Hold-up Measurement based on Fast Coincidence Counting and Neutron time-of-flight

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Abstract

Reprocessing, nuclear fuel fabrication, or uranium enrichment require large facilities that contain many glove-boxes, tanks and pipes where nuclear materials can build up. The quantity of fissile materials needs to be evaluated for nuclear material accounting and control, criticality safety, radiation protection and waste management. The location of the source and attenuation of radiations (neutrons and gamma-rays) are two key factors for the evaluation of nuclear material quantity. Indeed, if the source's location is unknown and/or radiations attenuations are not estimated, they become important sources of uncertainties for the evaluation of nuclear material quantity. A new system based on fast coincidences between inorganic and organic scintillators is studied to reduce uncertainties associated with the source localization and attenuations. An array of scintillators is simulated around the inspected object. MCNPX-PoliMi code is used to model a mix of plutonium and americium oxide in a large object. Then a post-processing macro based on ROOT simulates the realistic response of the acquisition system (real and accidental coincidences, energy threshold, energy resolution, time jitter, etc.). The attenuation estimation and the discrimination between prompt fission neutrons and neutrons from (alpha,n) reactions are obtained by multiplicity measurement. Then the neutron time-of-flight technique is performed to identify fission signatures and locate the source. Afterwards, plutonium mass could be evaluated.

Keywords: holdup, plutonium, neutron, time-of-flight, simulation

1. Introduction

Reprocessing, fuel fabrication, or enrichment require large facilities that contain many gloveboxes, tanks and pipes where plutonium can build up. Holdup measurement is essential to verify nuclear material inventories in nuclear process facilities, but also for safeguards, and criticality safety. Gamma-ray spectrometry based on the Generalized Geometry Holdup method, is widely used [1][2]. Another common NDA technique is based on neutron coincidence counting with large polyethylene-moderated ^{3}He slab detectors [2][3]. For these two methods, measuring the nuclear materials deposit challenges the capabilities and compromises the accuracy of measurement because of very large attenuation effects by equipment and deposits, and the necessary assumptions to describe the deposits (point, line, area, or volume). The paper presents the study of a system based on neutron time-of-flight event detection and fast coincidence counting for plutonium holdup measurement without any assumption on the source localization or distribution.

2. Measurement Principle

The principle is based on fast coincidence measurement between gamma rays and neutrons produced by spontaneous fissions and (alpha, n) reactions.

2.1. The spontaneous Fission and (alpha,n) Reaction

Two fast process can produce correlated gamma-ray(s) and neutron(s) :

- Fission : the fission fragments de-excitation takes place at early stage after scission through successive emission of neutrons and gamma-rays, namely the prompt fission neutrons and the prompts fission gamma-rays. Prompts fission neutrons and gamma-rays can be considered as simultaneously emitted. In average, 2.15 prompt neutrons and 6.99 prompt gamma-rays are emitted per spontaneous fission of ^{240}Pu.

- (alpha,n) reaction : leads to the emission of one alpha particle and gamma rays from unstable daughters. The alpha particle can produce correlated gamma-ray(s) - neutron through (alpha,n) reaction with light elements. One neutron and at least one gamma-rays are accompanying reactions on the light nuclei (example: $\alpha + {}^{18}O \rightarrow n + {}^{21}Ne^{*} \rightarrow \gamma + {}^{21}Ne$). Neutron and gamma-ray(s) can be considered simultaneously emitted [4].

2.2. Measurement Principle

The system concept considered consists of many plastic scintillators (slave detectors) and one NaI(Tl) scintillator (trigger detector) placed around the inspected object, as showed in Fig.1. The slave detectors and the trigger detector are located around the inspected object without any knowledge of the nuclear material localization or distribution.

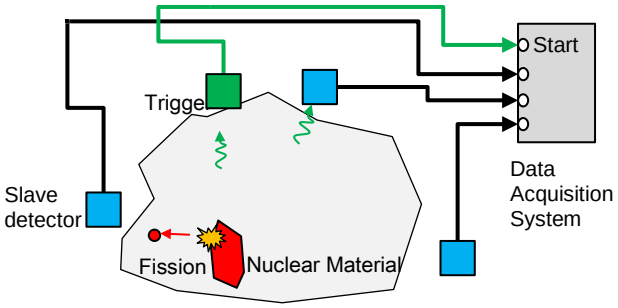


FIG. 1. Schematic drawing of the system concept.

