Testing and verifying information barrier methodology on spent fuel assemblies

Naeem Syed*, Styrkaar Hustvei*, Tore Ramsøy**, Kjell Johansen**

*Norwegian Radiation Protection and Nuclear Safety Authority (http://dsa.no)

**Institute of Energy Technique, IFE (http://ife.no)

Abstract: The UK-Norway initiative (UKNi) information barrier method for the determination of isotopic ratio of ^{239}Pu / ^{240}Pu has been tested. The test specimen was spent fuel assemblies with varying plutonium content. The same gamma -ray data were analyzed and compared using commercially available PC/FRAM code.
The preliminary results show a good agreement of the results determined by UKNi method and PC/FRAM method. However, some discrepancies are observed on the deduced results, due to the presence of other radioactive nuclei, high count rate and high radiation background. These results are important and would help us in understanding the spoofing the IB results due to the presence of other radionuclides, due to the presence of other radionuclides.

Introduction

The UK-Norway initiative (UKNI) has been investigating technologies and methodologies that could contribute to a future nuclear disarmament verification effort. A key project has been the development of an Information Barrier (IB), a system that is designed to verify a set of attributes assigned to a nuclear warhead, whilst not revealing the information that is being regarded as being sensitive by the host party. To be useful, the host and inspecting party must have the confidence that the IB does what it is specified to do (and nothing else). This sets strong requirements on the certification and authentication of the employed software and hardware.

The efforts of the UKNI-IB were centered around the automated analysis of a γ-ray assay for attributes like the presence of plutonium and its isotopic ratio. For a plutonium based nuclear warhead, the isotopic ratio of ^{240}Pu / ^{239}Pu is commonly assumed to be indicative of weapon suitable material. The goal of the IB development is, therefore, to verify explicitly whether the isotopic ratio of an inspected item is below an agreed upon threshold, without revealing the total γ-ray spectrum of the tested specimen.

In 2017, the International Partnership for Nuclear Disarmament Verification (IPNDV) identified [1] testing and exercising potentially promising technologies and procedures. In this framework, Belgium proposed and organized an international exercise, held on 9 – 26 September 2019 at the site of the Belgian Nuclear Research Centre (SCK•CEN) in Mol, Belgium. Participants from Australia, Belgium, Canada, the European Union, Finland, Hungary, Japan, Norway, Switzerland, and the United Kingdom, participated to the experimental campaign. The objective of the exercise was to investigate the performance of various measurement methods with respect to their capabilities to verify the presence or absence of nuclear material originating from a nuclear weapon and to distinguish weapons-grade from reactor-grade nuclear material.

Norwegian team (IFE and DSA experts) participated to measure the isotopic plutonium composition (^{240}Pu / ^{239}Pu) in mixed oxide fuel pins arranged in compact hexagonal configuration. The methods used for this purpose was UKNi developed method. This article summarizes the preliminary results of the IPNDV exercise campaign. A brief introduction of the tested UKNi algorithm [2] for the isotopic ratio (^{240}Pu / ^{239}Pu) has been given under along with the summary and discussion on preliminary results.

Experimental Setup

Under the IPNDV test campaign, the material to be assayed was mixed oxide (MOX) fuel pins arranged in a compact hexagonal configuration. The plutonium isotopic composition, fuel pin types with a ^{239}Pu content between 61%wt and 96%wt were used. The number of rods is 19 with length of either 50 cm or 100 cm. The fuel assembly with 50 cm long fuel rods had different isotopic composition along the axial direction, middle portion having 96 % ^{239}Pu content. Norwegian team tested only fuel assemblies containing 61 % and 96 % enriched ^{239}Pu.

A high-resolution p-type coaxial HPGe γ-ray detector (ORTEC GEM-30185) was connected to the Data Acquisition system using FAST ComTec MCA4, with 32k ADC resolution, to acquire the γ-ray data from test samples. For each test specimen γ ray spectra were collected for half an hour. In order to observe the effects of count rate of (irradiated) fuel assemblies lead absorbers of 0 mm, 5 mm and 10 mm were used. Additionally, a lead collimator on the crystal probe of the detector, with a 3 mm opening in the center, as shown in Fig. 1, was used to obtain better peak to Compton scattering ratio. The algorithm [2] was applied to following data:

