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Adrien Sari, Frédérick Carrel, Frédéric Lainé, A. Lyoussi. Design of a neutron interrogation cell based on an electron accelerator and performance assessment on 220 liter nuclear waste mock-up drums. IEEE Transactions on Nuclear Science, 2014, 61 (4), pp.2144 - 2148. 10.1109/TNS.2013.2288240 . hal-01894714

HAL Id: hal-01894714

<https://hal.science/hal-01894714>

Submitted on 25 Jul 2023

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Design of a neutron interrogation cell based on an electron accelerator and performance assessment on 220 liter nuclear waste mock-up drums

Adrien Sari, Frédérick Carrel, Frédéric Lainé, Abdallah Lyoussi

Abstract—Radiological characterization of nuclear waste drums is an important task for the nuclear industry. The amount of actinides, such as ^{235}U or ^{239}Pu , contained in a package can be determined using non-destructive active methods based on the fission process. One of these techniques, known as neutron interrogation, uses a neutron beam to induce fission reactions on the actinides. Optimization of the neutron flux is an important step towards improving this technique. Electron accelerators enable to achieve higher neutron flux intensities than the ones delivered by deuterium-tritium generators traditionally used on neutron interrogation industrial facilities.

In this paper, we design a neutron interrogation cell based on an electron accelerator by MCNPX simulation. We carry out photoneutron interrogation measurements on uranium samples placed at the center of 220 liter nuclear waste drums containing different types of matrices. We quantify impact of the matrix on the prompt neutron signal, on the ratio between the prompt and delayed neutron signals, and on the interrogative neutron half-life time. We also show that characteristics of the conversion target of the electron accelerator enable to improve significantly measurement performances.

Index Terms—Electron accelerator, neutron interrogation, photoneutron, nuclear waste drum, uranium.

I. INTRODUCTION

RADIOLOGICAL characterization of nuclear waste drums is an important task for the nuclear industry. One of the main steps of such characterization studies consists in determining the amount of actinides contained in the drums. Non-destructive methods based on the fission process are often required to address this issue. Passive non-destructive methods consist in detecting prompt and delayed particles spontaneously emitted by the actinides contained in the drum. However, these methods can be limited by strong background noise or self-attenuation in the drum. Non-destructive active methods are based on two steps. In the first step, the drum is irradiated with a beam of neutrons or photons in order to induce fission or photofission reactions. In the second step, prompt and delayed particles emitted following the fission or photofission reactions are detected. Neutron interrogation [1]-[3] commonly uses a deuterium-tritium neutron generator whereas photon interrogation [4]-[6] requires the use of a

photon beam produced by Bremsstrahlung in the conversion target of an electron accelerator.

Neutrons commonly referred to as “photoneutrons” are produced by photonuclear reactions in the target of an electron accelerator. Photoneutrons are seen as spurious particles during photon interrogation measurements. However, previous studies have shown the possibility of carrying out neutron interrogation measurements using photoneutrons generated by an electron accelerator [7]-[11]. One of the main advantages of this method is that the intensity of the photoneutron flux reached can be on the order of 10^{11} neutrons per second [12], [13]. Moreover, an electron accelerator enables to improve nuclear waste characterization studies by combining neutron interrogation, photon interrogation, and high-energy imaging, on a single facility. In a previous paper, we characterized the photoneutron flux produced by two electron accelerators [14]. In this new study, we design an electron accelerator-based neutron interrogation cell with MCNPX [15], [16], and perform photoneutron interrogation measurements on uranium samples positioned at the center of 220 liter nuclear waste mock-up drums containing different types of matrices. We also show impact of the photoneutron source on measurement performance.

II. DESIGN OF A NEUTRON INTERROGATION CELL BASED ON AN ELECTRON ACCELERATOR

We designed a neutron interrogation cell based on an electron accelerator by MCNPX simulation. A scheme of such a cell is shown in Fig. 1. Our study is divided into three parts: the cavity of the cell, the neutron source, and the detection blocks. Six performance criteria were retained in this study: the fission rate in a uranium sample placed in the cell, the interrogative neutron half-life time, the half-life time of fast neutrons in the detection blocks, the absolute detection efficiency of prompt neutrons from thermal fission, the absolute detection efficiency of delayed neutrons from thermal fission, and the absolute detection efficiency of delayed neutrons from photofission. Among the main requirements set, we emphasize on the fact that the cell is designed to enable to:

- carry out measurements on 220 liter nuclear waste drums (cylindrical shape, 60 cm of diameter and 80.9 cm of height, steel thickness of 0.9 mm);
- move rapidly the drum between the photon and the neutron interrogation measurement positions;
- keep the same neutron detection blocks for both the neutron and the photon interrogation measurements.

