A measurement campaign in support of technologies for disarmament verification

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ABSTRACT

The performance of measurement technologies play an important role in view of developing a verification strategy in a nuclear disarmament scenario. In recent times, the International Partnership for Nuclear Disarmament Verification (IPNDV) has identified testing technologies and procedures as an important step.

In this framework, Belgium proposed and organized at end of 2019 an international exercise at the site of the Belgian Nuclear Research Centre (SCK CEN) in Mol, Belgium. The aim of the exercise was testing and comparing measurement techniques to be potentially used for the verification of nuclear material in the framework of the dismantlement of nuclear weapons. SCK CEN made available well characterized nuclear material in form of Mixed Oxide fuel previously used to benchmark neutron transport codes and data. The relative content of Pu was up to 14%$_{wt}$ and the isotopic abundance of ^{239}Pu was up to 93%$_{wt}$. The amount of nuclear material being assayed could be chosen by the participants as well the type of an optional shielding material.

A total of ten measurement teams participated in the exercise deploying different gamma-ray and neutron measurement devices.

In this paper we report about the content of the measurement campaign and some results that were obtained when deploying a Low-Energy Germanium detector and neutron coincidence ^{3}He based slab counters.

Keywords: Nuclear Disarmament Technologies; Measurement campaign; International Partnership for Nuclear Disarmament Verification (IPNDV); Low-Energy Germanium detector, Neutron coincidence ^{3}He based slab counters

INTRODUCTION

At the end of Phase I in December 2017, the International Partnership for Nuclear Disarmament Verification (IPNDV) identified “testing and exercising potentially promising technologies and procedures” as a key next step.

In this framework, Belgium has proposed and organized an international exercise at the site of the Belgian Nuclear Research Centre (SCK CEN) in Mol, Belgium. The aim of the exercise was testing and comparing measurement techniques to be potentially used for the verification of nuclear material in the framework of the dismantlement of nuclear weapons.

The exercise was meant to complement a second Nuclear Disarmament Verification exercise called “NuDiVe” focused on testing procedures related to a simulated nuclear warhead dismantlement, hosted in Germany jointly by German and French teams.

This report summarizes the content of the exercise and the experimental campaign carried out between 9 and 26 September 2019 and reports about the results of the Belgian delegation who deployed a Low-Energy Germanium detector and neutron coincidence ^{3}He based slab counters. Measurements with a Cadmium Zinc Telluride detector were also carried out but are not reported in this paper.

THE MEASUREMENT CAMPAIGN

Objectives and types of measurements

The main goal of the measurement campaign was to investigate the performance of different non-destructive passive measurement technologies with respect to their capabilities to: verify the presence or absence of nuclear material originating from a nuclear weapon; distinguish weapon grade from civil grade nuclear material.

The material to be assayed was unirradiated Mixed Oxide (MOX). The plutonium is mixed with uranium, chemically in the form of oxide. The total plutonium content is up to 14%$_{wt}$ with a relative ^{239}Pu amount up to 96%$_{wt}$. The fuel pins were arranged in a compact hexagonal configuration as shown in Fig .1 to maximize the average density of the item to be assayed.