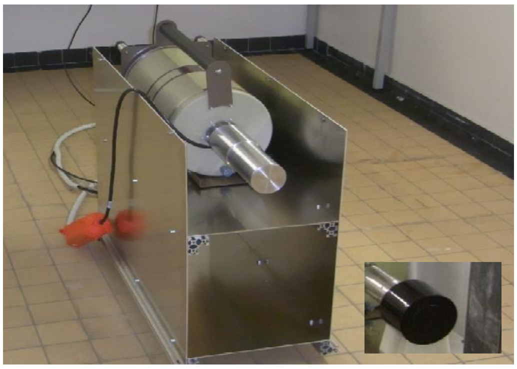


Figure 1: Experimental setup for gamma-ray detection

- Raw decay γ-ray data collected from fuel bundle containing 100 cm long fuel pins with ^{239}Pu content ~ 62 %.
- Raw decay γ-ray data collected from fuel bundle containing 50 cm long fuel pins. This MOX fuel bundle had three different Pu content, with the middle part having ^{239}Pu content ~ 96 %.

Analytical Method:

UKNI-IB is designed to determine the presence of ^{239}Pu and the isotopic ratio of ^{240}Pu/^{239}Pu in a plutonium sample. An isotopic ratio of 0.1 has been chosen as reference level to verify the tested sample as "weapon grade".

In our analysis we concentrated on the determination of the isotopic ratio from the γ-ray spectra in the energy region 630 – 680 keV. This methodology is the same as described in the reference [3]. The algorithm [2] is implemented in C++ using the ROOT data analysis framework [4]. The main features of the UKNI algorithm [2] are described under:

1. Calibrating the spectra using ^{152}Eu decay peaks at energies 121.782 keV and 77.904 keV. The IB system assumes a linear energy distribution between these two energies.
2. Identification and location of the centroids of ^{241}Am peak at 662 keV and of ^{239}Pu peak at 645.94 keV.
3. Calculating the average background at energies greater than 662 keV
4. Determining the Full Width Half Maximum (FWHM) of ^{241}Am peak at 662 keV. Assuming Gaussian distribution, the peak is calculated: $n_x = k_1 e^{K_2(x-c')^2} + b_x$, here

n_x	modelled count in channel x	x	channel number
K_1	normalization factor	c'	calculated location of centroid
K_2	Gaussian constant	b_x	background in channel x

   a. A χ^2parameter is calculated for each channel: $\chi_x^2 = \frac{(n_x - m_x)^2}{m_x}$, here n_x is the modelled count and m_x is the measured count in channel x
   b. A χ_{red}^2 is thus calculated for the peak region: $\chi_{red}^2 = \frac{\sum \chi_x^2}{\nu}$, here, ν is the degrees of freedom.
   c. An optimized value of $\chi_{optimal}^2$ is estimated by calculating the χ_{red}^2 of adjacent channels and selecting the lowest value.
   d. Since, $\chi_{optimal}^2 = g(FWHM)$, the optimized FWHM gives the peak height of 662 keV.
   e. The final χ^2 value should pass two cirteria, (i) $\delta\chi^2 < 0.001$ and (ii) $\chi^2 < 16$.
5. Using the same method as above χ_{red}^2 is calculated for ^{239}Pu peak at 645.94 keV. However, FHWM is held constant and an optimal $\chi_{optimal}^2$ is calculated as a function of background continuum $\chi_{optimal645}^2 = g(b_{645})$.
6. A multiplet incorporating the ^{239}Pu, ^{241}Am, and ^{240}Pu peaks is de-convoluted for 642.35 keV γ-ray photopeak. This is done by estimating the contributions from other nearby prominent peaks.
   a. The estimated contribution is given by: $n = \frac{I_{\gamma 637}}{I_{\gamma 645}} h_{645}\, e^{(x-c_{637})^2} + \frac{I_{\gamma 640}}{I_{\gamma 645}} h_{645}\, e^{(x-c_{640})^2} + \frac{I_{\gamma 641}}{I_{\gamma 662}} h_{662}\, e^{(x-c_{641})^2}$, here

$I_{\gamma x}$	Emission prob at x KeV	x	channel number
h_x	Height of measured γ-ray photopeak at x keV	c_x	channel of centroid of modelled γ-ray peak at x keV

   b. If m' is the remaining pulse height distribution once modelled peaks are subtracted: $m' = m - n$
   c. χ_{red}^2 value of our region of interest is evaluated and optimized
7. The isotopic ratio, ^{240}Pu/^{239}Pu is derived using: $n = \frac{\left[\frac{h_{642} \cdot t_{1/2} \cdot (^{240}Pu)}{I_{\gamma 642}}\right]}{\left[\frac{h_{645} \cdot t_{1/2} \cdot (^{239}Pu)}{I_{\gamma 645}}\right]}$, where

h_x	net peak at x KeV, and	$t_{\frac{1}{2}}(^{y}Pu)$	half-life of Pu isotope

Results and Discussion:

