Concepts for dismantlement verification and neutron multiplicity measurements for plutonium mass attribute determination

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Abstract:

For meaningful and effective verification of nuclear warhead dismantlement, a combination of warhead authentication measures along with a robust chain-of-custody would be instrumental. In this context, requirements and concepts will be discussed that could tie together warhead authentication and chain-of-custody measures to a meaningful overall regime. For this purpose, it will be assessed at what stages information barriers and other technologies could be applied. This serves as an overall introduction into the topic of dismantlement verification.

A more technical part focuses on passive neutron multiplicity measurements with He-3 detectors for the purpose of verifying attributes that are helpful for authenticating warheads. With a given isotopic composition (e.g. deduced from gamma spectroscopy measurements), neutron multiplicity counting is suited for determining plutonium mass which is considered one attribute. Plutonium measurements have been performed in the PERLA laboratory in Ispra that were used to validate MCNPX PoliMi simulations. Preliminary simulation results will be presented.

Keywords: disarmament, verification, information barrier, neutron multiplicity counting

1. Arms Control Verification

As part of verified fissile warhead component inventory declarations and verified warhead dismantlement, two elements will most likely be key to a sound verification regime. On the one hand, this is the authentication of warhead (components). Authentication in this context is the process during an on-site inspection through which it is assessed by measurements whether a specific item is a nuclear warhead (or component). On the other hand, a robust Continuity of Knowledge, the other key element, could be defined as providing means to effectively demonstrate over a certain time or process, e.g. during warhead dismantlement, the unchanged identity of the treaty-accountable item (i.e. that it remains the same item) and the integrity (i.e. that no undeclared changes to the item occurred). Technologies relevant for providing a Continuity of Knowledge are for example seals and tags that can be applied on the items directly or their storage containers and monitoring equipment.

Due to the classified nature of the items under investigation, direct measurements for authentication purposes will most likely not be possible as they would reveal information that is considered sensitive for nonproliferation, national security and possibly other reasons. The use of information barriers could overcome this problem. An information barrier takes classified measurements but converts the results to an unclassified output (such as a binary yes/no signal) while protecting the sensitive data from the inspector's view. The range of possible measurement techniques includes some non-nuclear type measurements and gamma spectroscopy, though this paper will focus on neutron detection.

Recognizing that other reasonable approaches exist, issues relating to the attribute approach are considered here. In the attribute approach, the inspecting and host parties agree on a set of attributes that the items would be checked against and on an analysis algorithm. This set should be defined in a way that it allows for an assessment whether a declared warhead component is genuine. One of the attributes that could be considered is a threshold fissile mass.

The technical focus of this paper will be on authentication of dismantled fissile warhead components that would be stored in appropriate containers with the attention being directed towards passive neutron multiplicity measurements for determination of plutonium fissile mass. Neutron interactions will be shortly introduced, then plausible container configurations will be described to then analyze what physical interactions occur in the containers that have an influence on the analysis. This will be done by simulations that are also checked against experimental results.

Neutron Multiplicity Counting

The neutron flux emitted by a fissile sample is affected by a number of possibly unknown properties [1]:

- spontaneous fission rate (the goal of neutron multiplicity counting to deduce fissile mass)
- sample self-multiplication / variation across the sample, in particular through induced fission
- (α, n) reaction rate if oxides are present

Other properties can be eliminated by careful calibration and counter design or are small or constant as described in [1]**.** Given that spontaneous fission, multiplication and (α, n) reactions all have an influence on neutron emissions, the goal is to determine the spontaneous fission rate since – through the known fission rate per Pu-240 atom – the fissile mass can be deduced if the isotopic composition of the sample is known through other means, e.g. gamma spectroscopy.
Passive neutron multiplicity counting can separate the contributions of these three effects to the overall neutron emission without requiring representative reference materials. Multiplicity counting measures the multiplicity distribution (i.e. neutron correlations) and calculates three parameters (Singles, Doubles and Triples rate) so that the three unknowns can be solved. Initial determination of detector parameters can be done with a Cf-252 source alone. Multiplicity counting based on this calibration can be slightly biased because of a detector's different efficiencies between Cf-252 and Pu fission neutrons [2], but this effect is minor.