Δ_{gamma} nanoseconds after the fission of the nuclear material, if at least one gamma-ray reaches the trigger detector, the acquisition by the slave detectors starts. The time of flight (TOF) between the detection in the trigger and the slave detectors is measured. If Δ_{gamma} is small enough, the TOF can be considered equal to the time between the fission and the detection of a prompt fission particle.

The TOF measurement allows the discrimination between gamma-rays and neutrons. For example, for the TOF-distribution presented in Fig. 2, gamma-rays TOF is distributed between 0 and 15 ns, whereas neutrons TOF is distributed between 15 ns and 150 ns.

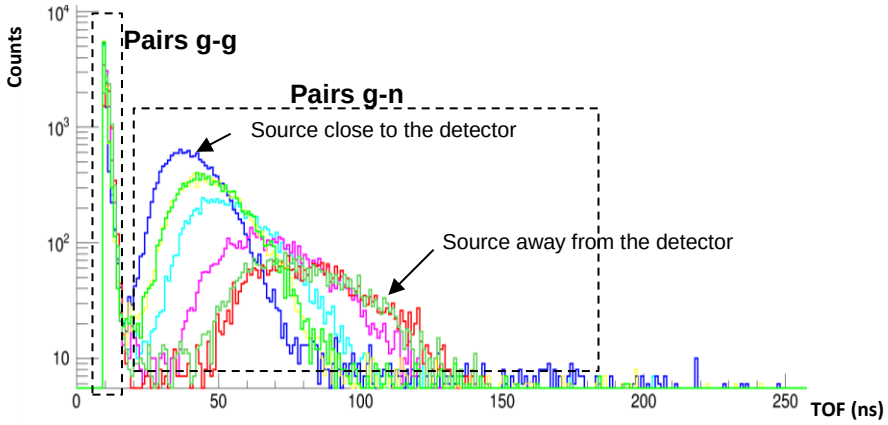


FIG. 2.Example of Time-of-Flight distribution (simulation with a punctual source / detector distance between 30 cm and 80 cm).

In addition, the system must discriminate neutron(s) produced by (alpha,n) reaction and fission. The speed difference between (alpha,n) neutrons and fission neutrons is not large enough to allow a discrimination. But multiplicity is pertinent to discriminate both reactions: (alpha,n) reaction produces only one neutron per reaction whereas fission produces many neutrons. Thus, only coincidence between one gamma-ray (in the trigger detector) and at least two neutrons (in slave detectors) is associated with a fission event.

For the source(s) localization, the volume of inspected item is meshed in voxels. When one gamma + 2 prompts fission neutrons are detected, for each voxel, the steps described below are done:

1. The distance between the voxel and the activated slave detectors are calculated;
2. The speed and the energy of the neutrons are calculated;
3. The probability of emission of prompt neutrons with these energies are calculated;
4. The probability to measure the energy deposits are calculated;
5. Based on these probabilities of emission and detection, the probability of presence of the source within the tested voxel is calculated and incremented.

This process is repeated each time prompts fission neutrons are detected. It allows source(s) localization without any assumption on the source(s) distribution(s) or location(s). After this first step, each voxel has a weight proportional to the probability of presence of the source. Nuclear material is supposed to be present in voxels having the highest probability of presence. Then, the localization of these voxels are used to calculate TOF distribution based on the following equation :

$$C^{i}{}_{gn}(tof) = \sum_{vox=0}^{N} Fiss_{vox} \times \overline{\nu_{gn}} \times Att_{g} \times Att_{n} \times \left(\varepsilon^{trig}{}_{geo} \times \varepsilon^{trig}{}_{int}\right) \times \left(\varepsilon^{i}{}_{geo} \times \varepsilon^{i}{}_{int} \times Prob^{i}(tof)\right)$$