The results of IB algorithm on IPNDV test samples are shown in Figure: 2 - 4. The arrows indicate the peak values of the 645.96 keV ^{239}Pu and 662 keV ^{241}Am. The continuous red lines give the χ^2 minimized fit to the data and the dashed lines highlight the estimated background. The ^{240}Pu contribution to the multiplet at ≈ 642 keV is fitted after subtraction of the contribution by ^{239}Pu and ^{241}Am and the whole resulting multiplet is drawn with the dashed green line.

The method has previously been successfully tested on PIDIE Pu samples and Pu test samples [3]. At IPNDV test campaign, it had been observer that background radiation and count rate was high. This put a limitation on the throughput of measured gamma-ray spectra, as the detector dead time was observed to be high. This resulted to relatively low data statistics at region of interest in a given time.

The multiplet peak at 640 keV γ-ray energy region is more prominent in 50 cm long fuel assembly spectra than the one in 100 cm long fuel assembly γ-ray spectra.

The stored γ-ray spectra of two tested fuel assemblies were also analyzed with the commercially available code PC/FRAM [5] to determine the Pu content and isotopic ratio. The preliminary results are summarized in Table 1. The best determined ^{239}Pu content ~ 91.13 % for the 50 cm long fuel assembly with no absorber as compared to PC/FRAM giving 93.15 % given ^{239}Pu content of 96 %. Similarly, best estimated ^{239}Pu content ~ 62.86 for the 100 cm long fuel assembly with no absorber as compared to PC/FRAM giving 55 % and given value of ^{239}Pu 62%.

The investigation on the deviations of determined isotopic ratio using UKNi methods in comparison to the given nominal values is still underway.

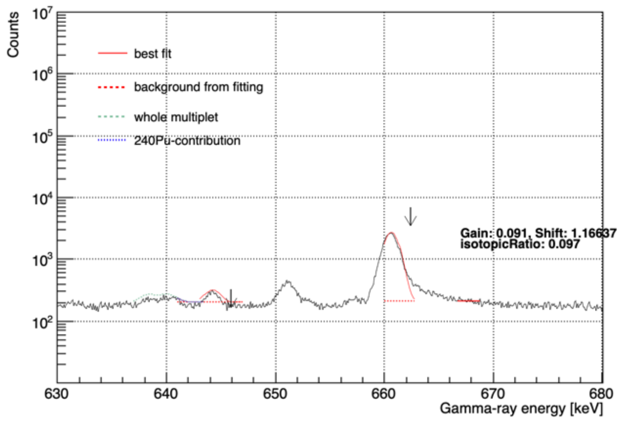
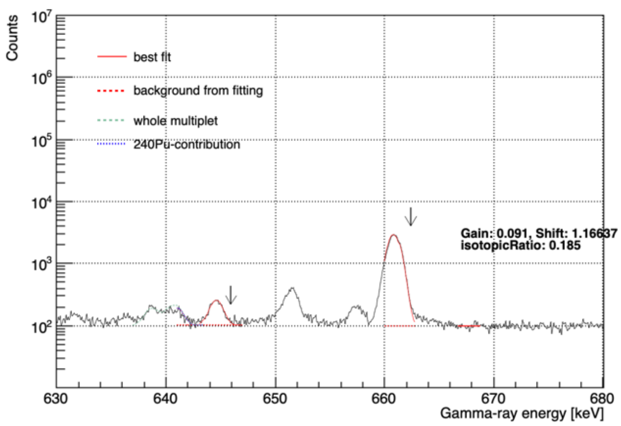


Figure 2:UKNi algorithm applied on 50 cm long fuel assembly with no absorber.