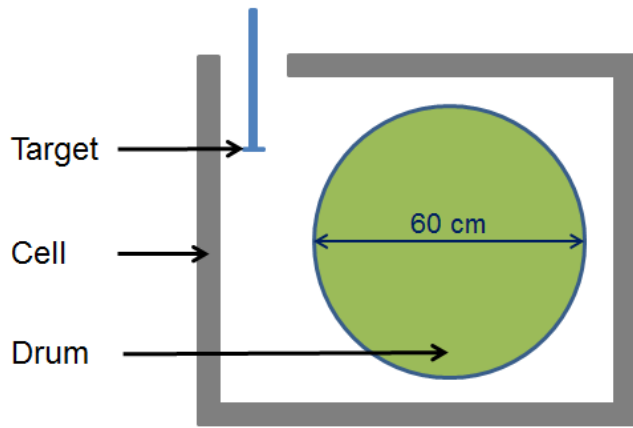


Fig. 1. Scheme of an electron accelerator-based neutron cell.

As a D-T generator emits 14 MeV neutrons and since the mean energy of photoneutrons produced by an electron accelerator is 1 MeV [14], we compared the performance of the polyethylene cell obtained with an electron accelerator and a D-T generator. Fig. 2 shows the obtained results (F4 tallies in MCNPX, isotropic neutron sources). It is important to notice that, on the latter and the following figures, error bars are not visible as they are smaller than symbols. We can see that the performance achieved with the electron accelerator is more than three times higher than the one obtained with the D-T generator. This could be due to the fact that 14 MeV neutrons are more likely to escape from the measurement cell as they interact deeper in the polyethylene walls than the 1 MeV photoneutrons. However, a 10 cm thick tungsten converter around the D-T generator could make its response curve similar to the one obtained with the electron accelerator.

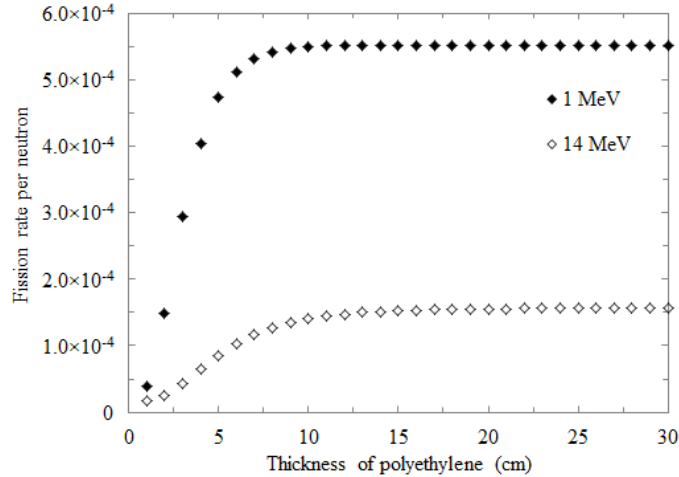


Fig. 2. Comparison of the performances of a polyethylene interrogation cell obtained with an electron accelerator or a D-T generator.

Fig. 3 shows the fission rate in the uranium sample according to the thickness of the walls of the cell. We compared results obtained considering walls made of polyethylene or graphite. For a polyethylene cell, we can see that the fission rate reaches its highest value for 10 cm thick walls. Large walls of graphite are necessary to achieve a

higher fission rate. Therefore, we chose to make our interrogation cell with polyethylene as this material is a good compromise in terms of mass, cost, and performance.