With

- $C^{i}{}_{gn}(tof)$ is the number of coincidences between one gamma-ray detected in the trigger detector and one prompt fission neutron in the slave detector *i*, with a time of flight equal to *tof*;
- $Fiss_{vox}$ is the number of fissions in the voxel *vox*;

- $\overline{\nu_{gn}}$ is the average number of gamma-neutron pairs per fission ;
- Att_g and Att_n are the attenuations of gamma-rays and neutrons;
- ${\varepsilon^{trig}}_{geo}$ and ${\varepsilon^{i}}_{geo}$ are the geometric efficiencies of the detectors (trigger or slave number *i*);
- ${\varepsilon^{trig}}_{int}$ and ${\varepsilon^{i}}_{int}$ are the intrinsic detection efficiencies of the detectors (trigger or slave number *i*);
- $Prob^{i}(tof)$ is the probability to detect one neutron "*tof*" nanoseconds after the detection of an associated prompt gamma-ray. This probability depends on the energy distribution of the prompt fission neutrons described by the Watt spectra.

The coefficients $Fiss_{voxel}$ are calculated based on comparison between calculated and measured TOF distribution for many sources distributions. Among tested distributions, only the one which leads to the best Khi2 minimization is selected and supposed to be close to the real source distribution and activity. For this study, attenuations of gamma-rays and neutrons is neglected (coefficients Att_g and Att_n are set at 1, i.e. no attenuation). The total number of fissions is estimated as follows:

$$Fiss_{tot} \approx \sum_{vox=0}^{M} Fiss_{vox}$$

With M the voxels where nuclear material has been detected. Thus, the ^{240}Pu equivalent mass estimation is proportional to the total number of calculated fission events. In the same way as neutron counting, the Pu mass is calculated taken into account the Pu isotopy.

3. Simulation

3.1. MCNPX-PoliMi and Post-Processing

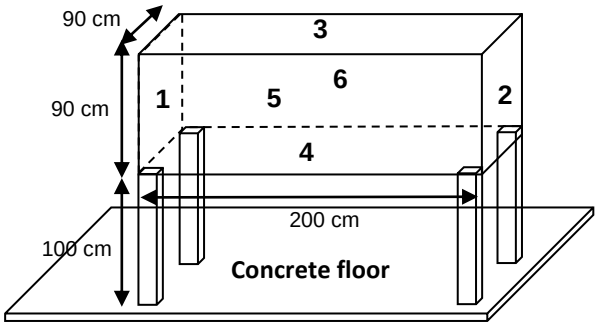
MCNPX is a Monte Carlo code used for the simulation of neutrons and photons transport [5]. The PoliMi extension [6][7] simulates particles interactions one by one. Neutrons and photons multiplicity distributions and correlations are implemented for nuclear reaction, such as fission and (alpha,n) reaction. The code output gives information about each collision within cells defined as detectors. A first post-processing macro based on the ROOT analysis platform [8] treats MCNPX-PoliMi outputs and models specific detector characteristics:

1. It converts the energy deposit of each particle's interaction in detectors (in MeV) in light output (in MeVee) [9][10];
2. It smears the light output by the energy resolution and applies an energy threshold;
3. It applies a time resolution;
4. It randomly mixes in one pulse train the different sources events (fissions, (alpha,n) reactions, beta reactions, etc.).

Once the pulse train of a realistic system obtained, a second post-processing macro, also based on ROOT, applies the steps described in the previous paragraphs for source(s) localization and Pu mass calculation.

3.2. Setups Description

A generic "light" glovebox and a generic "shielded" glovebox have been defined. Fig. 3 shows a description of gloveboxes.



Faces	Light glovebox	Shielded glovebox
1, 2, 3, 4	1 cm of stainless steel	0.47 cm of stainless steel 1.27 cm of lead 8.89 cm of WEP* 0.47 cm of stainless steel
5, 6	1 cm of acrylic glass	0.95 cm of safety glass 6.35 cm of acrylic glass 2.54 cm of leaded glass 0.32 cm of safety glass

* WEP shielding material consists of a water/ethylene glycol mixture, and a boric acid/sodium hydroxide mixture

FIG. 3. Gloveboxes models.