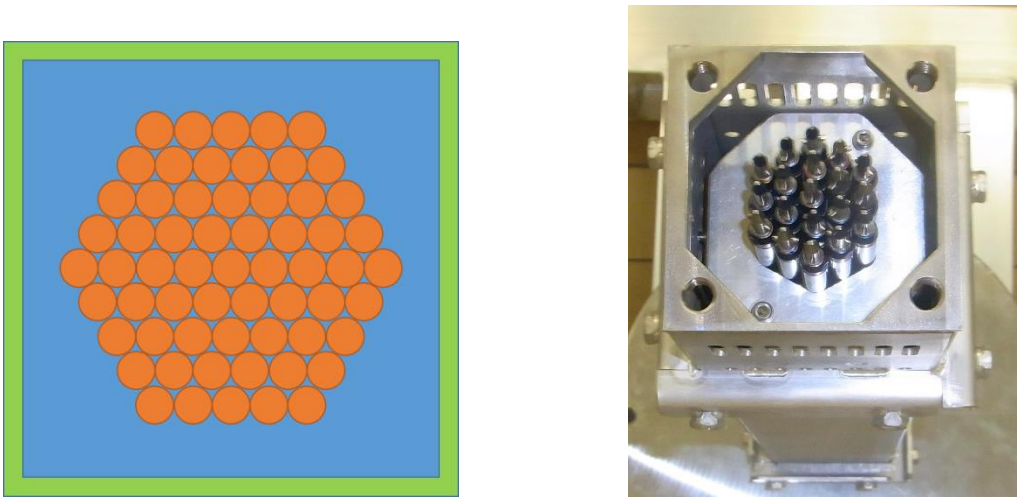

Figure 1. Horizontal cross-section of the container with 61 fuel pins (left). The top view of the 19 pins configuration (right).

The composition and geometry of the MOX fuel as well as the design information of the measurement setup was well known. Hence, the data acquired in the exercise provide a sound benchmark to validate developed models for the deployed technologies.

During the measurement campaign it was possible to study the influence on the performance due to: the amount of nuclear material; the type of nuclear material; the type of shielding material. For example, by performing measurements on samples with various amounts of plutonium mass and a fixed isotopic composition one could study the sensitivity of the methods to the amount of nuclear material. In addition, by measuring samples with different plutonium isotopic compositions one could assess whether a certain technology was capable to distinguish between reactor-grade and weapon-grade plutonium. Finally, the performances of the technologies could be assessed in presence of shielding material such as lead, cadmium, and polyethylene. This could represent natural shielding of the fissile material but could also imply a possibility to divert nuclear material during the dismantling process.

Characteristics and significance of the chosen materials

A total of five assemblies with given composition were available for the experiments. Emission rates for most intense gamma-rays and neutron emission rates for the considered assemblies are shown in Table 1 and 2. The hexagonal bundle was hosted in a 3 mm thick square container of stainless steel to reduce the gamma-ray dose rate and facilitate the manipulation. The dimensions of the horizontal cross-section of the container were 10 cm × 10 cm. The following individual shielding could be installed on the container: Lead 5 mm and 10 mm thick; Cadmium 1.1 mm thick; Polyethylene (PE) 50 mm thick; a combined shielding of polyethylene (50 mm thick on the inside) and lead (10 mm thick on the outside).

The assemblies *62–1, 62–19, 62–61* were fuel pins with a plutonium content of 12.6%$_{wt}$ and with a ^{239}Pu amount of 61%$_{wt}$. The number of rods were 1, 19 or 61; these numbers allowed to keep a hexagonal arrangement and to have a ^{239}Pu mass range from 0.1 to 2.6 kg. The uranium enrichment was 0.4%. These assemblies could be used for measurements dedicated to the study the mass determination or sensitivity.

The assemblies *62–19*, *79–19* and *96–19* have a ^{239}Pu relative content in the PuO_2 of 61%$_{wt}$, 79%$_{wt}$ and up to 96%$_{wt}$ respectively. The *96–19* is an axially heterogeneous assembly ^{239}Pu relative content of 79% at the top and bottom and 96%$_{wt}$ at the mid of the assembly. Amongst these assemblies the Pu content ranged between 4%$_{wt}$ and 12%$_{wt}$ and the uranium enrichment was between 0.4% and 2.0%. These assemblies could be used for measurements dedicated to determine the identify the isotopic composition.

Assembly ID	^{240}Pu 160 keV	^{235}U 186 keV	^{239}Pu 413 keV	^{239}Pu 646 keV	^{240}Pu 642 keV	^{241}Am 662 keV	^{238}U 1001 keV
79–19	1605	161	7482	75	50	3567	386
96–19	660	319	5598	56	20	1345	373
62–1	539	7	1436	14	17	2364	30
62–19	10243	134	27292	273	318	44907	576
62–61	32887	430	87621	876	1022	144174	1850

Table 1. Emission rates for most intense gamma-rays for the considered assemblies. Values are expressed in 10^3 photons/s.