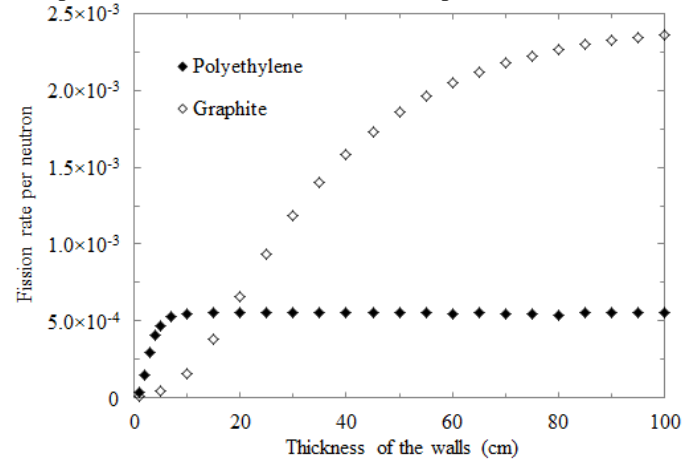


Fig. 3. Optimization of the thickness of the polyethylene or graphite walls in terms of fission rate.

In order to keep the same detection blocks for both the neutron and the photon interrogation measurements, we chose to place detection blocks against both sides of the cell. In such a way, the polyethylene cell would be easily removed when moving the drum to the photon interrogation position. Fig. 4 shows the evolution of the method sensitivity, proportional to the product between the absolute detection efficiency and the fission rate in the uranium sample (F4 tallies), according to the thickness of the polyethylene walls. This example is given for the case in which the sample is positioned at the center of the drum in a 0.22 g/cm³ iron matrix. We compared results obtained for the prompt neutrons and the delayed neutrons from thermal fission. We can see that respectively 3 cm and 4 cm thick walls enable to maximize the product for the delayed neutrons and the prompt neutrons. Several configurations have been investigated according to the composition and the density of the matrices. Finally, we chose to make our polyethylene cell with 5 cm thick walls, which corresponds to a satisfying tradeoff.

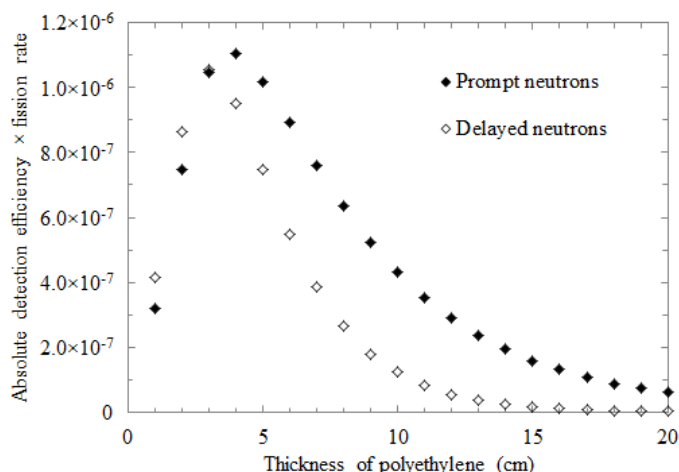


Fig. 4. Optimization of the method sensitivity adjusting the thickness of the polyethylene walls. Simulation results obtained considering a 0.22 g/cm³ iron matrix.

III. PERFORMANCE ASSESSMENT ON 220 LITER NUCLEAR WASTE MOCK-UP DRUMS

We carried out measurements with the electron accelerator of the SAPHIR facility [5] at CEA Saclay, shown in Fig. 5, and the neutron interrogation cell built further to its design by MCNPX simulation. Internal dimensions of the neutron cell cavity are the following: 70 cm × 90 cm × 92 cm. The average value of the electron energy spectra was 17 MeV. Pulses of 2.5 μs duration were generated at the frequency of 50 Hz. The electron peak current was 100 mA. Due to budget constraints, we used detection blocks at our disposal (containing three ³He detectors each), even though these detection blocks are not optimized for being used with the neutron cell. During 10 min, we irradiated uranium samples positioned at the center of a 220 liter nuclear waste mock-up drum. Fig. 6 shows the experimental setup. We carried out measurements with different types of matrices: a 0.02 g/cm³ vinyl matrix, a 0.22 g/cm³ iron matrix, and a 0.45 g/cm³ polyethylene matrix. These matrices are respectively shown in Fig. 7, Fig. 8 and Fig. 9.



Fig. 5. The electron accelerator of the SAPHIR facility.