2. Technical Implications of Component Storage Containers

The determination of fissile mass using neutron multiplicity counting requires information on isotopic composition that could come from gamma spectroscopy measurements. Gamma rays are shielded in particular by materials with a high atomic number. This paper only looks at the influence of neutron shielding but it is acknowledged that gamma shielding can complicate the quantitative analysis significantly as well.

Neutron interactions

Neutrons can react via elastic or inelastic scattering as well as neutron-induced nuclear reactions. Elastic scattering slows them down and changes their direction. The average energy loss due to elastic scattering is given as $2E \cdot A/(A+1)^2$,where A is the mass number of the nucleus and E the energy of the neutron [3]. It is clear that the energy loss decreases with increasing atomic number A. The probability of many neutron-induced reactions drops off rapidly with increasing neutron energy and is usually high for low energy neutrons.

Containers

The container's purpose is mainly to ensure safety and radiation protection. It appears that the main requirements for container certification are criticality control, energy absorption (shielding), thermal insulation and fire resistance [4, 5]. Also, containment and impact limitation (shock mitigation) must be guaranteed [5, 6]. A range of certified containers exist. They might contain some lead shielding which is very relevant to gamma radiation, but less to neutrons. However, usually a material like Celotex is used which consists of hydrogen, carbon and oxygen contents. Celotex has a density of 0.24 - 0.29 g/cm^3 [5]. While this material is suited for absorbing neutron energy via elastic collisions, it does not naturally contain significant neutron poison such as boron.

The container used for pit storage at PANTEX, the US site for warhead production and the only site where they are dismantled, which hosts a warhead component storage site, is the AL-R8 container which uses Celotex.[1] Other containers that are also certified are the 9975 and AT-400 container.[2] The 9975 container can be seen in Figure 1.

For the purpose of analyzing the physical processes that occur in the container, total neutron counting will be looked at, bearing in mind that those physical processes could have an influence on the neutron multiplicity counting measurements. In this study, the 9975 container will be investigated as it has the thickest neutron moderator compared to the AT-400 and AL-R8.

Code check and simulations

At the PERLA Laboratory of the Joint Research Centre site in Ispra, Italy, neutron measurements have been performed on a 2861 gram PuO_2 powder sample (70% Pu-239 and 24% Pu-240 and other plutonium isotopes) that was placed inside the 9975 container. A neutron detector system was designed to enclose the container. A model of the measurement setup has been built in MCNPX PoliMi. Experimental data will be compared to the simulated results to serve as a check of the simulation code and the implemented detector and container geometry.

The detector consisted of 8 slabs (4 lateral slabs, 2 back slabs and 2 front slabs. Each of these slabs contained six He-3 detector tubes (cylindrical tubes with a 2.44 cm diameter and 35.8 cm lengths). The slabs were made of polyethylene to moderate the neutrons coming in. The geometries of the slabs were not identical but similar in shape. They were about 23.5 cm broad and 9 cm long. The detector (MCNPX PoliMi model) is sketched in Figure 2.

The experiment yields the singles count rate S=81765±302. In the simulated result, we obtain S=83070±289. This shows that the simulated count rate overestimates the experimental by 1.6%±0.5%. All errors are statistical. The simulated lies outside the experimental result with its uncertainty, but can still be considered very small. Therefore this MCNPX PoliMi model seems to be consistent with the experiment in this regard and is used for further simulations.

Since the ultimate goal is to authenticate nuclear weapon components, we simulate weapons-usable fissile material. The sample simulated is a 2 kg solid sphere filled with 95% Pu-239 and 5% Pu-240 metal (19.8 g/cm^3). The goal here is to simulate a sample somewhat similar/comparable to a weapon component so that conclusions drawn from the simulations can in general terms hold for assessing the physical processes that would occur during verification.

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[1] More information on the design of the AL-R8 is found in [5, 7, 8]

[2] More information on the design of the AT-400 is found in [7]. More information on the design of the 9975 container is found in [6, 9, 10].

