November 20, 2019, A. Borella, Belgium

Name of Experimental Campaign:

Belgium exercise to investigate performance of measurement methods

Technology Name: Gamma-ray Spectroscopy with Cadmium Zinc Telluride Detectors

Physical Principle/Methodology of Technology: Medium resolution gamma-ray measurements between 0 and 1600 keV

What Does the Method Determine/Measure (e.g., presence of nuclear material, isotopics, mass): Because traditional gamma-ray spectroscopy approaches are difficult to apply with this detector, this technology was tested to see if qualitative information could be obtained in terms of being able to discriminate weapons-/civil-grade Pu. The sensitivity to mass is studied by looking at count ratios in defined region-of-interests (ROI).

What Is the Applicability to IPNDV: Verification of NED material

Type of Data Collected by the Technology:

Gamma-ray spectra

Constraints (e.g., time to install the equipment, measurement times including distance from object, dose rate required, required Cd shielding to limit the count rate):

Room temperature detector, measurement times were between 20–120 minutes with a distance between 0–25 cm. The distance was chosen to limit the dead time to less than 5%. Dose rate and measurement time are not required for the analysis. Cd shielding (1 mm) is advised to limit the count rate due to the 60 keV gamma-ray line of 241Am. The method was also tested with 5 cm CH_2 and 10 mm Pb shielding.

Physical Description/Diagram/Photos of the Experimental Setup/Layout:

A 10 mm×10 mm×5 mm quasi-hemispherical CZT detector (Model SDP500S from Ritec Ltd.) with a preamplifier embedded in the detector head was used. The data acquisition was done with an MCA527 from GBS-Elektronik connected to a laptop computer.

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Specific Objects Measured (which of the experimental objects were measured; if not described elsewhere, describe experimental objects here):

Fuel with composition as specified in "Design information IPNDV_v03.pdf."

Mechanical design of the fuel container as given in "105761 - Measurement setup IPNDV.zip." The fuel type is indicated as follows:

- 96 mid: 50 cm fuel pins, dismountable, U_{enr} 0.72%, Pu content 4.37%, ²³⁹Pu 96%
- 96 bottom: 50 cm fuel pins, dismountable, U_{enr} 2.00%, Pu content 3.03%, ²³⁹Pu 79%
- 79 mid: 50 cm fuel pins, pelleted, U_{enr} 0.72%, Pu content 5.1%, ²³⁹Pu 79%
- 62 mid H-MOX: 100 cm fuel pins, H-MOX, U_{enr} 0.4%, Pu content 12.6%, ²³⁹Pu 62%

All measurements were carried out without a lead collimator and with 1.1 mm Cd around the fuel assembly. The "62 mid H-MOX" was measured in 1, 19, and 61 pins arrangements.

"62 mid H-MOX" 61 pins was also measured with 5 cm CH2 and 10 mm Pb shielding. These two measurements were carried out also with a lead collimator.

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Process Required to Analyze the Data (include any software used):

Net counts in given ROI were determined for each spectrum. The ROI were selected to include strong peaks.

Preliminary Results (qualitative, not quantitative; e.g., did the method perform as expected, if not how was it different):

Final Results (if available; if not, estimate of when final results will be available):

Spectra

The left figure shows the obtained spectra in the 19 pin configurations for the 96 mid, 79 mid, and 62 mid H-MOX fuel. The figure on the right compares the detector response obtained with CZT with the one obtained with LEGE detectors. The worse energy resolution results in peak overlapping.



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We could determine that the observed difference and apparent peak shift is due to 241Am, which is significantly contributing to the observed response (98.95 keV, 101.07 keV, 102.96 keV).

ROI Ratio Analysis

We determined the subtracted counts $C_{i,i,k,m}$ for different ROI and fuel types.

The background of each ROI was determined by linear interpolation using the average spectral values over three channels before and after the beginning and end of the ROI, respectively, and subtracted. The indexes *i*, *j*, *k* and *m* have the following meaning:

- *i* ROI
- *j* fuel type (see above)
- *k* number of pins (1, 19, and 61)
- *m* type of shielding and collimator (Cd, CH2+Pb, Cd+coll, CH2+Pb+coll)

The ROI were chosen around 100, 208, 335, 414, 662, and 723 keV.

