. Note that while the angle reproducibility is very good, the absolute angle of the mirrors with respect to the fuel is not well known. This is both due to uncertainties of the system geometry as defined by the hot cell wall and fuel pin manipulator, and the fact that the stage did not go all of the way to 0°. Overall, coarse alignment was easily established and was sufficient to get results with the system.

2.3. Detector

Data were collected using a mechanically cooled, double-sided-strip, HPGe detector manufactured by PHDS Co. [9]. The detector itself is 1 cm thick and has an active diameter of 8.6 cm. Charge is collected by strips that have a 5-mm pitch, with the strips on opposite faces of the detector orthogonal to each other. Approximate event locations are determined by the strips that collect the electrons and holes generated by the gamma-ray interaction within the germanium. The system electronics also record the transient signals seen by the strips adjacent to those that collect the charge. By comparing the size of the signals seen by these spectator electrodes, the event location is refined to obtain a spatial resolution of ~ 1 mm. Only single-site interactions were included in the analysis. Data acquisition was controlled from a laptop computer that also controlled the angle of the mirror stack. Events were saved to a list-mode data file for later analysis and displayed in real time on a 2D plot of the detector face. Events for display could be selected based on both location and energy, allowing one to obtain spectra from different regions of the detector.
Table 1. Fuel properties

<table>
<thead>
<tr>
<th>Rod</th>
<th>$^{235}$U/U (%)</th>
<th>Pu/(Pu+U) (%)</th>
<th>Burn Up GWD/MTU</th>
<th>Time since Discharge</th>
</tr>
</thead>
<tbody>
<tr>
<td>Three Mile Island (TMI)</td>
<td>0.57</td>
<td>1.3</td>
<td>50.9</td>
<td>17 years</td>
</tr>
<tr>
<td>MOX</td>
<td>0.25</td>
<td>4.9</td>
<td>47.3</td>
<td>62 months</td>
</tr>
</tbody>
</table>

3. Measurements

Measurements were made on two different fuel rods in one of two modes: a rocking-curve mode in which the mirror assembly was stepped through a series of angles, with data collected for a fixed time at each angle; and a long-dwell mode where the mirror was fixed at a set angle and data collected for the entire time at that angle. Typical rocking-curve dwell times were one hour per step while the long dwell modes ran from overnight to 65 hours. In addition, a short, direct exposure was obtained on one of the rods with the lead beam stop normally used to block the direct beam removed. The properties of the two fuel rods are summarized in Table 1. The first rod was a section of Three Mile Island (TMI) fuel that had undergone 50.9 MWD/MTU (MegaWatt Days/Metric Tons Uranium) in the reactor and had subsequently cooled for 17 years. It is primarily U with 1.3% Pu. The second rod was metal oxide fuel (MOX) containing 4.9% Pu. It had been discharged only 5 years before the measurement. The background rate observed with the detector when no fuel was present was 525 Hz.

4. Results

The results obtained with the different rods are qualitatively very similar and are selectively presented to illustrate the overall features of the measurement approach. Differences are pointed out where they are thought to be significant.

4.2 Bare Fuel

The results of a 100-s exposure to the TMI fuel without the lead beam stop are shown in Fig. 3. The estimated count rate on the detector face was 21 kHz, which was enough to paralyze the display software while the data were being collected. In fact, the overall count rate is only an estimate since the detector was stopped manually, and the response was sluggish. The dead time of the overall system at this rate is not known, so this represents a lower bound on the flux. Nevertheless a viable spectrum was obtained. It is clearly dominated by the 662-keV emissions from $^{137}$Cs. The K-fluorescence lines from the uranium are also visible. For comparison the spectrum with the beam stop in place is also shown. Note that the fluorescence lines are missing from that spectrum. The overall count rate with the beam stop was 574 Hz representing a 36-fold reduction in rate. The figure also shows intensity maps of the detector face. The direct beam only strikes the lower portion of the detector and this region was blocked with 5 cm of lead during the runs using the mirrors. Note also that these data were collected with the mirror in and at its shallowest angle (~ 0.15°).

