SIMULATION OF THE PHOTON INTERROGATION OF NUCLEAR MATERIALS

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ABSTRACT

The detection of shielded highly enriched uranium is a challenging problem that is being addressed by numerous researchers. The measurement systems that are being investigated require fast, accurate, and simple read-out responses to be used at ports of entry. A number of currently proposed techniques make use of photon sources to induce fission in the nuclear material, and to detect the subsequent gamma rays and neutrons from fission. The design of such devices and the analysis of the measurement results rely on Monte Carlo codes to simulate the interaction of neutrons and photons with the nuclear material, the shielding, and the radiation detectors. However, currently available Monte Carlo codes lack the ability to accurately simulate the emission of correlated particles from photofission. Handling the whole photofission yield multiplicities and spectra is vital for implementing novel efficient techniques capable of identifying nuclear materials and estimating their amount. The goal of this study is to provide a tool that addresses and solves these deficiencies. We investigate methodologies that rely on the acquisition of correlated signals from prompt and delayed neutrons and photons emitted whenever a photofission event takes place. We are also evaluating the sensitivity and efficiency of organic scintillators for measuring the signals from impinging fast neutrons and photons.

In this paper, we describe the currently implemented photofission simulation in MCNPX, and propose an extension to improve the simulation of secondary particles. Our approach is based on the use of two existing and well-benchmarked codes, MCNPX and MCNP-PoliMi.

We present the application of the proposed method to determine the presence of nuclear material in luggage transported on a conveyor belt.

INTRODUCTION

Recent studies have been performed to investigate non-invasive inspections of cargos using photon sources [1,2]. For any fixed LINAC operational energy, delayed neutrons are identified in measurements in-between accelerator pulses on a basis of their time-dependent response. These delayed neutrons, originated by fission events, are separable in time from the prompt neutron signature. Therefore the detection of any gross delayed neutron emission is a direct indicator of the presence of nuclear material. It was proved that nuclear material identification is possible by comparing the delayed neutron emissions per beam coulomb measured at different electron beam energy operations [3].

Our project is concerned with the design of a photonuclear inspection device for non-intrusive detection of shielded nuclear material. The application is based on photon-induced fission for nuclear material detection and characterization, in particular highly enriched uranium (HEU). In this paper we will show that the analysis of the prompt radiation from photofission can be used to identify nuclear materials. The proposed active interrogation technology for this nuclear smuggling detection application relies on the ability of modern linear accelerators to generate very penetrating photons which induce photofission in fissile materials. A LINAC produces copious quantities of photons (~10^9 photons/s). The endpoint energy of the LINAC electrons is selected to maximize photon absorption in the target nuclei, thereby inducing photofission in fissile material. Photofission

is a consequence of the absorption of electric dipole radiation by the vibration of the neutrons against the protons and the subsequent damping of this motion into intrinsic excitation. This Giant Dipole Resonance phenomenon can be observed in every nucleus throughout the periodic table. Very little structure difference can be inferred in the energy dependence of the absorption cross-section, except for the lightest nuclei. Photonuclear interactions such as (γ,n), (γ,2n), (γ,f) have very close energy thresholds in the heaviest nuclei. For example, the minimum amount of photon energy required to induce a (γ,n) reaction is 5.3 MeV for ^{235}U, 6.1 MeV for ^{238}U, and 7.4 MeV for ^{208}Pb. Conversely, for a (γ,f) reaction we need 5.8 MeV for ^{235}U, 5.8 MeV for ^{238}U, and 27.4 MeV for the most abundant lead isotope, ^{208}Pb. It follows that the detection of neutrons and gamma rays does not constitute by itself a key factor for material identification and discrimination because of the similarity of the (γ,n) reaction threshold energies in actinides and in other high-Z materials. Instead, the fission energy barrier is much lower in the actinides than in other heavy elements. Our interrogation method is based on this physical property.