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Change in ROI wrt Mass

For the fuel 62 mid H-MOX fuel assembly with 1, 19, and 61 pins, measured with Cd and without collimator, the following quantity was determined:



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In this way $\alpha_{i,62 \text{ mid H}-\text{MOX},1,Cd}$ equals 1 for every *i* and the trend of $\alpha_{i,62 \text{ mid H}-\text{MOX},k,Cd}$ can indicate a change in the detection efficiency due to a different mass and therefore a different self-absorption with respect to the reference (1 pin). Given the different number of pins, the geometry and therefore the solid angle seen by the detector are also different. This phenomenon comes into play in the observed trend of $\alpha_{i,62 \text{ mid H}-\text{MOX},k,Cd}$.

Change in ROI wrt Composition

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We considered fuel with 19 pins with types 96 mid, 96 bottom, 79 mid, 62 mid H-MOX measured with a Cd shield and without collimators and determined:

$$\alpha_{i,j,19,Cd} == \frac{\frac{C_{i,j,19,Cd}}{C_{i,96 \text{ mid},19,Cd}}}{\frac{C_{i,96 \text{ mid},19,Cd}}{C_{723,96 \text{ mid},19,Cd}}}$$

Given the high content 241Am, it would be interesting to carry out a "spectrum stripping" by subtracting a spectrum from a pure 241Am source; in this manner the differences versus 239Pu content could be more evident. In the considered cases, the different 241Am may come into play when looking at different composition.



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In this way $\alpha_{i,96 \text{ mid},19,Cd}$ equals 1 for every *i* and the trend of $\alpha_{i,96 \text{ mid},19,Cd}$ can indicate a change in the detector response due to a different composition with respect to the reference (96 mid). The red and black dots should agree within the uncertainties although the composition of 96 bottom and 79 mid is not the same due to a different Pu content and U enrichment, which could impact the results.

Change in ROI wrt Shielding and Collimation

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The data with CH2 (5 cm) and Pb (10 mm) shielding indicate that the signatures above 350 keV start to become visible. The use of a collimator is recommended to improve the peak to background below 300 keV and limit the influence of a non-uniformly space distributed gamma source (e.g., axial profile, other sources in the room). However, when looking at the $\alpha_{i,j,k,m}$ the impact due to the collimator does not seem to be significant for the considered cases.

All Data

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So far, we analyzed a subset of data where only one characteristic (composition, shielding, mass) was changed. Now we consider all spectra together to see if the characteristics (composition, shielding, mass) can be identified by studying $\alpha_{i,i,k,m}$. The reference was the 96 mid fuel and $\alpha_{i,i,k,m}$ was defined as:

$$\alpha_{i,j,19,Cd} == \frac{\frac{C_{i,j,19,Cd}}{C_{i,96 \text{ mid},19,Cd}}}{\frac{C_{i,96 \text{ mid},19,Cd}}{C_{723,96 \text{ mid},19,Cd}}}$$

Grouping all data together reveals that it is not evident to distinguish the influence of composition, mass, and shielding/collimator, although it is possible to identify ROI ratios that are more sensitive to variation in the material change.



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The line at around 560 keV is probably due to n,γ reaction in Cd of the CZT detector because its intensity is enhanced with the CH2 shielding. There is also a line at about 1600 keV with an intensity that is decreasing with collimation and shielding. This line should correspond to the 1588 keV line of 228Ac decaying in to 228Th as part of the 232Th decay chain initiated by 240Pu in the fuel.

Lesson Learned (e.g., what went well, what went wrong or not as expected, do the results confirm what we said in the technology tables?):

This technology was not explored in the technology table and traditional approaches based on peak analysis are not well developed for medium-resolution gamma-ray detectors with complex spectra.

The advantage of CZT detectors is that they can be operated at room temperature and are compact.

The proposed simplified data analysis approach revealed that ROI ratios are sensitive to mass and composition changes as well as shielding and collimation. It is to be seen how this approach can be deployed in a real scenario.

A 241Am peak stripping could be applied in an attempt to clarify the sensitivity to fuel composition.