Fig. 3. Left (right) shows the intensity distribution on the detector face for the direct (deflected including beam stop) beam. (Center) The spectra for the two different cases are shown normalized to equivalent acquisition times. The regular square array on the detector face is due to the reduced efficiency between the electrodes of the detector; they have a 5 mm pitch. The gray-scale of the images are each scaled independently.
4.3 Rocking Curve Measurements

Sample results of a rocking curve measurement obtained with the MOX rod are shown in Fig. 4. The figure shows the full face of the detector obtained with a tight spectral cut on the U Kα lines. As the mirror angle is increased, the amount the spectral lines are deflected increases as well. This is visible as the vertical shift of the dark band (higher intensity) region on the detector. By restricting the data that are analyzed to a height commensurate with the height of the line (rectangle on the left image), we create an artificial slit that can be used the same way a real slit would be in a standard rocking-curve dispersive measurement (Fig. 5).

A plot of the counts in a spectral band centered on a peak and obtained in the region of the detector defined by the virtual slit provides a classic rocking curve response (Fig. 5). In such a system the dispersive nature of the measurement is evident from the fact that the peak in counts from different energy lines occur at different angles.

While the rocking curves are informative, we obtain enhanced results using the full spatial and spectral nature of the detector. This is illustrated in Fig. 6 which shows the spectra obtained in the virtual slit for each of the detector faces shown in Fig. 4. As the region of enhanced transmission enters the slit, first the Kα and then the Kβ lines show up in the spectra. By selecting the appropriate slit height we can optimize the SNR for a given line.

4.4 K-shell emission rates

The quantitative nature of the results are highlighted by performing a detailed analysis of the relative areas of the K-shell lines from both the TMI and MOX rods. Details of this work are reported elsewhere [10] and summarized in Table 2. In short, the data from a virtual slit encompassing the lines of

![Fig. 5. Rocking curve obtained from the MOX rod. To generate each curve the number of counts obtained in a spectral band centered on the Kα (93-100.5 keV) and Kβ (109-119 keV) lines in a fixed virtual slit (4.8 mm high) on the detector are recorded as a function of the mirror angle. Errors are less than the height of the plot markers.](image)
Fig. 6. Spectra obtained from the spatial region given by the rectangle in Fig. 4. The left plot shows the spectra from the left two images of Fig. 4, while the right plot shows the right two. Note that the atomic lines only appear when the dark band falls within the virtual slit defined by the rectangle.

interest are analyzed by removing an estimate of the background taken from an exponential fit to the background above the line energies. The lines are then each fit to a Gaussian shape and the area of the line calculated from the fit results. When compared to the expected line ratios from x-ray fluorescence, the line areas are found to agree to ~ 1%.

Table 2. Relative line intensities

<table>
<thead>
<tr>
<th>Uranium Line</th>
<th>Energy (keV)</th>
<th>Expected Intensity (%)</th>
<th>Measured (%) TMI</th>
<th>Measured (%) MOX</th>
<th>Difference (%) TMI</th>
<th>Difference (%) MOX</th>
</tr>
</thead>
<tbody>
<tr>
<td>$K_{\alpha1}$</td>
<td>98.4</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>$K_{\alpha2}$</td>
<td>94.7</td>
<td>61.9</td>
<td>62.2</td>
<td>62.7</td>
<td>0.3</td>
<td>0.8</td>
</tr>
<tr>
<td>$K_{\beta1} + K_{\beta3}$</td>
<td>111.0</td>
<td>33.6</td>
<td>34.4</td>
<td>34.3</td>
<td>0.8</td>
<td>0.7</td>
</tr>
<tr>
<td>$K_{\beta2}$</td>
<td>114.5</td>
<td>12.3</td>
<td>11.3</td>
<td>11.3</td>
<td>1.0</td>
<td>1.0</td>
</tr>
</tbody>
</table>

4.5 Pu $K_{\alpha}$ line detection
Both the TMI and MOX rods contain a few percent Pu and detection of this material is one of the goals of developing the band pass filter technique. The $K_{\alpha1}$ line from Pu falls at 103.7 keV between the $K_{\alpha}$ and $K_{\beta}$ lines of uranium and just below the 105.3 keV nuclear line from $^{155}$Eu. Fig. 7 shows the results from long-dwell measurements on the two rods and clearly shows a double peak structure at the correct energy for the lines.