Assembly	Spontaneous fission			(α,n) reaction		
ID	Pu	U	Am	Pu	U	Am
79–19	51	0.1	<0.1	17	0.5	21
96–19	21	0.1	<0.1	10	0.5	8
62–1	23	<0.1	<0.1	13	<0.1	14
62–19	439	0.1	<0.1	241	0.7	262
62–61	1410	0.3	<0.1	774	2.1	841

Table 2. Neutron emission rates for the considered assemblies. Values are expressed in 10^3 neutron/s.

The plutonium used in the measurement campaign differs from the envisaged material in a nuclear explosive device to be inspected during a disarmament verification process for the aspects outlined in Table 3.

	Exercise	Warhead
Material	Civil/weapon grade Pu mixed with depleted/natural/low enriched uranium	Weapon grade Pu metal (+depleted uranium as reflector)
Chemical form	Oxide	Metal
Geometry	Hexagonal bundle of pins	(quasi)spherical
Impurities	^{241}Am from ^{241}Pu decay (30÷85 mg/g$_{Pu}$)	^{241}Am from ^{241}Pu decay (0 ÷ 30 mg/g$_{Pu}$)

Table 3. Characteristics of the materials measured in the exercise and in a nuclear warhead.

When considering a detection system based on gamma-ray spectroscopy, the differences outlined in Table 3 have an impact on the detector response. While the gamma ray emission of plutonium does not depend on the chemical form, its density, composition and geometry affects the degree of attenuation to which the gamma radiation undergoes. The data in Table 1 indicate that the gamma-lines from both uranium and ^{241}Am should be visible in the spectra and this would complicate the spectral analysis, when compared to the one associated with a real warhead.

When considering a detection system based on neutron coincidence counting system, while the neutron emission due to spontaneous fission does not depend on the chemical form there is a significant (α,n) component associated due to the fact that the material is in oxide form; the data in Table 2 reveal that this contribution is estimated to be comparable to the contribution from spontaneous fission. The total neutron emission is therefore depending on the chemical form. Due to the non-negligible multiplication, the (α,n) component is expected to have an impact also on the number of coincident neutrons being detected. We expect therefore that time uncorrelated component associated to the (α,n) reaction makes the measurements and interpretation of the data more difficult when compared to a plutonium in metal form.

Given these considerations, the verification of well characterized MOX fuel may even be more challenging than the one of a nuclear explosive device. In addition to testing the technologies in a complex scenario, the proposed exercise provides a sound benchmark to validate developed models for verification devices based on the deployed technologies.

Participating teams

About 30 persons representing the delegations of Australia, Belgium, Canada, the European Union, Finland, Hungary, Japan, Norway, Switzerland, and the United Kingdom, participated to the experimental campaign. The Nuclear Threat Initiative (NTI) was present as observer at the beginning of the campaign. Germany provided support for calculations and support during the Belgian measurements.

Participants deployed different measurement instruments. We can categorize them as follows: Imaging devices (gamma-rays and neutrons); Total neutron counters; Neutron coincidence counters; High resolution gamma-ray detectors; Low/Medium resolution gamma-ray detectors.

MEASUREMENTS WITH A LOW-ENERGY GERMANIUM DETECTOR

The goal of the measurement was to verify Special Nuclear Material (SNM) in bare form or with minimal amount of shielding by deploying a Low-Energy Germanium (LEGE) detector and analysing the collected gamma-ray spectra. In particular, the performance of the technology was assessed with respect its ability to determine the Pu isotopics, time since last separation, ^{235}U/Pu and ^{238}U/Pu.

Detector and setup

High resolution gamma-ray measurements between 0 and 300 keV were carried out with a LEGE detector from Canberra [1], cooled with liquid nitrogen. 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 the Genie software in combination with an Inspector 2000 [2] from Canberra/Mirion.

A lead collimator was used to limit scattered radiation as shown in Fig. 2. Measurement times were between 12 and 30 minutes with a distance between 2.5 cm and 25 cm. The distance was chosen to limit the dead time to few percent.