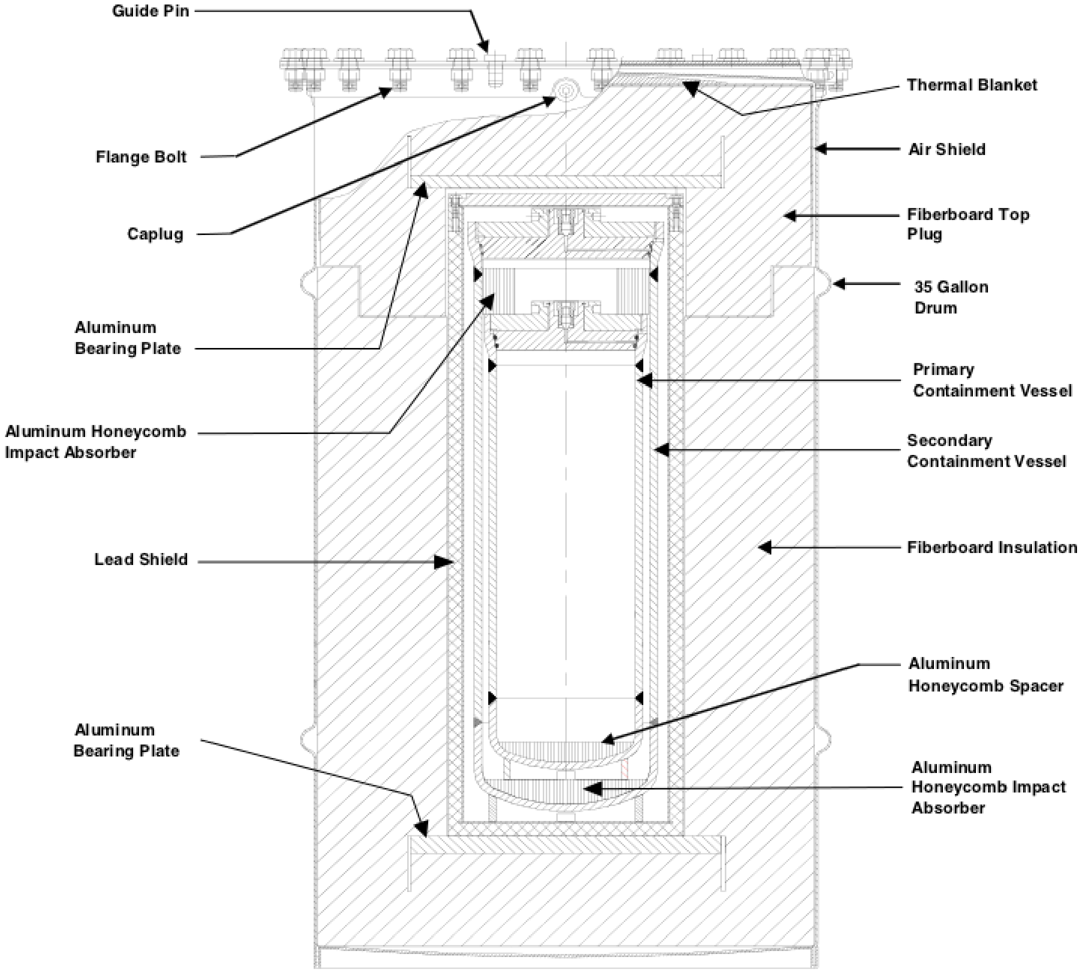

Figure 1: Sketch of 9975 container from [9]. The fiberboard insulation consists of Celotex. The Celotex component is 11.9 cm thick, the overall container has a diameter of 46.4 cm.

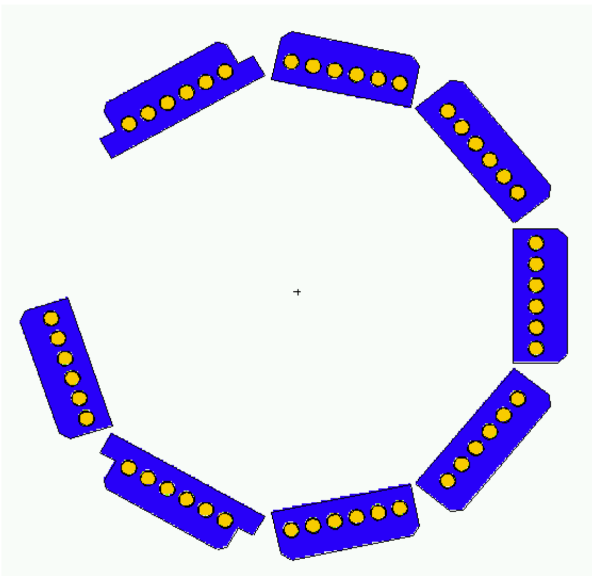

Figure 2 Cross-sectional sketch of the He-3 slab detector from the top. Polyethylene is depicted in blue; the He-3 tubes are marked yellow. The tubes have a length of 35.8 cm, items with a diameter less than 35.8 cm can be placed in the detector.