TECHNOLOGY OVERVIEW

The technique we propose infers the presence of concealed nuclear material by correlating the signals from pairs of particles that reach either detector within the pre-selected time window.
The interrogation device consists of an electron accelerator to produce pulsed Bremsstrahlung photon beams and a detection system, designed for scoring time-correlated events, that is sensitive to neutrons and gamma rays, as much as scintillators. Neutrons and gamma rays, reaching either detector within the selectable time window, are tagged as time-correlated. The Monte Carlo simulation generates four distinguished signals coming from four different particle pairs belonging to the same source history. The possible pairs are: n-n, n-γ, γ-n, and γ-γ. Since we assume the signals coming from neutron and photon interactions are indistinguishable in the measurements, then only a selection by different delays can be performed. In this paper we show that some of the signals, and/or a combination of them, carry enough information to identify unambiguously the concealed nuclear material. Both photoneutron and photofission events can take place in nuclear materials, whereas only photoneutron reactions can occur in other materials at the energy being used in our investigation. Photofission reactions, similarly to neutron-induced fissions, emit both neutrons, which are evaporated from the fission fragments, and gamma rays. The prompt neutrons can be slowed down and induce further fissions that generate more radiation emissions.
In this work we assume that the measurements are performed when the source beam is on. Then if fissile material is present, photofission events will occur and many prompt photons and neutrons will be produced and predominate over the delayed particle yield. In addition, the useful signals will be contaminated by the background noise due to the interaction of source gamma rays both with the object under investigation and the environment, but also then generating spurious and false correlated events as source photons reaching the detectors can be mistakenly time-correlated with secondary neutrons. If the measurements are performed when the source beam is off then the number of photofission events will be negligible. Both the time-correlated signals and the noise will be weak. In this case the signals from time-correlated particles is mainly due to reactions induced by photo-neutrons, which were generated when the beam was on, or by delayed neutrons from fission events that had taken place when the beam was on. In fact, neutrons can survive capture in the surrounding medium for times ranging from several micro-seconds to milliseconds.

DESCRIPTION

At the onset of this project we evaluated the most established and robust radiation transport Monte Carlo codes looking for a detailed implementation of photofission yield multiplicity, spectra, and particles emission time reference. No currently available code satisfied our requirements, so we customized MCNPX 2.4.0 [4] and MCNP-PoliMi [5] to simulate the required physics. The calculation methodology is split into three steps as follows:

1) the modified MCNPX code is used to transport the interrogating photons throughout the problem geometry. It determines the type and position of photonuclear interactions that occur within the system that is being modeled. This information is saved to an output file.

2) MCNP-PoliMi [5] reads the file created in the previous step and transports the secondary photons and neutrons regarding them as new sources. In our application, the interactions occurring in each of two detectors are stored, together with the arrival time at the detector, in an external file.

3) a post-processor reads the file generated in the previous step, calculates and plots the time-correlation of the signals from the two detectors.

DATA SOURCES FOR PHOTONUCLEAR REACTIONS

The current customized MCNPX version simulates only the beam-on data acquisition. The photo-fission yield is dealt with as follows:

- The average number of neutrons is read out of the Bofod01u library. No distinction is made between prompt and delayed neutrons average. This value is used to calculate the neutrons multiplicity by the Terrel's Formula [6].
- No information is provided about the photon yield in the Bofod01u library. The average number of photons is calculated by T. Valentine's formula [7]. The photon multiplicity is sampled from a negative binomial distribution as indicated in T. Valentine's paper [7], the same way as the photon multiplicity is obtained for neutron-induced fission.
- No delayed neutron is generated at this time. This is work-in-progress for the next release.
- Delayed photons are optionally generated at the user's discretion. If this option is chosen, a delay is sampled from a distribution adjusted to the delayed gamma rays emitted by ^{252}Cf spontaneous fission.

The photon energy is sampled independently from the emission delay. The same rules apply to the delayed photons from neutron-induced fission.

Gamma rays can be emitted together with neutrons during the evaporation stage of the reaction and even during the pre-equilibrium stage, before evaporation, and before fission [8]. Currently, the implemented description of prompt gamma and neutron multiplicities and spectra, for any of the investigated actinides, makes use of the emission yield from neutron-induced ^{235}U fission. This choice is justified by the similarity of photon emission spectra at energy values around 10^2 keV or higher. In addition, photons in this energy range contribute the greatest amount to the total detected signal as they are less likely to be captured.