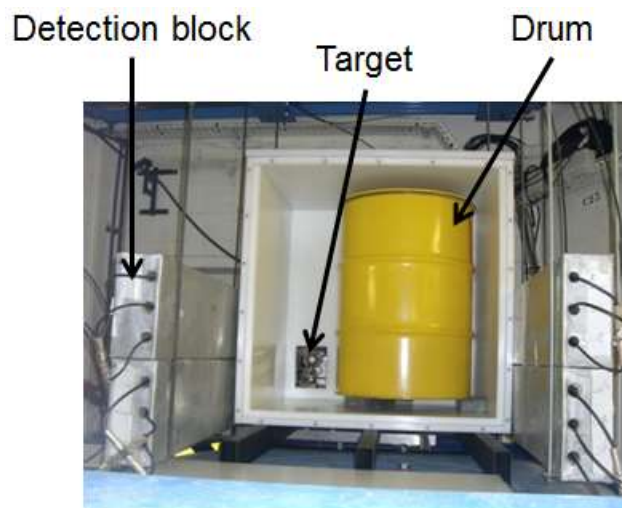


Fig. 6. Experimental setup (back of the cell removed).



Fig. 7. Mock-up drum containing a 0.02 g/cm³ vinyl matrix.



Fig. 8. Mock-up drum containing a 0.22 g/cm³ iron matrix.



Fig. 9. Mock-up drum containing a 0.45 g/cm^3 polyethylene matrix.

For this measurement campaign, we have used the neutron signal obtained with a single detection block (the nearest block from the centre of the drum). Examples of signals obtained are shown in Fig. 10. It is important to notice that the 0.45 g/cm^3 polyethylene matrix did not enable to achieve significant signals. Moreover, the background noise shown in Fig. 10 is the one measured without any drum in the cell. We detected prompt neutrons between 3 ms and 8 ms after each pulse, and delayed neutrons between 15 ms and 20 ms. Interrogative neutron half-life times have been deduced from the decay of the prompt neutron signals. Half-life values vary from $635 \mu\text{s}$ for the drum containing an iron matrix to $1026 \mu\text{s}$ for the cell without any drum. In comparison with the value of $1120 \mu\text{s}$ obtained for the neutron cell PROMETHEE 6 [2] located at CEA Cadarache, our electron accelerator-based neutron cell presents satisfying performances according to this criterion.

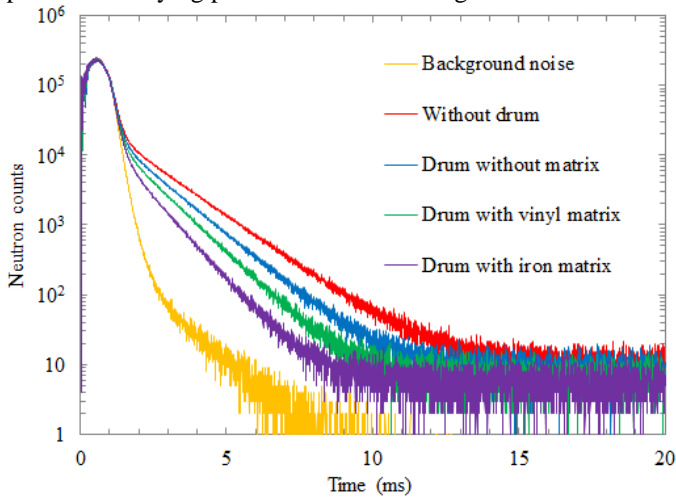


Fig. 10. Examples of neutron signals obtained with the different matrices.

Fig. 11 shows the prompt neutron signal obtained according to the ^{235}U mass for the different experimental configurations studied. We can see that measurement sensitivity decreases when the matrix density increases, which corresponds to the expected behaviour. We deduced the mass detection limits from the prompt neutron signals. We can detect 3 mg of ^{235}U in 10 min with only three ^3He detectors (without any drum).

When considering the drum without any matrix, or the drum with the vinyl or the iron matrix, this value respectively changes to 6 mg, 8 mg and 18 mg.