Regarding fissile material mass, 2 kg can be considered a reasonable fissile material quantity for a weapon, as for example discussed by Thomas Cochran in [11] who estimates that 2 kg weapon-grade plutonium is needed for a 10 kt yield weapon under a high technical capability. A solid sphere was chosen as opposed to a hollow sphere since the effect of multiplication becomes larger which is one of the physical processes that have an influence on and complicates the analysis of the measurement results. The rate of the neutrons emitted from spontaneous fission in this source is 1.02×10^5 n/s.

Simulations with the original 9975 container with Celotex are performed. For comparison, a simulation without the container was done. Furthermore, a simulation was run with 5 weight percent borated Celotex to study the effects of a neutron poison if it should be included in a container for whatever reasons.

Results

As can be seen from Table 1, the neutron detection events (through neutron capture) in the He-3 counter increase by 33% when the 9975 container is present. With 5% borated polyurethane, the count rate is still 22% above the rate without container, but 8% below that of the 9975 container with Celotex. Three different effects lead to this result and will be discussed, namely neutron moderation in the container, neutron capture in the container (with a negative effect on the count rate) and additional induced fission in the plutonium sample due to back-scattering from the container.

Neutron moderation

Celotex consists of low-Z materials that effectively reduce the neutron energies due to elastic scattering. This effect is in addition to elastic scattering that takes place in the detector polyethylene, so it further contributes to slowing down the neutrons to increase their detection probability in the He-3 detectors since the He-3 capture cross-section is high for thermalized neutrons, see Figure 4. This effect can be seen in Figure 3 which shows the significant increase of the neutron surface current in the thermal region. Also the configuration without container has a significant thermal contribution caused by the polyethylene which is part of the detector. It is however still clear that the moderation in the container increases the thermal surface current. Table 1 shows the surface current of neutrons below 1 eV and above 1 eV for the different configurations. It can be seen that the detection rate and the rate of neutrons below 1 eV are roughly proportional. Fast neutrons don't have a significant contribution to the detection rate. Thus the container moderation in addition to the polyethylene detector moderation results in an increase of the count rate.

configuration	Neutron detection rate in He-3 detector [1/s]	Neutrons < 1eV entering He-3 tubes [1/s]	Neutrons > 1eV entering He-3 tubes [1/s]
Bare component	1.18×10^4	2.15×10^4	5.56×10^4
9975 container with Celotex	1.57×10^4	2.87×10^4	5.06×10^4
9975 container with5% borated Celotex	1.45×10^4	2.65×10^4	5.08×10^4

Table 1: Neutron detection in the He-3 detector tubes for the different container configurations and neutron rates entering the He-3 tubes