Glovebox are inspected by a system composed of 12 slave detectors (plastic scintillators 20×20×10 cm^3) and 1 trigger detector (NaI(Tl) scintillator 20×20×10 cm^3). The number of detectors placed around the glovebox is limited, due to the presence of other instruments or other gloveboxes:

- Faces 1 and 2: No detectors are installed
- Faces 3 and 4: Only 2×2 slave detectors are installed
- Faces 5 and 6: 2×4 slave detectors are installed + 1 trigger detector

In order to limit the size of the MCNPX-PoliMi outputs to maximum value of 10 Go, only 1 hour of measurement and an amount of 500 g of 12 years old weapon grade plutonium (PuO_2 + AmO_2) is simulated. The modelled setup is presented in Fig. 4.

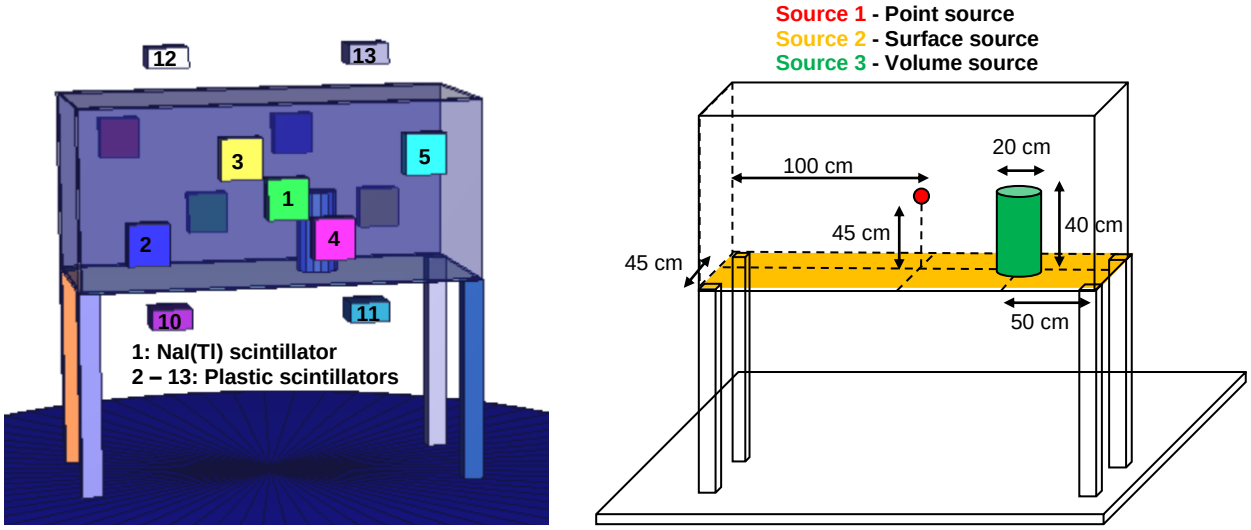


FIG. 4. View of detectors distribution around the inspected glovebox, and sources distributions.

To simplify the simulation, many assumptions are made in the paper:

- Only radiations produced by PuO_2 and AmO_2 through spontaneous fissions and (alpha,n) reactions are simulated;
- β-decays are not simulated;
- Induced fissions are not simulated;
- The external background is not simulated.

3.3. TOF distributions results

Setup and source previously described are simulated by MCNPX-PoliMi. The code output is processed by a first ROOT macro to convert the simulated output to a realistic pulse train. The TOF distributions between detector n°1 (the trigger detector) and other detectors, resulting from this first post-processing are presented in Fig. 5.

