November 20, 2019, A. Borella, Belgium

#### Name of Experimental Campaign:

Belgium exercise to investigate performance of measurement methods

## Technology Name: Gamma ray Spectroscopy with Low Energy Germanium Detector

## Physical Principle/Methodology of Technology:

High-resolution gamma-ray measurements between 0–300 keV. The assayed sample should have little or no shielding.

## What Does the Method Determine/Measure (e.g., presence of nuclear material, isotopics, mass):

The Pu isotopics, time since last separation, <sup>235</sup>U/Pu and <sup>238</sup>U/Pu is determined with the FRAM code version 4. An appropriate configuration file ("parameter set") is needed to process the data.

#### What Is the Applicability to IPNDV:

Verification of NED bare material or with minimal amount of shielding

## 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):

Need LN<sub>2</sub> cooling and periodical filling (with our detector every 24 h). The detector weighs about 5 kg. Measurement times were between 12–30 minutes with a distance between 2.5–25 cm. The distance was chosen to limit the dead time to few percent. The knowledge of the dose rate and of the measurement time are not required. Cd shielding (1 mm) is advised to limit the count rate due to the 60 keV gamma-ray line of 241Am. The method works only with samples with limited shielding.

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

The detector is from Ortec. The resolution at 59.5 keV is 0.62 keV, at 208 keV is 0.83 keV, which is worse than recommended for the data analysis (0.55 keV at 122 keV).

The data acquisition is done with Genie in combination with an Inspector 2000 from Canberra.

A lead collimator was used to limit scattered radiation. The collimator allows also being closer to the sample and limiting a possible impact spatial dependence in the isotopic composition.

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

Fuel assembly with 19 pins with 1.1 mm Cd 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<sub>enr</sub> 0.72%, Pu content 4.37%, <sup>239</sup>Pu 96%
- 96 bottom: 50 cm fuel pins, dismountable, U<sub>enr</sub> 2.00%, Pu content 3.03%, <sup>239</sup>Pu 79%
- 79 mid: 50 cm fuel pins, pelleted, U<sub>enr</sub> 0.72%, Pu content 5.1%, <sup>239</sup>Pu 79%
- 62 mid H-MOX: 100 cm fuel pins, H-MOX, U<sub>enr</sub> 0.4%, Pu content 12.6%, <sup>239</sup>Pu 62%

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

- FRAM version 4
- "UPu60\_210SolidX" parameter set for Planar detector (0.075 keV/ch, Equ. w U, U235/Pu < 1, 60-210keV with physical model of the efficiency). Default settings used.

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):

The figure below shows spectra obtained with the LEGE detector for the "96 mid," "79 mid," and "62 mid H-MOX."

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The obtained results in terms of isotopics composition, U/Pu ratios, and time from separation (*Sep*) are given in the following table. *N* stands for nominal value, *C* stands for the calculated value. The nominal value was

obtained from the "Design information IPNDV\_v03.pdf" corrected for decay by using Nucleonica.

96 mid			79 mid			96 bottom			62 mid H-MOX				
Nuclide		Ν	C/N	$\sigma_{\text{C/N}}$	Ν	C/N	$\sigma_{\text{C/N}}$	Ν	C/N	$\sigma_{\text{C/N}}$	Ν	C/N	$\sigma_{\text{C/N}}$
Pu238	% mass	0.00			0.05	1.28	0.15	0.05	1.25	0.18	1.05	1.07	0.03
Pu239	% mass	95.88	0.99	<0.01	81.61	0.98	<0.01	81.61	0.98	<0.01	66.94	0.99	0.01
Pu240	% mass	4.09	1.13	0.06	17.64	1.08	0.02	17.64	1.11	0.02	25.36	1.03	0.02
Pu241	% mass	0.02	1.22	0.08	0.24	1.06	0.02	0.24	1.09	0.02	1.89	1.04	0.02
Pu242	% mass	0.01	3.00	0.65	0.46	1.01	0.07	0.46	1.03	0.08	4.76	0.96	0.03
Am241	mg/gPu	2.09	1.10	0.05	27.28	1.07	0.02	27.28	1.09	0.02	75.44	1.11	0.02
UX-ray/Pu		20.97	0.09	<0.01	18.36	0.14	<0.01	31.98	0.07	<0.01	6.48	0.95	0.01
5U/Pu		0.15	1.11	0.20	0.13	1.01	0.06	0.64	1.00	0.05	0.03	1.27	0.42
8U/Pu		20.81	0.88	0.17	18.23	0.88	0.31	31.32	1.07	0.20	6.45	2.91	2.31
Sep /y		51.92	0.96	0.02	52.86	1.00	0.01	52.86	1.00	0.01	33.84	1.03	<0.01

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In the graph, the uncertainties are not shown for clarity but can be found in the table.

Notice that the C/N for 242Pu is out of the scale for the 96 mid case.

The relative uncertainty on 239Pu is 0.2-0.8%, the relative bias is maximum 2.5%.

The obtained net peak areas used for the activity ratios calculations in FRAM are given below. These represent only a subset of the peaks that are observed in the spectra.

		96 mid		79 mid		96 bottom		62 mid H	I-MOX
Nuclide	energy	area	error	area	error	area	error	area	error
U238	92.792	482	20%	323	36%	478	20%	164	79%
Uxray	94.658	16865	3%	18157	1%	14257	2%	19275	1%
Uxray	98.441	37541	3%	41609	1%	24142	1%	44310	1%
Pu241	101.066	851	11%	7472	2%	5255	2%	26183	1%
Am241	102.961	10367	3%	99365	1%	70275	1%	133702	1%
Pu240	104.242	5315	3%	16569	1%	11837	2%	10773	2%
Am241	125.292	1586	8%	16137	2%	11210	2%	24782	1%
Pu239	129.294	23168	5%	14630	2%	10671	2%	6441	2%
Pu241	148.567	268	33%	3585	4%	2328	4%	15760	1%
Pu241	164.597	147	60%	927	11%	476	17%	5110	3%
U235	185.715	3214	18%	2135	6%	6907	5%	309	33%
Pu239	203.545	5615	7%	4075	3%	2096	5%	1893	6%
Pu241	208	2406	6%	22444	2%	16388	2%	97021	1%

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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?):

In the future it would be interesting to measure with a different gain settings to be able to use the *Planar\_Widerange3* parameter set that uses data up to 430 keV.

The 242Pu was estimated using correlation curves available in FRAM. The estimate of 242Pu for weapons grade Pu from correlation curve does not work well. However, the 242Pu content is less than 0.1%. Fine tuning of the parameter set could improve bias.

Good results were obtained despite the high presence of 241Am.