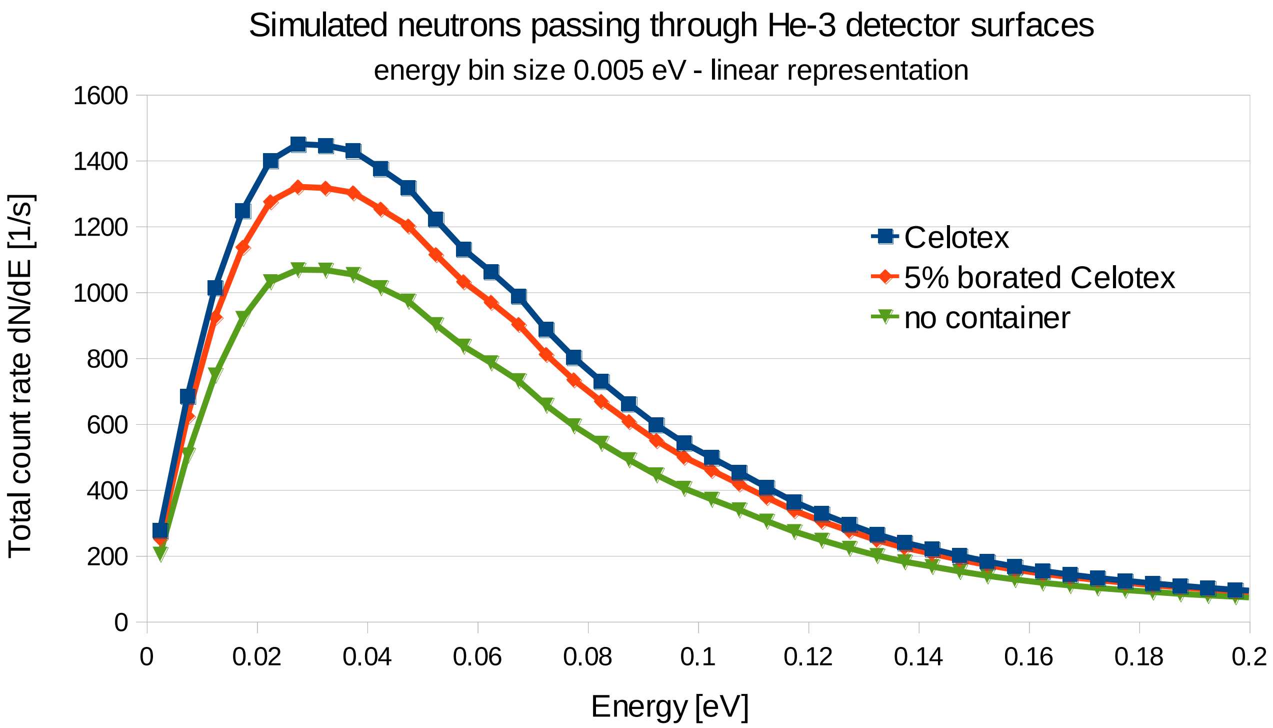

Figure 3: Energy spectrum of the low-energy neutrons (MCNPX PoliMi simulations) entering the He-3 tubes