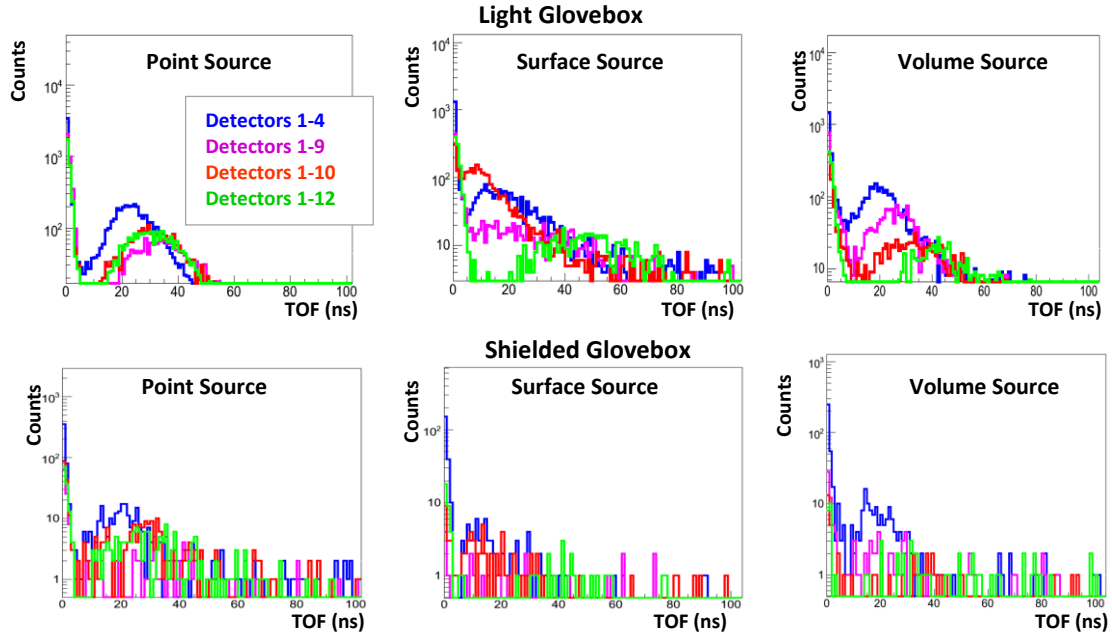


FIG. 5. Impact of the source distribution on the TOF distributions for coincidences between detector 1 (master) and 4, 9, 10 and 12 (simulation setup: 500 g of Pu within the inspected glovebox 1, 1 hour of measurement). Only coincidence g-n-n are used.

The gamma-rays TOF is distributed between 0 ns and ~5 ns, whereas neutrons TOF is distributed between ~5 ns and 100 ns. The shape of the "neutron part" depends on the distance between the source and the detector, and the source distribution. Due to gamma absorption and neutron slowing down by radiological protections of the shielded glovebox, few coincidences are recorded and TOF distributions are difficult to read. In addition, in the case of the surface source, detectors 9 and 10 are closed to the source and so gamma-rays and neutrons are difficult to discriminate based on TOF measurement.

Once the output of a realistic system is obtained, a second post-processing macro estimates the source localization and the Pu mass.

3.4. Source localization, TOF distributions deconvolution and mass calculation

The inspected volume is divided in 1620 voxels (1voxel = 10×10×10 cm^3) [11], and processes described previously are applied. The best agreement between calculated and measured TOF distributions, which lead to the source localization, are presented in Fig.6, 7 and 8, and the associated localization are presented in Tab. 1.