5. Discussion
The results presented above clearly indicate that the mirror works as a band-pass filter. Further, as shown in Fig. 8, the technique clearly improves on the SNR when compared to the results obtained with the direct beam. However, the detection of the Pu line is marginal and not as clean as that ob-

Fig. 7. Spectra showing both Pu $K_{\alpha}$ radiation and a nuclear line from $^{155}$Eu. The spectrum on the left is from a 22-hour integration on the TMI rod while the one on the right is from a 65-hour integration on the MOX rod. An exponential background was subtracted from the spectrum on the right.
Fig. 8. Comparison of spectra obtained with the direct beam from the fuel rod (gray) and in a narrow slice of the detector after deflection of the beam by the mirror (blue). The spectra are each self-normalized to their respective peak intensities.

tained in the work of Charlton et al. [11] who used a carefully constructed collimator to observe the same lines. A comparison of their spectra to those obtained in this work show they had a peak to valley for the U Kα line of ~ 6.2 while our results were factor of two worse at 3.4. On the other hand, their detector was operating at dead times up to 16% with comparable count times (1 to 16 hours.) In short, those results represent the upper limit of what can be achieved with current technology, while the results presented here represent a first proof-of-concept experiment.

There are two issues that must be addressed to advance the use of gamma-ray mirrors with spent fuel. The first is to develop optics that provide a greater solid angle for the radiation. The current mirrors are simple flats that only deflect the radiation. Without focusing to gather the gamma rays, the solid angle of the detector falls off as the radius squared. Using the distance to the detector and including the 50% of the stack aperture that is blocked by the mirror substrates and the finite reflectivity of the coatings, the solid angle to a single location on the fuel is ~ 5.5 x 10^-7 Sr. More advanced optics designs can be constructed using the same techniques developed for gamma-ray astronomy [6] and medical imaging [4]. With the same number of focusing shells one could boost this by almost 3 orders of magnitude. The net signal rate in the current configuration is ~ 150 Hz so even a few shells would solve the solid angle problem. We note that the charge collection time in the current detector is ~ 100 ns and that it is segmented so that instantaneous rates up to 150 kHz should be possible without signal degradation (although the software and probably firmware would need to be upgraded.)

While it should be relatively easy to upgrade the solid angle, the issue of the noise floor remains. It is clear from the work of Charlton et al. that at least a factor of two improvement in the peak to valley should be possible. One of the primary sources of background is Compton scattering in the detector of the 662-keV line that dominates the unfiltered spectrum. While the mirrors should keep the line from hitting the detector, on examining the count distributions of the full energy spectrum on the detector face, we noticed the presence of distinct geometric structures unrelated to the lines of interest. When spectral cuts were applied to use only 662-keV radiation, the structures were enhanced (Fig. 9), overlapped with regions used to harvest the K-shell radiation, and changed as the mirror angle was changed. Hence they are due to 662-keV radiation interacting with the mirror structure. Their exact origin is currently being researched. However, by requiring two bounces (needed to increase the solid angle) they should be significantly reduced.

Fig. 9. Upper detector faces at 662 keV for angles (left to right) of 0.35°, 0.38°, 0.40°, and 0.43°.
The second technique for reducing background should be effective for the higher energy nuclear emissions from $^{239}$Pu (375 and 414 keV), that are the ultimate goal of this research. At these energies a significant fraction of the events should undergo Compton scattering in the detector and this can be used to restrict radiation to arrive only from specific directions. Through the use of Compton imaging and by rejecting single site interactions, improved SNR should be possible. The goal is to achieve a SNR that is dominated by what happens in the fuel and not the detection system.

6. Conclusion
Proof of concept experiments conducted on spent nuclear fuel with simple optics indicate that gamma-ray mirrors can be successfully applied as a band-pass filter to reduce the radiation from the fuel reaching a detector. Significant improvements in solid angle and enhanced background suppression should be possible with more advanced optics designs.

7. Acknowledgements
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8. References
9. PHDS Co., 3011 Amherst Rd, Knoxville, TN, USA.