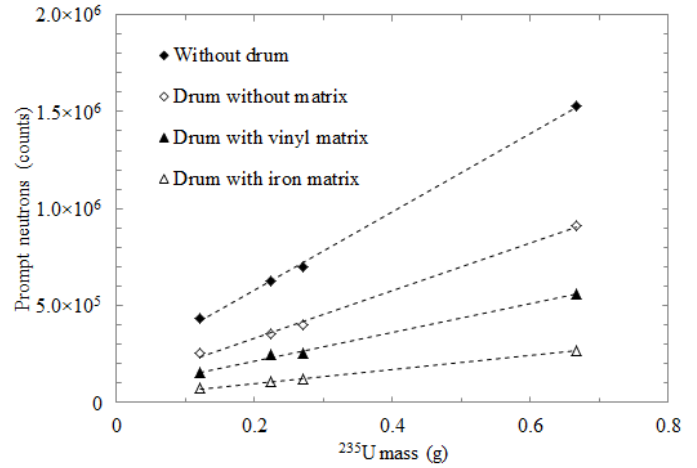


Fig. 11. Comparison of the prompt neutron signals obtained with the different matrices.

We also studied the ratio between the prompt and the delayed neutron signals. Results obtained are visible in Fig. 12. The value of the ratio, independent of actinide mass, varies significantly according to the matrix density. In the case of a real drum and knowing the matrix density, this value would enable to achieve information on the nature of the actinides.

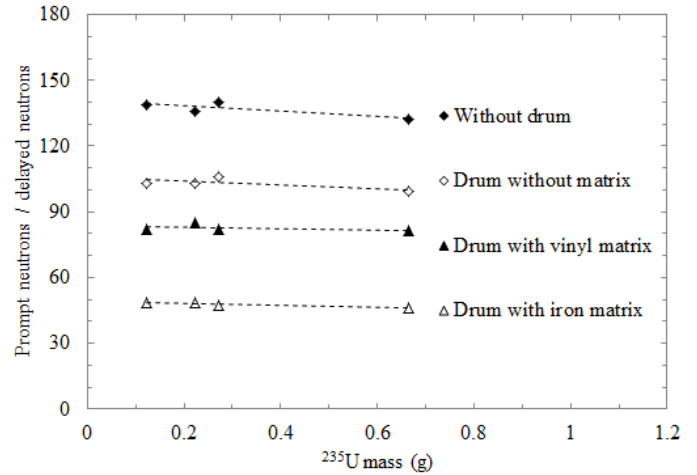


Fig. 12. Comparison of the ratio between the prompt and delayed neutron signals obtained with the different matrices.

IV. OPTIMIZATION OF THE PHOTONEUTRON FLUX AND PERFORMANCE ENHANCEMENT OF MEASUREMENTS

We carried out measurements with conversion targets made of tungsten or tantalum, targets of different thicknesses, and irradiated by either 14 MeV or 17 MeV electrons. Fig. 13 shows a few targets used in the frame of this measurement campaign.



Fig. 13. Tungsten or tantalum targets of different thicknesses.

Fig. 14 compares results obtained for a 1 cm thick tantalum target irradiated by 14 MeV or 17 MeV electrons. Signals obtained at 17 MeV are 3.2 times higher than the ones obtained at 14 MeV. We emphasized on the fact that this experimental result is in very good agreement with simulation results presented in a previous paper. Indeed, we obtained a factor of 3.2 between average emission intensities produced by a 1 cm thick tantalum target irradiated by 14 MeV or 17 MeV electrons [13].

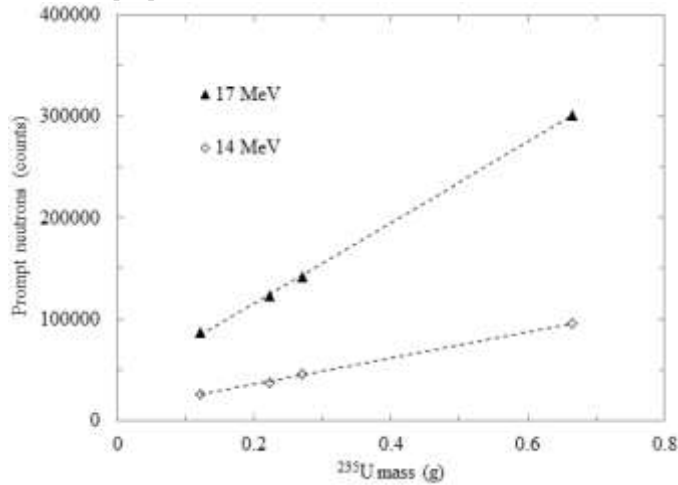


Fig. 14. Comparison of the prompt neutron signals obtained using a 1 cm thick tantalum target irradiated by 14 MeV or 17 MeV electrons.