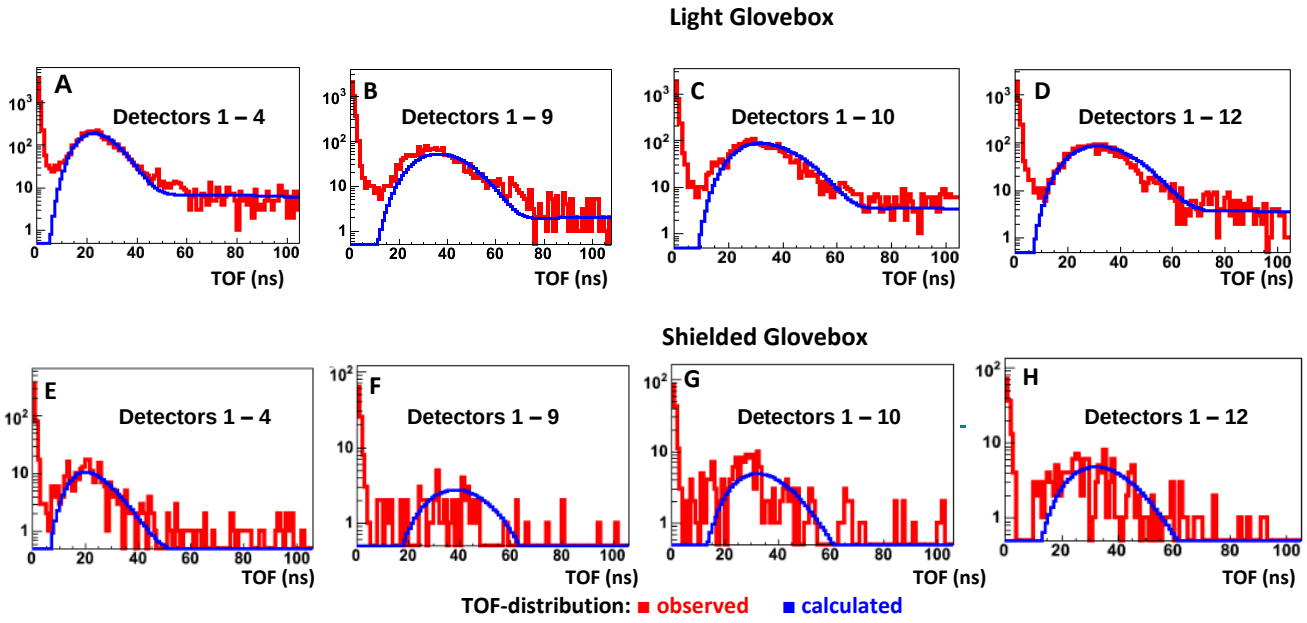


FIG. 6. Comparison between the calculated TOF distributions for the point source.