Replacement of the photofission gamma yield with the neutron-induced fission yield is justified by the fact that once the scission of the target nucleus has taken place, the deexcitation channels of the daughter fragments do not carry any memory about the fission type. Modeling the unknown photofission yield through the known neutron-induced fission yield has been proved a good enough

approximation by M. B. Chadwik, W. B. Wilson at Los Alamos National Laboratory where they deal with photonuclear reactions and transmutation of long-lived nuclear waste in high photon fluxes. The same hypothesis turned out to be reasonably sustainable in a study comparing the yield from photofission, and thermal neutron-induced fission of plutonium isotopes [9]. The same study shows also that the spectra of plutonium and uranium isotopes are similar. For this reason we plan to obtain the missing data for ^{238}U by interpolating the prompt photon multiplicities and spectra for ^{235}U and $^{239}P_u$ measured by Verbinsky, Weber, and Sund [10]

For what concerns the photonuclear interactions (γ,n) and (γ,2n), an energy balance is applied to determine the total energy available for the deexcitation gamma rays that accompany the neutron emission. The photo-nuclear neutron and gamma ray yield is dealt with as follows:

- If the photon emission data is available for the material where the reaction has taken place then this information is used. If only the Q-value is available, then some photons are generated with isotropic direction in such a way as to fulfill the energy balance. The number of photons that are emitted is then input as an adjustable parameter by the user.
- Neutrons multiplicities and spectra for actinides are read out of the Bofod01u library

RESULTS

In order to test the efficiency and accuracy of our proposed material interrogation system, we applied it to the real problem of detecting nuclear material concealed in packages. The scenario consists of a luggage belt-conveyor lying on a concrete stand. In the concrete a cavity, whose radius is 10 cm, has been designed to accommodate a photon source placed at 50 cm below the belt plane. The photon beam is collimated to a conical shape in such a way as to irradiate one piece of luggage at a time or none. The beam cone size is ~76.2 cm in the x-y plane. On each side of the belt, in correspondence of the beam cone, there are two scintillation detectors collecting the signal from the package. Each detector size is 100 x 8 x 100 cm. The two detectors, which are sensitive to both neutrons and photons, are 228 cm apart. A schematic diagram of the measurement system configuration is shown in Fig. 1. The goal is to detect the presence of nuclear material, if any, inside the luggage.

In order to account for a large number of possible scenarios, we are examining and simulating fourty-six different cases stemming from different combinations of sizes of the problem geometry components and different materials in the target. In this paper we only present the results for a subset of our survey.

The package size that is considered here is 43 x 40 x 18 cm. It is made up of Celotex and can contain a 5 Kg HEU or 10 Kg HEU sphere. To test the discriminating capability of our system against high-Z material versus nuclear material, we kept the respective volumes of the 5 Kg U and 10 Kg U spheres constant and replaced the U with Fe or with Pb. Finally, we have picked the largest Pb sphere that can be fit in the small package which turns out to be 33.6 Kg Pb.

The results for the belt-conveyor project, presented in this paper, were obtained by averaging over 20 simulations ran with the modified MCNPX code, each transporting $2x10^9$ particles for each system configuration investigated.

ANALYSIS

Fig. 2 to 4 show the separate contribution of the photonuclear and photoatomic reactions from our simulations. We infer that the amplitude of the photoatomic signal, which is significant only in the range (-10,10) ns, is roughly the same for all material samples, whereas the intensity of the

correlation signal at higher delays, which is due to neutrons production, scales with the amount of the mass hit by the source photons. The photoatomic signal for various materials exhibits a sharp cusp around time t=0 due to time-coupled γ-γ detection, and two symmetrical side peaks due to Compton scattering, in-between the detectors. The two symmetric wings, in the photonuclear portion of the signature, are due to the contribution of neutrons.
Fig.5 shows the total correlation function. This signature cannot discriminate among the 5 Kg U and the 33.6 Kg Pb sphere. Fig.6 shows the γ-γ correlation function gives a significant contribution from non-nuclear materials only in the range (-20,20) ns. At higher delays there survive only the signal from the materials where the chain reactions give rise to radiation emission at longer times. Both figures show the superimposed signal from the photonuclear and the photoatomic reactions, as it is read out of the measurement device. Drawing on our analysis we calculated the Full-Width-At-5000_th-Max of the γ-γ correlation function. The results, shown in Fig.7, confirm that this feature stands out as a possible key for material detection.