Given that only low energy gamma-rays are considered, there is a significant self-absorption in the sample and therefore the method assays only the outer part of the sample; in combination with a narrow collimator, this limits the spatial region to which the detector is sensitive. Measurements with lead shielding were not carried out as almost no gamma-rays that could be detected would have reached the detector. However, a 1.1 mm thick Cd shielding was used to limit the count rate due to the 60 keV gamma-ray line of ^{241}Am.

Measured samples

The measured samples consisted of 19 pins hexagonal bundles, *96–19, 79–19* and *62–19* measured at the mid position. In addition, the *96–19* was measured at the bottom position where the ^{239}Pu relative content is 79%$_{wt}$.

Figure 3 shows spectra obtained with the LEGE detector for the *96–19*, *79–19* and *62–19* measured at the mid positions.

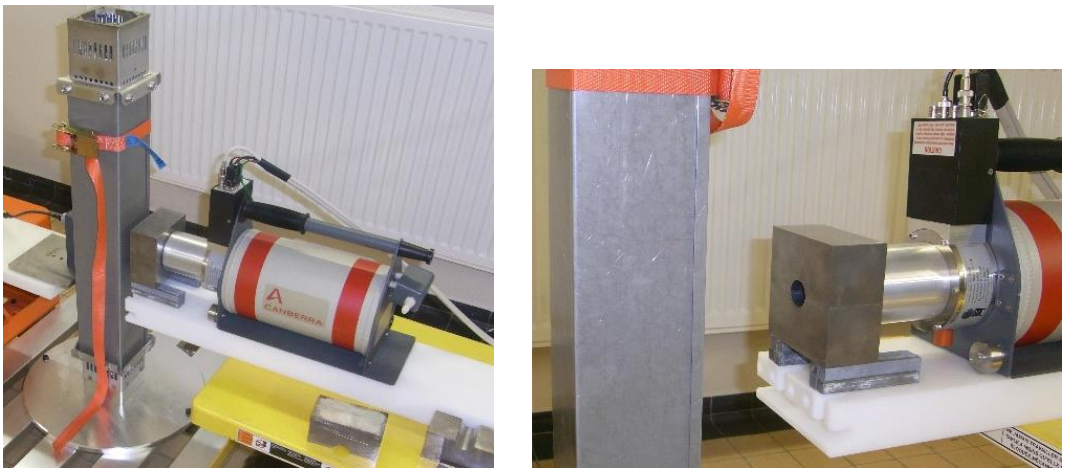

Figure 2. Setup of the measurement with the LEGE detector. A Cd shielding covers the assembly and between the detector and the assembly the lead collimator is present.