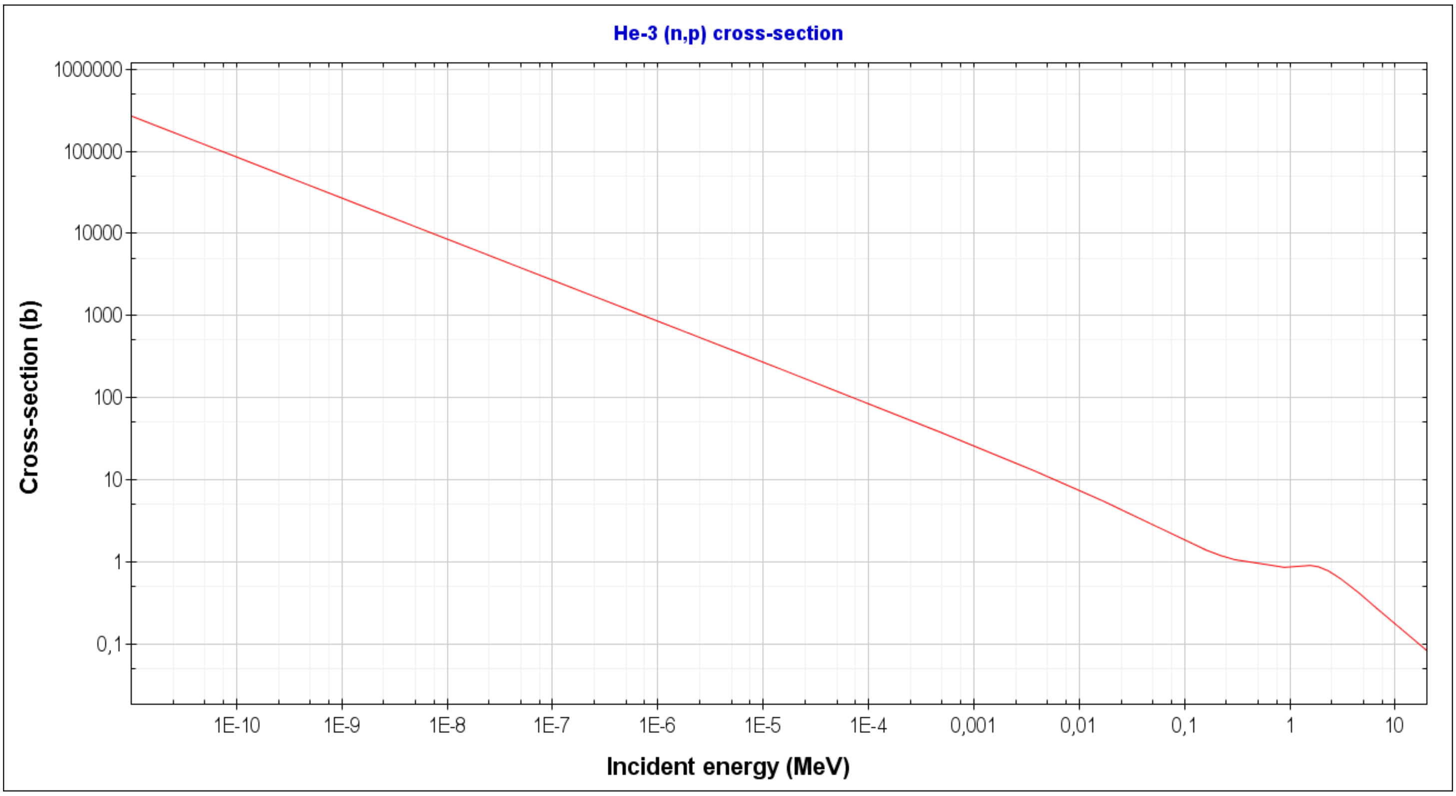

Figure 4: Cross-section of (n,p) reaction in He-3 from ENDF/B-VII.0

	Elastic scattering rate in container [1/s]	Neutron capture rate in container [1/s]
9975 container with Celotex	$1.38 \cdot 10^6$	$7.2x10^3$
9975 container with 5% borated Celotex	$1.02 \cdot 10^6$	$2.44x10^4$ (94% by ^{10}B)

Table 2: Elastic scattering and neutron capture events within the containers

Neutron capture in container

Besides elastic collisions, capture reactions occur in the container. Besides capture in Celotex component isotopes, in particular hydrogen, capture also occurs in concrete and stainless steel components of the container. The boron of the borated container has by far the most significant contribution to the overall capture rate. Neutron capture rates for the different container configurations can be compared in Table 2. One can see the large difference between the two configurations, which is due to the boron content which has the highest cross-section for neutron capture.

Looking at the neutron spectrum at the He-3 tube surfaces in Figure 3 and 4, the influence of neutron capture in the container can be seen: The thermal neutron rate of the borated container is 8% lower compared to the non-borated configuration. In the fast neutron energy region, neutron capture does not have any significant influence due to the low reaction cross-section; neutron moderation dominates here.

Reflection back into fissile material

Due to scattering, neutrons are scattered back from the container to the plutonium sample where they undergo additional interactions. The energy spectrum of the neutrons that are scattered back and enter the sample is shown in Figure 5. The reason why also the bare component-configuration has a lower but present thermal neutron contribution is that the detector polyethylene scatters back neutrons that can reach the sample.

The effect of back-scattered neutrons is additional induced fission by Pu-239 which creates additional neutrons on the one hand and neutron capture on the other. Looking at Table 3, it can be seen that the effect of induced fission outweighs the effect of neutron capture in terms of net neutron production. This results in a net increase of neutron multiplication. The neutron production rate from the borated Celotex container is rather high, this is under investigation.

	Neutron production rate from induced fission in fissile material [1/s]	Neutron capture rate in fissile material [1/s]
Bare component	1.02×10^5	1.5×10^3
9975 container with Celotex	1.06×10^5	1.7×10^3
9975 container with 5% borated Celotex	1.05×10^5	1.6×10^3

Table 3: Neutron gain from induced fission and neutron loss from capture in fissile material for different container configuration