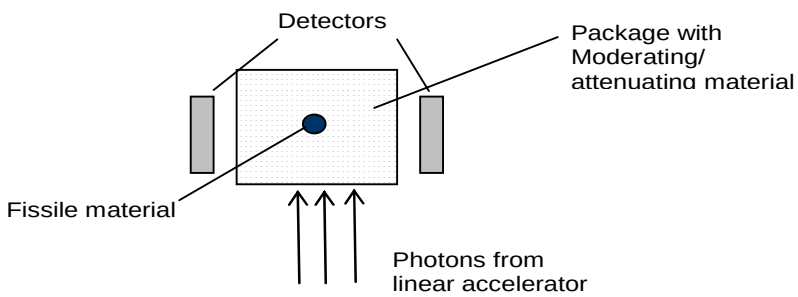

Fig. 1 Materials investigation device. Top view.

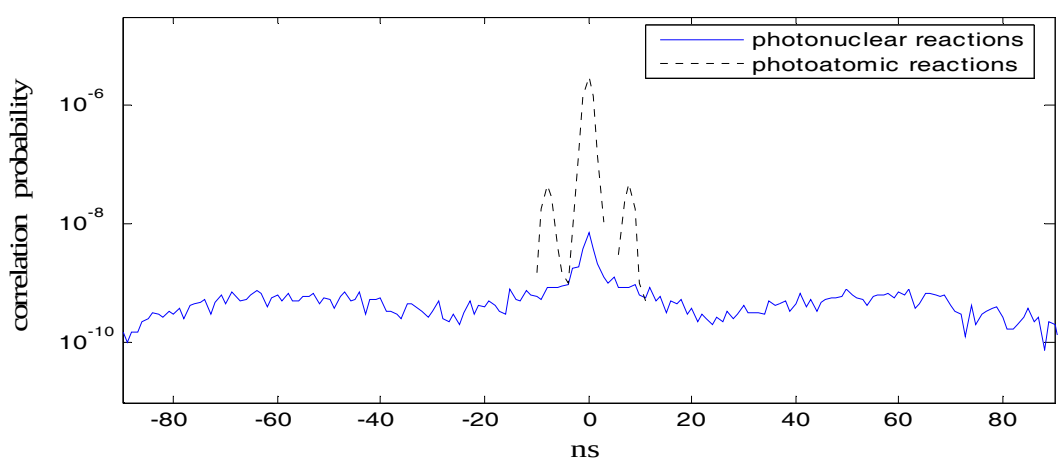

Fig. 2 Contribution of the photo-nuclear and photo-atomic reactions for the 5Kg U sphere and small package case.