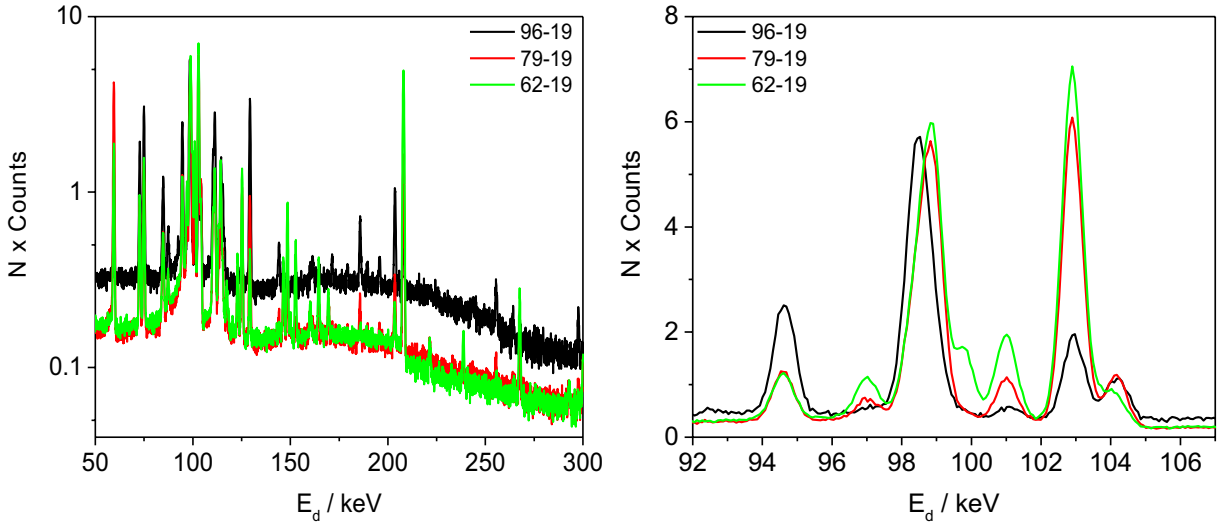

Figure 3. Obtained spectra with the LEGE detector for three samples.

Analysis and results

The data analysis was carried out with the FRAM [3] code version 4. The parameter set *UPu60_210SolidX* for Planar detector (0.075 kev/ch, ^{235}U/Pu < 1, 60-210 keV with physical model of the efficiency) was used with its default settings. The analysis is applicable only to spatially homogenously distributed samples containing SNM.

We could determine that the different content of ^{241}Am strongly affect the observed response in Fig. 3 for examples with its peaks at 98.95 keV, 101.07 keV and 102.96 keV.

The obtained results for all four sample revealed that that:

The relative uncertainty on ^{239}Pu is 0.2÷0.8%, the relative bias is at most 2.5%.

The relative uncertainty on ^{240}Pu is 2÷6%, the relative bias is at most 13%.

The relative uncertainty on the separation time is less than 2%, the relative bias is at most 3%.

The ^{242}Pu was estimated using correlation curves available in FRAM. The estimate of ^{242}Pu for the case *96–19 mid* case where ^{239}Pu relative content is the highest the ^{242}Pu content cannot be well determined. However, the ^{242}Pu content is less than 0.1%.

The ratio U/Pu from analysis of the X-ray region could not be determined in a satisfactory way.

Conclusions

We could conclude that the technology can distinguish civil from military grade Pu as well as the time from last separation, despite the increased background conditions (^{241}Am).

The method was tested in conditions with limited shielding (3 mm stainless steel + 1.1 mm Cd), but we did not explore systematically from which shielding thickness the method fails. The measurements were done only up to 300 keV; extending the range to 450 keV is a possibility that should be explored since in principle it should be less sensitive to shielding. The measurement time is in the order of 10 minutes which is acceptable for verification inspections. The fact that that the technology works only with limited shielding limits its applicability to the steps in the disarmament scenario where the SNM has been removed from the warhead and structural components.

Given the fact that sensitive information such as radionuclide vector of Pu can be revealed an information barrier similar to [4] should be out in place for its deployment.

MEASUREMENTS WITH NEUTRON COINCIDENCE ^{3}HE BASED SLAB COUNTERS

Neutron coincidence counting allows estimating the mass of a sample containing SNM provided that the radionuclide vector is known [5]. This technology was used to verify its ability to determine the mass of the available SNM and determine its performance with respect to shielding material and limit of detection.

The technology measures the time correlation between events measured with neutron detectors. In addition to determine the total count rate *T*, the reals rate *R* is determined from the number of detected neutrons in two time windows that are opened every time a neutron is detected. *R* is a measure of the amount of material emitting neutrons by spontaneous fission.

In the proposed approach we apply Hage's point model [6] to determine the intensity of the spontaneous fission (SF) source term and detection efficiency. From the spontaneous fission source term by knowing the isotopic composition of the sample (e.g. from gamma-ray spectrometry) it is possible to determine the ^{239}Pu mass.

Detector and setup

Two WM3400 slab counters [7] were deployed. The sample to be assayed is between the detectors as shown in Fig. 4.

Totals, "R+A", and "A" are measured with a JSR-12 shift register [8]. Each detector is equipped with a shift register. The logical "OR" of the signals are also processed by a shift register as well as an MCA527 from GBS-Elektronik with upgraded firmware [9]. The upgraded firmware allows saving the time stamps of the detected event for an offline analysis to determine the distribution of correlated events. By considering the two detectors as a whole one limits the impact of possible asymmetrical detector arrangements.