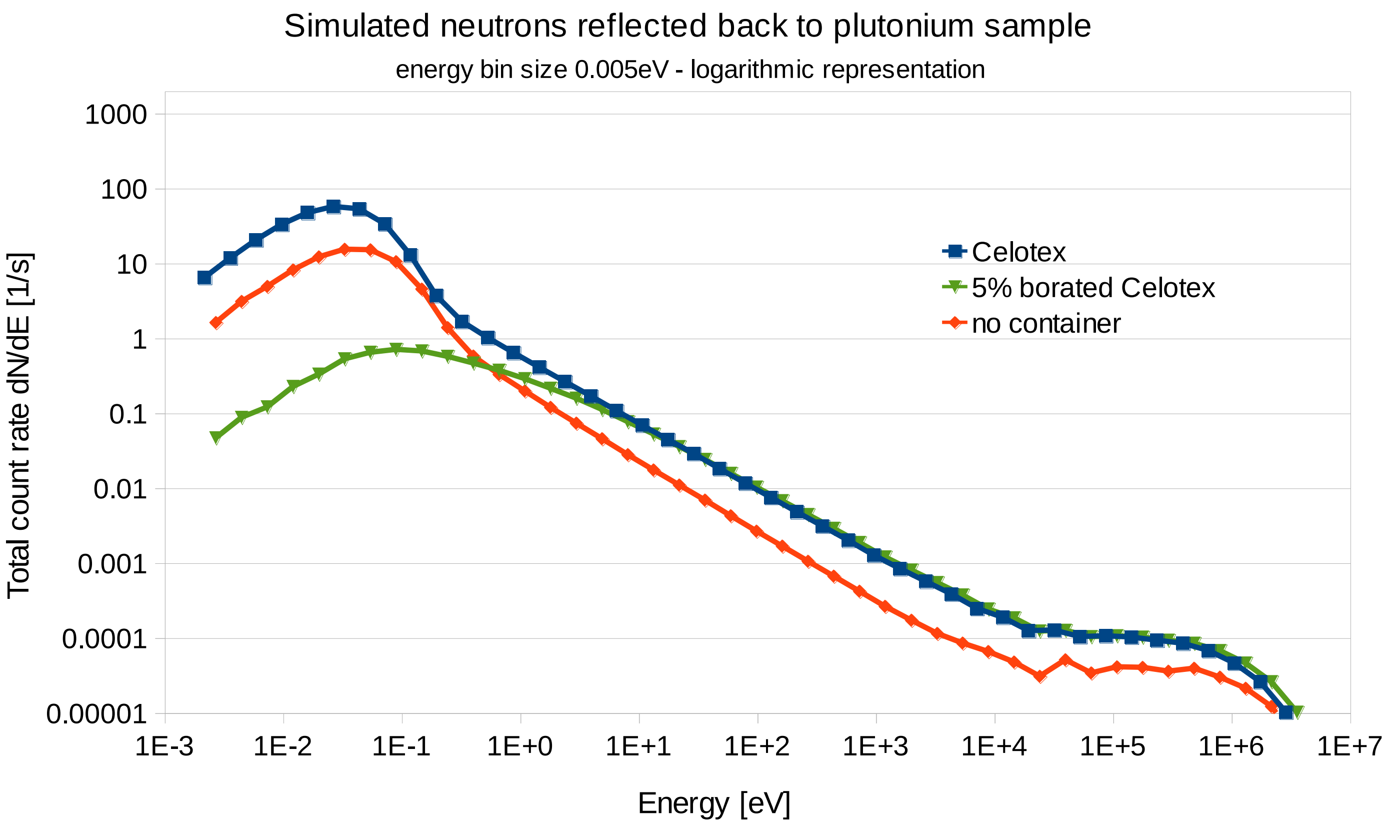

Figure 5: Logarithmic energy spectrum of all neutron (MCNPX PoliMi simulations) reflected back into fissile material for the different container configurations

3. Conclusion

This study has quantified the neutron processes that occur due the presence of the 9975 container containing a 2 kg weapon-grade plutonium sample. The relevant interactions were found to be neutron moderation in the container (see Fig. 3) that increases the count rate in a He-3 detector, neutron capture in the container that decreases the count rate (see Table 2) as well as reflection back into the fissile sample (see Table 3 and Fig. 5) where the neutrons induce further fission reactions.

These three effects have an influence on neutron multiplicity measurements that could be used for warhead component verification. In future, we shall study the effect of the presence of such a container on the automatic analysis behind an information barrier and to what extent it can decrease the reliability of the information barrier’s output.

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