Fig. 15 compares results obtained for a 1 cm thick tantalum or tungsten target irradiated by 17 MeV electrons. Signals obtained with the tungsten target are 14% higher than the ones obtained with the tantalum target.

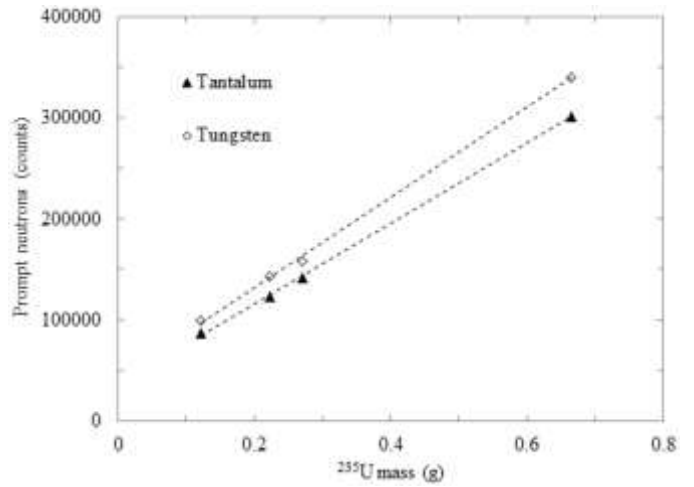


Fig. 15. Comparison of the prompt neutron signals obtained using a 1 cm thick target made of tungsten or tantalum and irradiated by 17 MeV electrons.

Fig. 16 compares results obtained for 5 mm to 4 cm thick tantalum targets irradiated by 17 MeV electrons. The 3 cm thick target enables to maximize the prompt neutron signal. This result enables to confirm experimentally previous simulation results which lead to the conclusion that a 3 cm thick target maximizes the average emission intensity of the photoneutron flux [13].

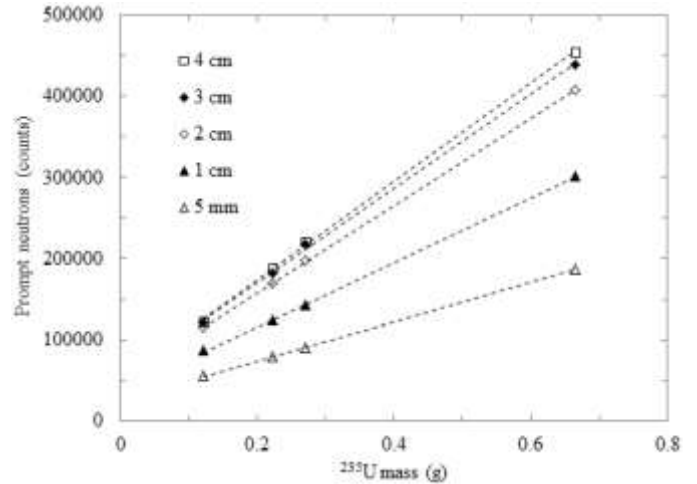


Fig. 16. Comparison of the prompt neutron signals obtained using a 5 mm up to 4 cm thick tantalum targets irradiated by 17 MeV electrons.

V. CONCLUSIONS AND OUTLOOK

We have designed an electron accelerator-based neutron interrogation cell intended to carry out measurements on 220 liter nuclear waste drums. This cell is characterized by an interrogative neutron half-life time of 1026 μs . We conducted neutron interrogation measurements on uranium samples placed at the center of 220 liter nuclear waste mock-up drums containing different types of matrices: vinyl, iron and polyethylene. We have quantified the impact of the matrices on the prompt neutron signal, the ratio between the prompt

and delayed neutron signals, and on the interrogative neutron half-life time.

Between 3 mg and 18 mg of ^{235}U can be detected in 10 min, depending on the experimental configuration. We have also shown that measurement performances can be improved significantly by increasing the electron energy and the thickness of the target. Based on our results, the use of a 3 cm thick target is recommended. Using optimized detection blocks, allowing detecting prompt neutrons earlier, would enable to expand mass detection limits even further.

Finally, our study demonstrates the possibility of building a platform, based on an electron accelerator and intended to characterize nuclear waste packages, which enables to perform neutron interrogation, photon interrogation, and high-energy imaging. Such a platform, combining these three complementary techniques, would enable to improve characterization of nuclear waste drums. The next step of this study would consist in evaluating the performance of such a cell on a real drum.

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