Figure 4. Measurement setup for the neutron coincidence counters.

Measured samples

All the five assemblies (*96–19, 79–19, 62–1, 62–19, 62–61*) were measured. All measurements were carried out with 1.1 mm Cd around the fuel assembly. For fuel types *79–19* and *96–19* the measurements were also carried out without Cd. The fuel type *96–19* was also measured with 5 cm CH_2 around the assembly and 1.1 mm thick Cd sheet on the detector and 5 mm Pb around the assembly and 1.1 mm thick Cd sheet on the detector.

Measurements were done at two distances 26 cm and 95 cm from the outer surface of the assembly. The measurement time depends on the neutron emission of the sample and was either 1800 s or 3600 s. The uncertainty on the reals rate R was about 2% at 26 cm distance and about 5-10% at 96 cm.

Analysis and results

The data analysis reported here considers only the ORred response of the two slab counters measured with the MCA527.

For the *96–19* assembly there was less impact due to (n,α) reaction, both totals and reals were insensitive to Cd, and almost insensitive to Cd+Pb (5% reduction on the totals), with CH_2+Cd the totals were attenuated by 30% and the reals by 43%.

In all the cases the uncorrelated background was relatively high with the ratio A/(R+A) ranging from 88.4% (96–19 with PE+Cd shielding) to 99.8% (*62–61* with Cd, at 1 m distance). In the worst case the reals rate could be determined with a relative uncertainty of 8%.

The equations of the Hage's point model equations in absence of multiplication

$$T = F_S \nu_{s1} \varepsilon (1 + \alpha) \qquad (1)$$

$$R/f = F_S \nu_{s2} \varepsilon^2 \qquad (2)$$

are solved being T and R the measured observables. In the equations ε is the detection efficiency, α is the ratio between the neutron emission due to (α,n) reaction and spontaneous fission, F_S the number of fission per seconds, ν_{s1} and ν_{s2} are the 1st and 2nd factorial moment of the distribution for the neutron emission through spontaneous fission. Knowledge of the die-away of the system, measured with a ^{252}Cf source (55.3±1.0 μs) or of the gate occupation factor *f*.

We used the composition from the fuel specifications to solve the equations; the α term in the Eq. 1 is determined from the composition and nuclear data of the fuel. We derived the calculated F_S source term and from the radionuclide composition the total mass of heavy metal was also calculated.

The obtained ratio between the calculated mass of heavy metal and the nominal one from the fuel specification for different cases and as a function of the mass of heavy metal mass is shown in Fig. 5.