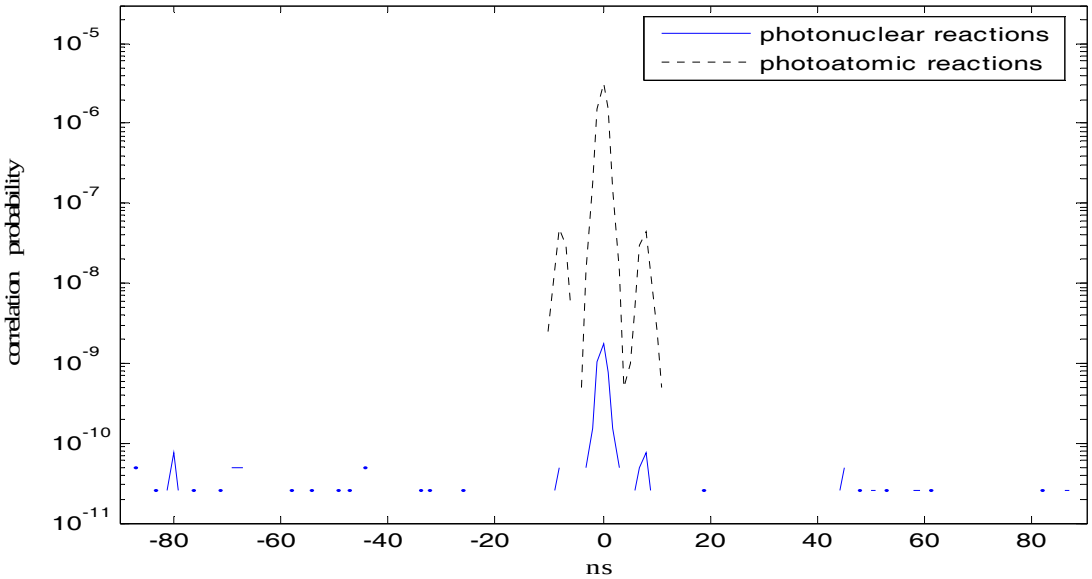

Fig. 3 Contribution of the photo-nuclear and photo-atomic reactions when no sphere is inside the small Celotex package.

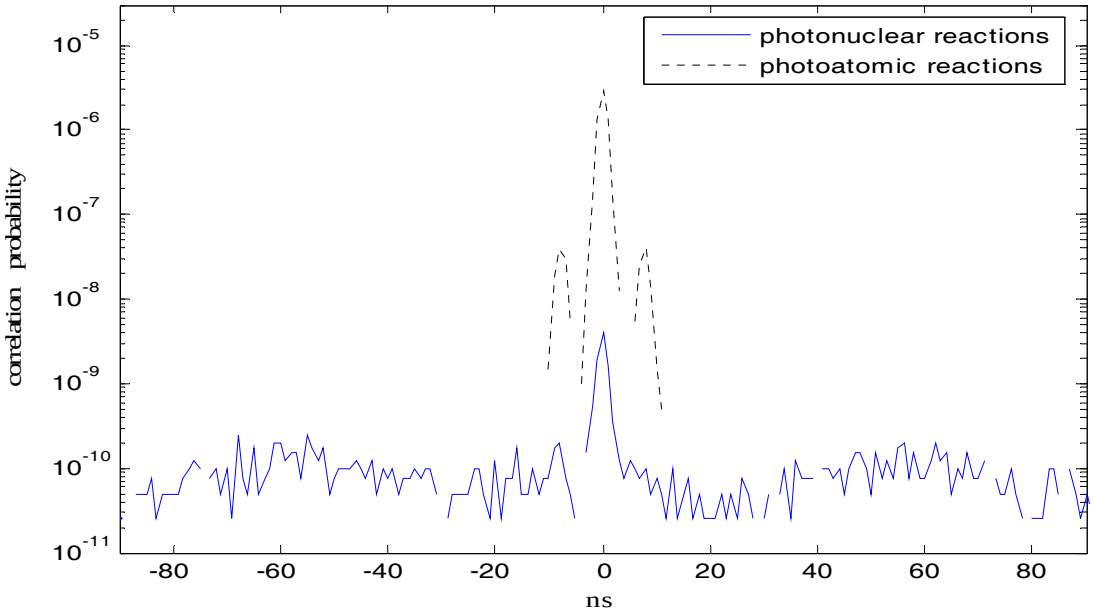

Fig. 4 Contribution of the photo-nuclear and photo-atomic reactions for the Pb sphere and small package case.