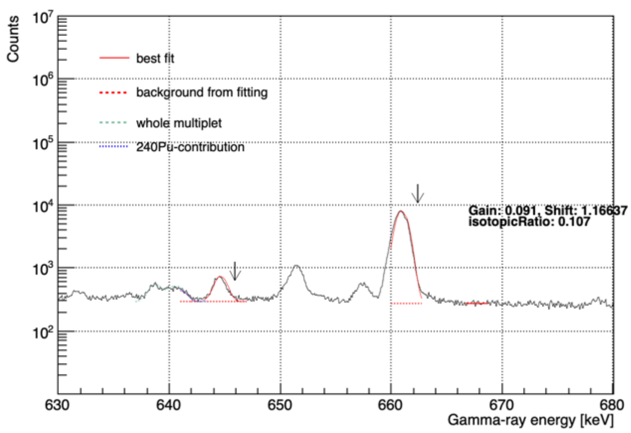


Figure 3: UKNi algorithm applied on 50 cm long fuel assembly with 5 mm and 10 mm lead absorbers repectively.

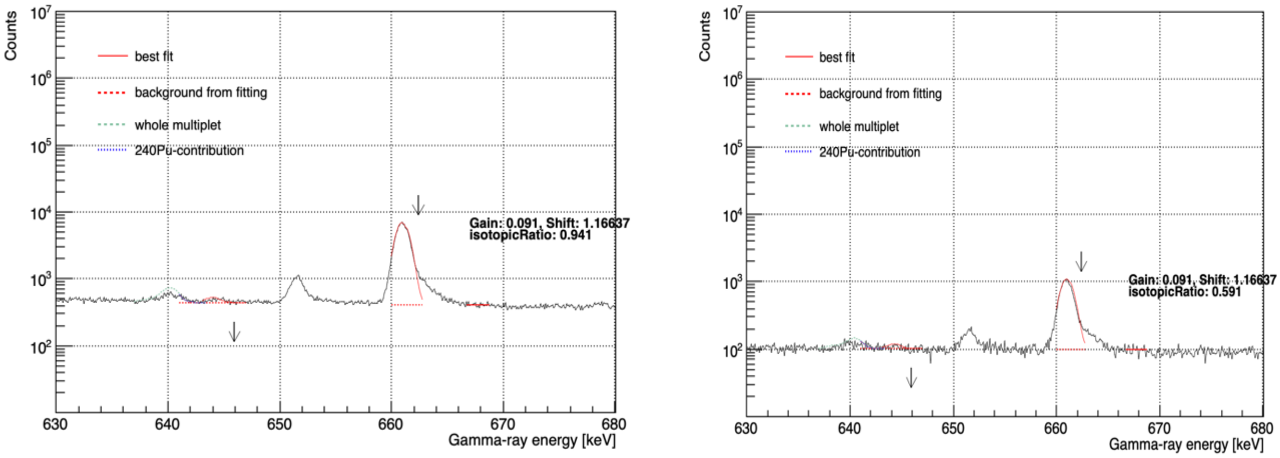


Figure 4: UKNi algorithm applied on 100 cm long fuel assembly with 5 mm lead absorber and no absorber respectively.

Table 1: Summary of the isotopic ratio determination using UKNi method and PC/FRAM method

Test Fuel Assembly (FA)	**Isotopic Ratio (^{240}Pu/^{239}Pu)**	**^{239} Pu %**	
	UKNi	**PC/FRAM**	**UKNi**
50 cm long FA with 5mm lead absorber	0.185	-	84.37 %
50 cm long FA with 10mm lead absorber	0.107	91.62 %	90.34 %
50 cm long FA with no absorber	0.097	93.15 %	91.13 %
100 cm long FA with 5mm lead absorber	0.941	74.78 %	51.51 %
100 cm long FA with no absorber	0.591	55 %	62.86 %

References:

1. https://www.ipndv.org/reports-analysis/working-group-6-belgian-measurement-campaign/
2. Edward Day. Information barrier software: Detailed algorithm overview. Technical report, UKNI, Version 1.0. Retrieved: 01.09.2016.
3. N. Syed, S. Hustveit, O. Reistad, S. Siem,, "Investigating the method of UK-Norway initiative information barrier", *INMM 60th Annual Meeting Proceedings*, California USA, 2019.
4. Rene Brun and Fons Rademakers. ROOT – an object oriented data analysis framework. *NIM A,* 389(1-2):81 – 86, Apr 1997. ISSN 0168-9002. doi: 10.1016/s0168-9002(97)00048-x. URL https://root.cern.ch/.
5. T.E.Sampson, T.A.Kelley, "PC/FRAM: a code for the nondestructive measurement of the isotopic composition of actinides for safeguards applications"*, Applied Radiation and Isotopes,* vol. 48, issue 10-12, 1997, p. 1543-1548