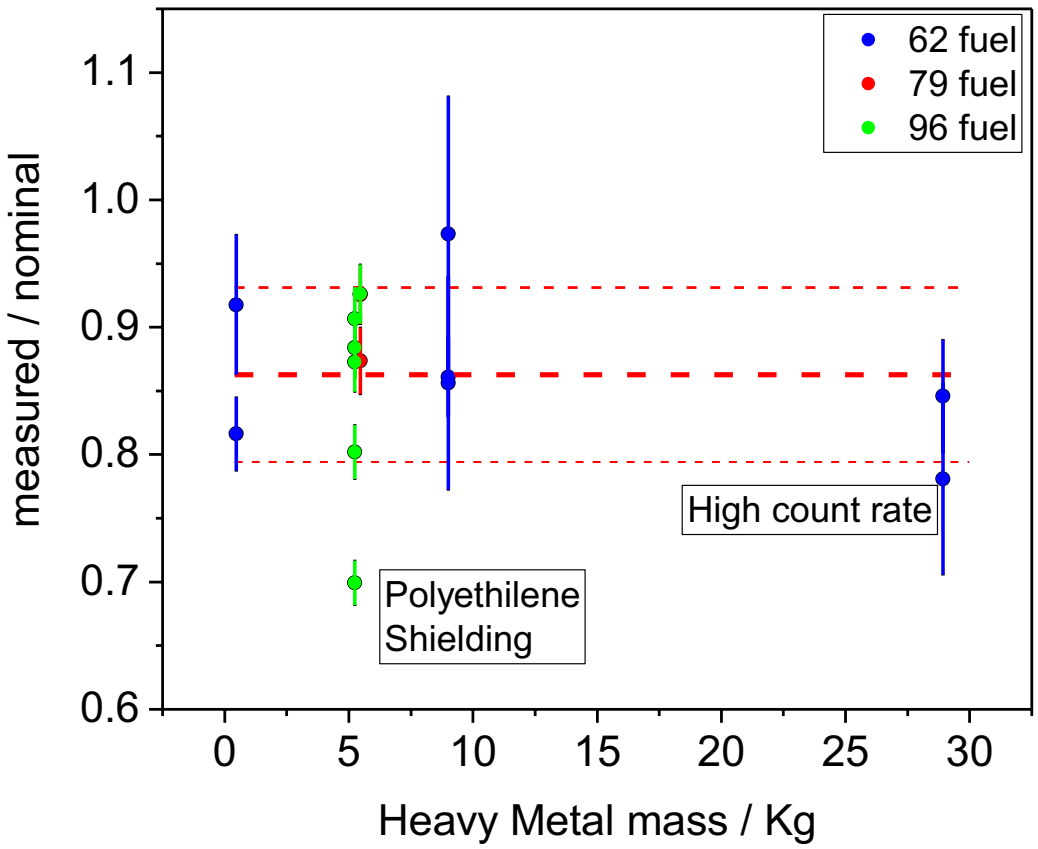

Figure 5. Measurement setup for the neutron coincidence counters.

The results show an average underestimation of the mass by about 14%. The fact that the multiplication factor is not accounted for can explain the observed bias as well as the fact that the conditions to apply the point model were not fully met. The two outliers are a case where we have a high count rate where the dead time has a potential impact on the observable that was not yet accounted for; with polyethylene shielding probably a correction for that multiplication in the assembly is needed when solving the point model equations.

Conclusions

The results indicated that measurements were challenging due to the very high (α,n) background. Such a background component is not expected to be present in verification scenario. With such unfavourable conditions we could detect ~20 g of $^{240}Pu_{eq}$ at 1 m distance in 30 minutes.

The limit of detection is however influenced not only by the (α,n) component but also by the accuracy of the background induced by neighbouring items and cosmic rays induced spallation in Pb or high Z material.

Despite the fact that the conditions to apply the point model equations were not fully met, relatively good results in terms of estimation of the total mass were achieved. The determination of the mass with the point model approach requires the knowledge of the radionuclide composition to determine the relative contribution (α,n) to spontaneous fission in the source term.

Being sensitive to spontaneously fissioning material is a signature very difficult to spoof; therefore the measurement of the reals rate can be used to detect absence/presence of spontaneously fissioning material whether with aim of quantifying the mass (with isotopics) or a quick verification of items.

SUMMARY AND CONCLUSIONS

We have presented a measurement campaign aimed at testing the performance of measurement technologies to be potentially deployed in a nuclear disarmament scenario. The measurement campaign was carried out at the premises of the Belgian Nuclear Research Centre SCK CEN. Well characterized nuclear material in form of Mixed Oxide fuel was used to test different measurements technologies with respect to their capabilities to detect the amount and the composition of SNM in presence or absence of shielding.

In the paper, we also presented the results of two measurement techniques, one based on high resolution low energy gamma-ray spectroscopy with a LEGE detector and one based on neutron coincidence counting. A technology based on a LEGE detector, could be implemented in a portable setup. Since the radionuclide composition of the items being assayed can be revelled, an IB should be in place. We expect that a technology based on neutron coincidence counting would require a fixed non portable setup where the items to be assayed is positioned as close as possible to the detectors as to maximize the detection efficiency and minimize measurement times. Since one measures count rates, an IB should be in place; however it is difficult to assess what type of requirement it would need to satisfy. One possibility could be that the systems simple confirms the absence/presence of a material emitting neutrons by spontaneous fission. If the radionuclides composition is somehow available, the system could also be used to establish a minimum mass is present, taking in account the observed bias of the point model.

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