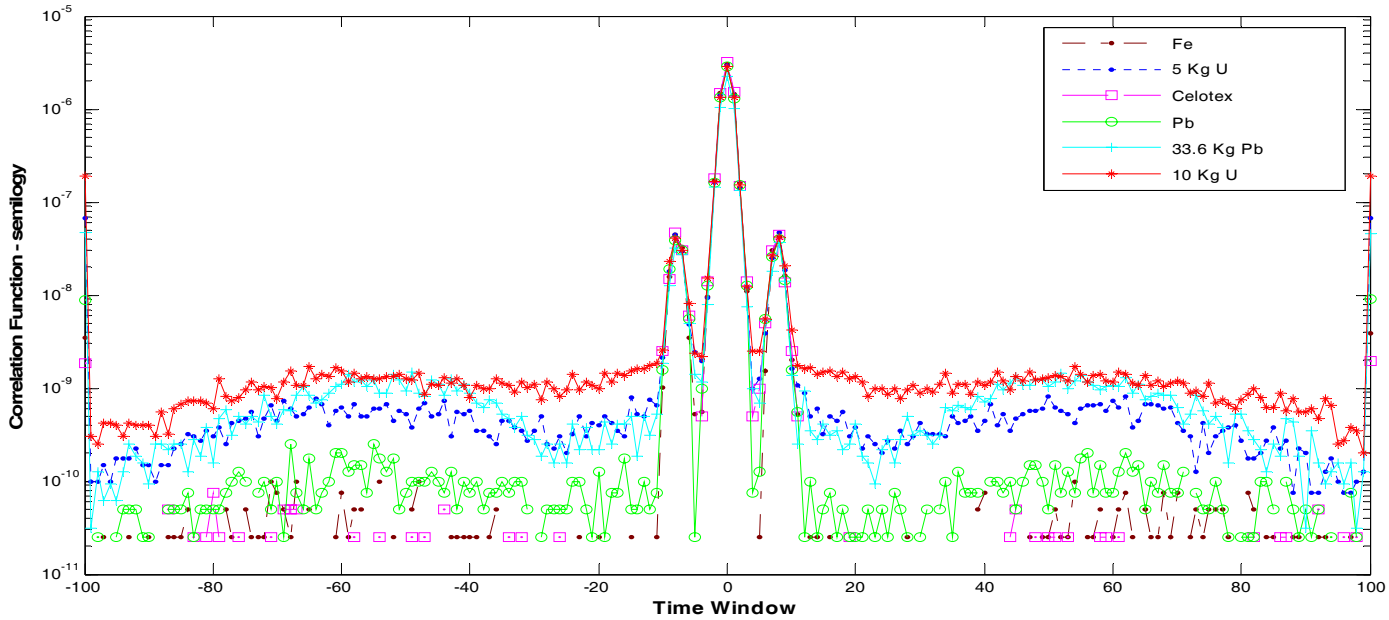

Fig. 5 The total correlation function is contaminated with the signal from the biggest Pb sphere that can be fit in the small package.