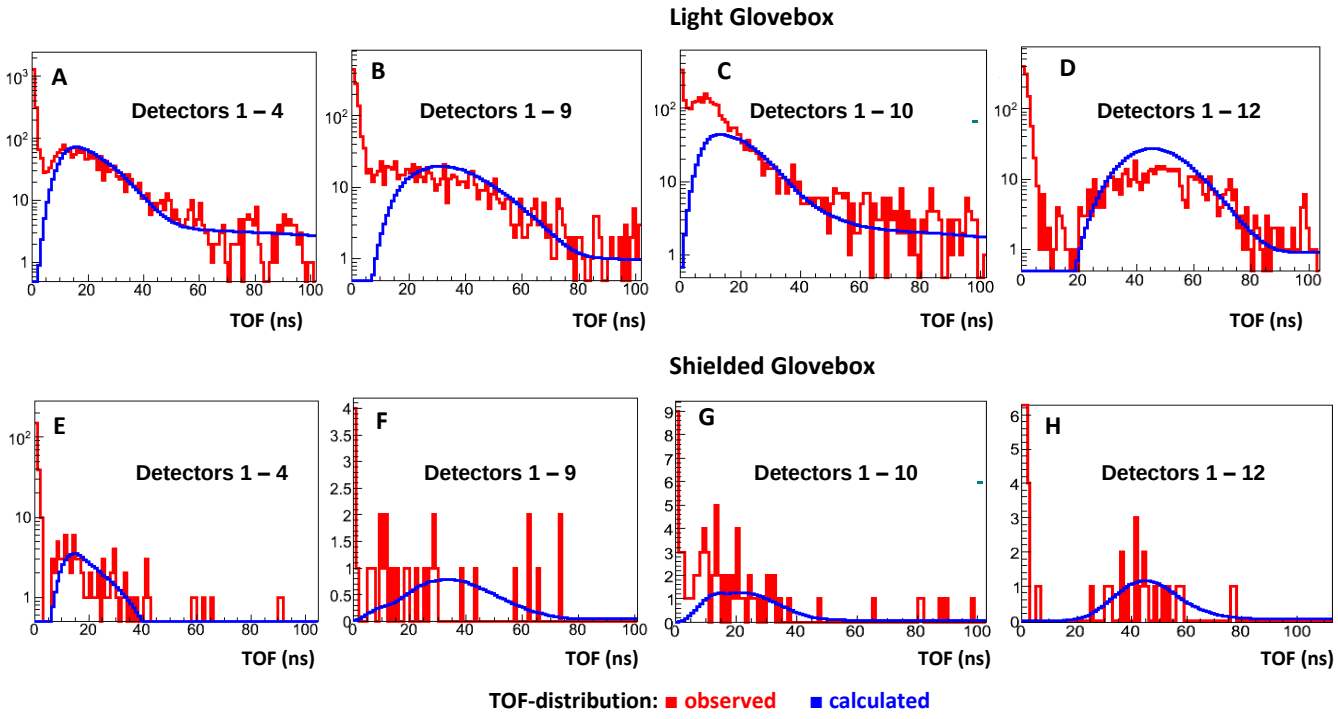


FIG. 7. Comparison between the calculated TOF distributions for the surface source.

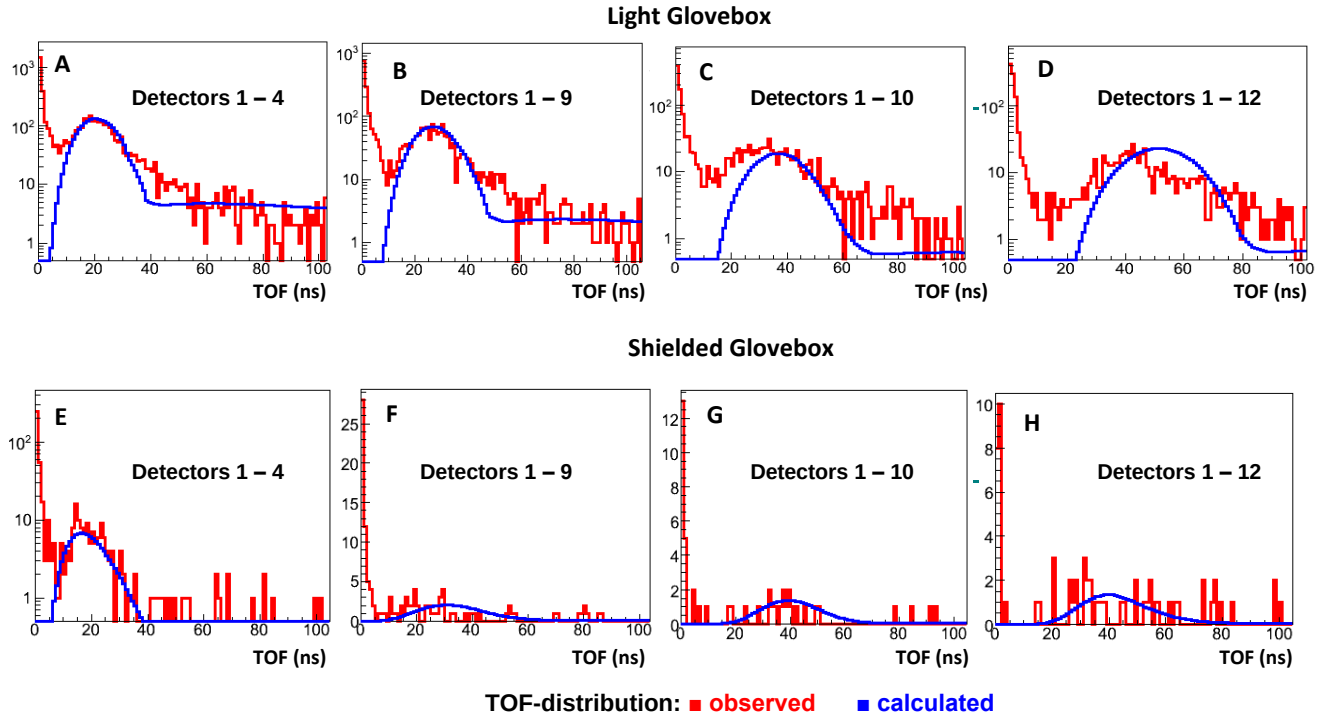


FIG. 8. Comparison between the calculated TOF distributions for the volume source.

