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Direct measurement of ^{235}U in spent fuel rods with Gamma-ray mirrors

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ABSTRACT

Direct measurement of plutonium and uranium X-rays and gamma-rays is a highly desirable non-destructive analysis method for the 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 is 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.

1. Introduction

A recent study [1] has suggested that one of the gaps to be addressed for safeguards in reprocessing plants is the direct measurement of the fissile content of spent fuel being introduced to the facility. The safeguards approach for reprocessing facilities generally uses a “shipper-receiver difference” evaluation that compares the quantity input in the spent fuel assemblies to that measured in the input accountability vessel and the head-end waste streams. The fuel assembly input quantity has traditionally been based on reactor calculated quantities (power history profile). A number of measurement technologies are being investigated to improve on the accuracy of these estimates and among those, direct gamma-ray measurements show significant promise. However, this measurement technique is time-consuming because the overall count rate at the detector must typically be kept below 1 MHz and only a small fraction of the counts originate from the species of interest. This poses a significant challenge for transition to a production environment where dwell times must be kept short. A grazing incidence optic, the subject of this paper, can dramatically improve the performance of the direct gamma-ray measurement technique by allowing only radiation in narrow energy bands of interest to reach the detector.

2. Approach

The optic is based on a grazing incidence mirror technology developed for astrophysical observations. The last 15 years have seen the rapid development of focusing hard X-ray optics for a variety of applications, including astronomy [2,3], medical imaging [4,5] and as beamline components [6–9]. The astrophysics community has spurred much of the development, and the maturity of the field is evident by current space missions such as NuSTAR and ASTRO-H [10,11]. Advances in coating technologies have resulted in the demonstration of efficient, narrow band (2 keV FWHM) reflectors at energies above 100 keV [12–14]. To advance the technology for these optics to the point where they will be effective at energies between 95 and 300 keV, relevant for measuring ^{235}U and ^{239}Pu contents, a series of basic science experiments have been performed, including the verification that optical constants are consistent with those tabulated using photon-interaction cross-sections for multilayer materials up to 300 keV and the experimental validation of the performance (peak energy reflectivity and width of energy bandpass) of the multilayer coated mirrors.

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

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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

$$m\lambda = 2d\sin\theta \simeq 2\,d\theta, \tag{1}$$

where λ is the wavelength of the photon, θ the scattering angle, d the bi-layer period and m 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 Fig. 1. As seen from Eq. (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.

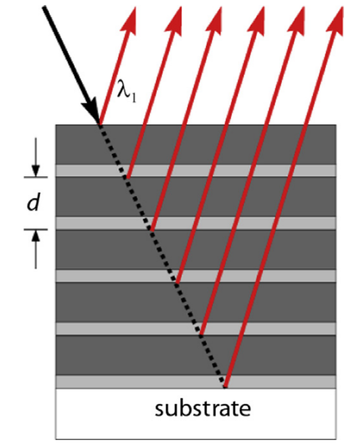

Fig. 1. Illustration of multilayer design: in a constant d-spacing design, the thickness of the alternating layers of high- and low-density materials is the same throughout the coating. This maximizes reflectivity for a unique combination of wavelength (λ) and grazing incidence angle (θ).

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

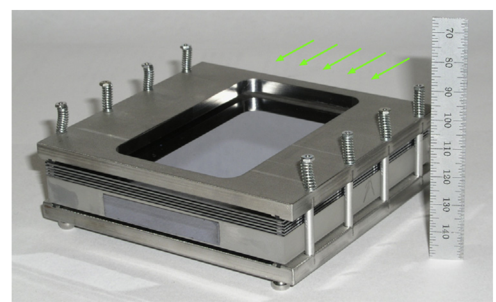

Fig. 2. Picture of the X-ray mirror stack.

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 National Laboratory. The collimator was designed and fabricated at Oak Ridge. The mirror stack was mounted downstream from the 30 in. lead collimator with an opening of 5 mm × 38 mm (see Fig. 3) centered on the fuel rod. The collimator and mirror assembly was installed in the hot-cell port.

Two spent fuel rod samples, TMI AG616E and MOX 605B, were investigated by using different grazing angles between the optic and the collimator. The measurement procedure consisted of acquiring data for 14,400 s at a fixed grazing angle. Measurements were acquired over angles ranging from 2.2 to 7.9 mrad, in increments of 0.2 mrad. Fig. 5 shows a sketch with the relevant parameters of our setup. The best results were seen for grazing angles that maximized the characteristic emission lines (95–120 keV), which from Eq. (1) correspond to grazing angles of 3.4–4.4 mrad.

4. Performance

The energy bandwidth of the optic is such that at a fixed grazing angle all K_α and K_β lines of interest are reflected onto the detector. Photons of different energies are specularly reflected at angles given by Eq. (1). In practice this means that the collimated beam is