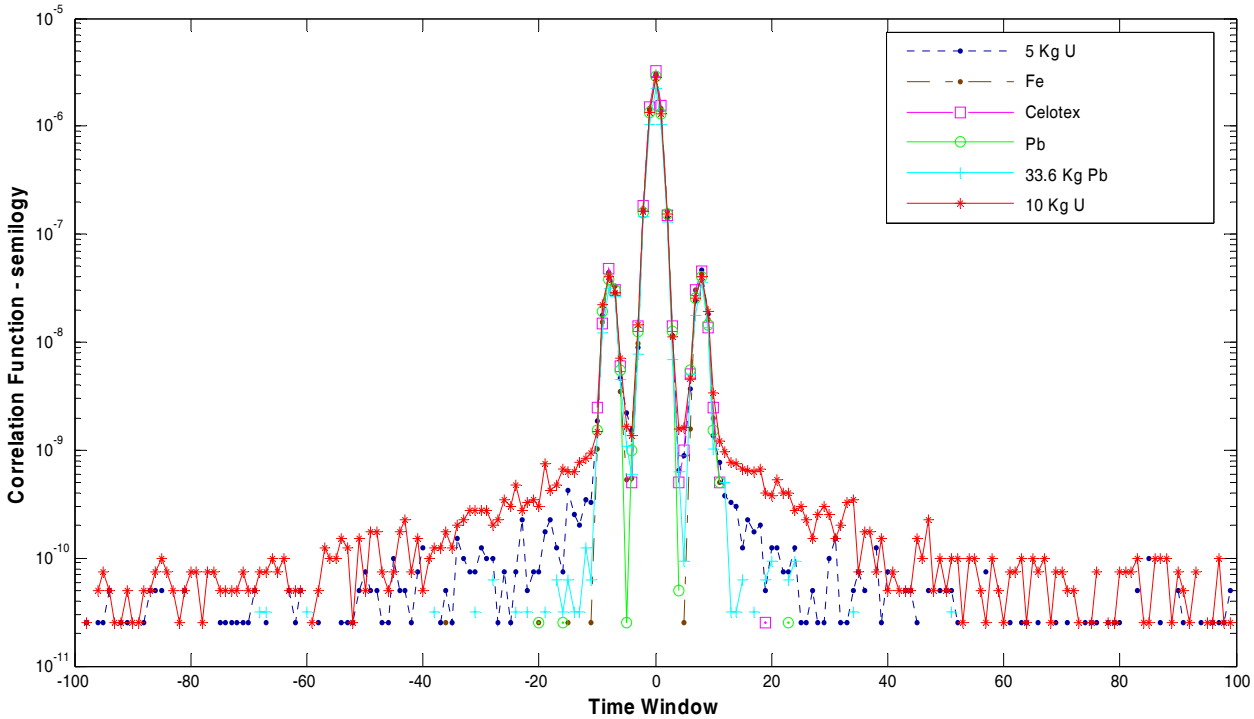

Fig. 6 The γ-γ correlation function benefits from the delayed neutrons from photofission contributing to the wings of the function, therefore discriminating between fissile and non-fissile materials.

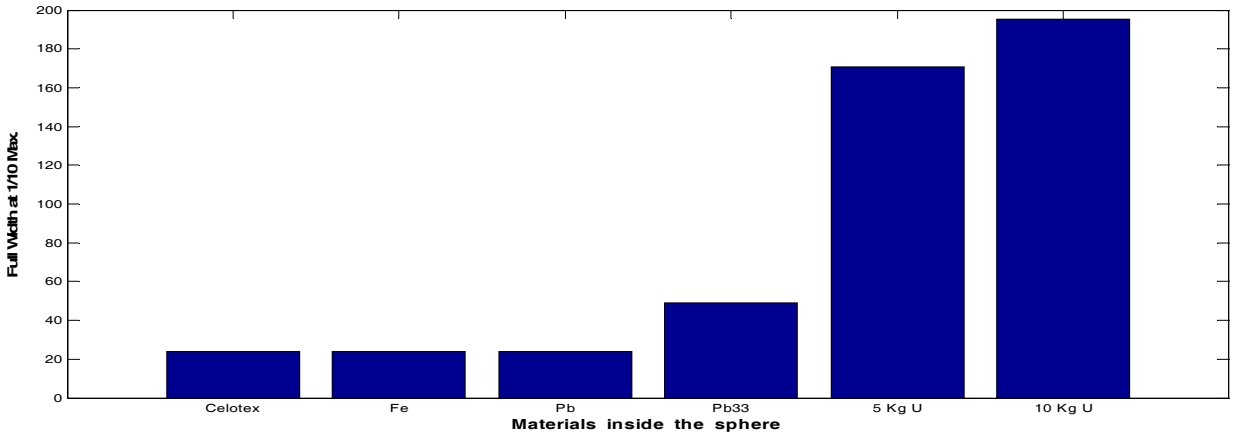

Fig. 7 Full width at 1/50000 maximim of the γ-γ cross-correlation function for different investigated materials.

CONCLUSIONS

We have shown that fissile material identification is possible by analyzing the photonuclear prompt radiation. We have validated our proposed investigation system through the analysis of a real scenario. Our preliminary results show that is possible to detect and extract the encoded material signature from the photonuclear prompt radiation . The proposed further enhancement to the PoliMi code to include the emission of delayed radiation from photonuclear events will complete the treatment and allow the code to be used to simulate measurements with the beam off. It is possible that we will find that the measurements with beam off are more sensitive to the presence of fissile material, or that they work better in certain scenarios.

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