If there are enough counts in the neutrons part of the TOF distribution, and if gamma/neutrons can be discriminated based on their TOF, there is a good agreement between calculated and observed TOF distributions. The fit between calculated and observed TOF distributions lead to the source localization in Tab. 1.

TABLE 1. SOURCE LOCALIZATION WITHIN GLOVEBOXES

Source	Simulated localization (cm)	Calculated localization (cm)	
		Light glovebox	Shielded glovebox
1 (point)	X = 0.0 Y = 0.0 Z = 0.0	X = [- 25, + 25] Y = [- 10, + 10] Z = [- 30, + 30]	X = [- 35, + 35] Y = [- 20, + 20] Z = [- 30, + 30]
2 (surface)	X = [- 100.0, + 100.0] Y= [- 45.0, + 45.0] Z = - 45.0	X = [- 65, + 65] Y = [- 40, + 30] Z = [- 40, - 20]	X = [- 65, + 85] Y = [- 40, + 40] Z = [- 40, - 10]
3 (volume)	X = [+ 14.0, + 35.0] Y = [- 10.0, + 10.0] Z = [- 34.0, +15.0]	X = [+ 15, + 45] Y = [- 10, + 10] Z = [- 40, + 10]	X = [- 5, + 65] Y = [- 30, + 20] Z = [- 30, + 20]

Even if the source distribution is not perfectly calculated, the localization of the source is coherent for the two gloveboxes and the three sources. But the dispersion of the source clearly increases in the case of the shielded glovebox. This bias is due to the organic shield:

- Some neutrons loose most of their energy and/or are captured by biological protections and cannot be detected;
- Some neutrons are slowed down (but can be detected), and so their speed distribution is different from the expected distribution described by the Watt distribution. This unknown speed distribution biases the source localization.

In addition, in the case of the surface source, some detectors are too closed from the Pu and gamma/neutrons cannot be discriminated based on TOF measurement. This bad discrimination hid the presence of Pu on the border of the glovebox.

The poor statistics -in the case of the shielded glovebox measurement- have an important impact on the estimation of attenuation coefficients and source distribution. These estimations lead to an important error on the Pu mass estimation. Table 2 shows the estimation of the Pu mass.

TABLE 2. PLUTONIUM MASS CALCULATION (EXPECTED MASS = 500 g)

Source	Light glovebox		Shielded glovebox	
	Pu mass (g)	Error (%)	Pu mass (g)	Error (%)
1 (point)	561	+ 12 %	40	- 92 %
2 (surface)	514	+ 3 %	16	- 97 %
3 (volume)	459	- 8 %	27	- 94 %

In the case of the light glovebox, the good source localization and the low attenuation of radiations lead to a good estimation of the Pu mass. But in the case of the shielded glovebox, the over-estimation of the source dispersion and the under-estimation of attenuation lead to an important underestimation of the Pu mass.

4. Conclusions

The paper shows the interest of measuring holdup using the combination of neutron time of flight and gamma-neutron coincidences measurement techniques. If the attenuation of neutrons and gamma rays are negligible, it is possible to estimate the localization and the Pu mass without any prior knowledge. Even if the outside background is neglected, this study used realistic assumptions and shows the limits of the technique:

- If organic material is present, many neutrons are thermalized, which biases the results.
- Additional simulations (not presented in the paper) without detectors below and above the glovebox were made. The results show that the source localization is possible only if most of the faces of the inspected object can be instrumented.
- The precision of TOF measurement has an important impact on the results. The electronics must have nanosecond resolution.

Despite these limits, the technic presents a great interest for the inspection of large objects without organic materials or radiological protection. The calculated uncertainty is close to uncertainties of already existing systems. Moreover, additional studies will be performed to improve the Pu mass calculation:

- Neutron and gamma attenuations coefficients will be calculated by simulation for generic gloveboxes;
- New algorithm will be developed to localize the source(s), and deconvoluate the TOF-distributions;
- New detectors distribution will be tested to optimize the detection efficiency and gamma/neutron discrimination based on TOF measurement;
- Small detectors will be tested and placed inside glovebox through glove ports.

5. References

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