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Nuclear Material Interrogation via High-energy β-delayed Gamma Rays from Photo-fission

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Abstract. An increased urgency has been associated with the development of new and improved means for the non-destructive characterization of nuclear waste or weapon grade materials that might be hidden in large storage blocks or cargo containers. Passive detection methods based on measurements of neutrons and/or photons are either inapplicable or impractical in many such cases. Active interrogation with neutrons or photons in a variety of forms currently depends upon the observation of β-delayed neutrons following induced fission to provide a unique signature for fissile material. However, the shielding provided by a thick waste container envelope, typically made of concrete of high density as well as with high water content, could be so large that this method will fail or will have very low detection sensitivity. Equally for identical reasons only high-energy neutrons or high-energy gammas have high enough range to activate the enclosed actinides.

In this paper we provide quantitative estimates on the detection limits of fissile material in the case of typical nuclear waste containers surrounded by massive concrete layers with variable contents of hydrogen. Signals of prompt photo-fission neutrons, β-delayed gammas and β-delayed neutrons are examined for direct comparison. Advantages and disadvantages detecting all these observables are discussed. We show that β-delayed gamma rays can offer an increase in sensitivity for the detection of fissile materials by as much as 2-3 orders of magnitude (depending on the hydrogen content in the waste envelope) when compared to the detection of β-delayed neutrons. Due to the lack of experimental data on decay photons from photo-fission fragments an experimental program is proposed to confirm these investigations.

Introduction

The material interrogation via photo-fission consists of irradiating radioactive matter with high-energy Bremsstrahlung photons in order to produce (γ,fiss) reactions in the enclosed actinides. Measurements are based on detecting either delayed neutrons (DN) [1], or prompt neutrons (PN) [2], or delayed photons (DN) [3] in order to establish the quantity of fissile material present as shown in Fig. 1. The major goal of this work is to provide quantitative comparison among the above three observables in terms of the expected signal of fissile material in the case of typical nuclear waste containers surrounded by massive concrete layers with variable contents of hydrogen.

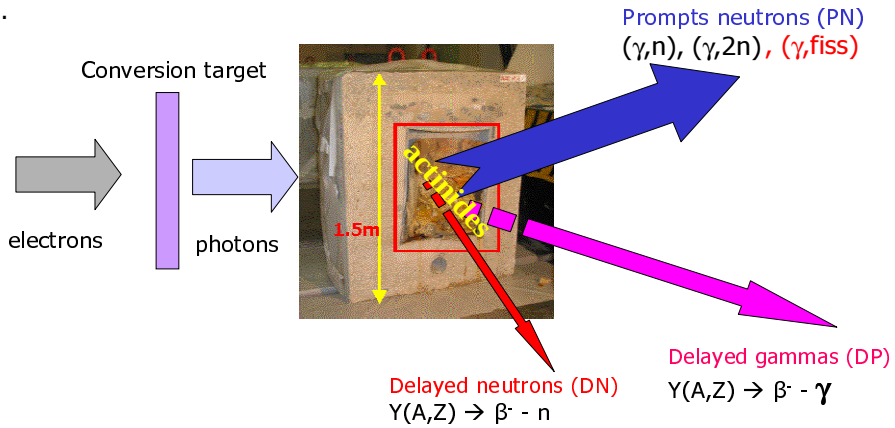

Figure 1. A schematic view of the photo-fission interrogation: emission of either PNs, or DNs , or decay DPs will take place following the photo-fission event.

Waste characterization with DNs, PNs, and DPs

The three measuring methods have their advantages and drawbacks. Below we try to summarize some of them:

- Delayed Neutrons

An advantage of this method is that the DN signal is proportional to the actinide mass in the waste. In principle, it is unaffected by photo-neutron interference and the gamma-flash [1]. However, the signal itself is rather weak, i.e. there are only ~ 1 % of DNs emitted per fission. DN averaged energy is ~ 0.6 MeV, i.e. attenuation level is high and emitted DNs will have strong dependence on hydrogen content in the waste matrix. The signal interference due to the reaction $^{18}O(\gamma,p)^{17}N$ (the threshold ~15 MeV) and emission of DNs ($T_{1/2}$ = 4.17 s) in some cases may become a limiting factor. Finally, the signal background due to thermal neutrons also might be an issue.

- Prompt Neutrons

This method is statistically satisfactory [2]. A number of prompt neutrons emitted in fission is typically by two orders of magnitude greater than the number of delayed neutrons. In addition, prompt neutron energy (~2.0 MeV) is somewhat higher than delayed neutron energy (~0.6 MeV), what favors higher permeability through the waste matrix to the detection system. On the other hand, (γ,xn) reactions on the converter target and also on different materials other than ACTINIDEs produce signal interference that increases proportionally to the energy of the incident photons. Further more, the interrogation photons may "blind" the measurement system for a period up to ~1 ms (the "gamma flash"), what limits the counting of the prompt signal. Finally, as in the case of DNs, PN signal will be very much dependent on the waste matrix and its hydrogen content in particular.

- Delayed Photons

The main advantage of this method is that it will not depend on the hydrogen content in the waste matrix. In addition, detection of delayed photons would increase the detection sensitivity due to the relatively high intensity of β-delayed γ rays (2-photons/fission. Large area and low resolution plastic scintillators are potential candidates for such a detection system [3]. Since, like in the case of DNs, one is detecting a delayed signal, this method is unaffected by prompt photo-neutron interference and the gamma-flash. On the other hand, decay photons from different activation products (other than actinides) may produce interference that would increase proportionally to the intensity of the incident Bremsstrahlung photon flux.

Calculation procedure

In order to compare the above three methods we employ the multi-particle transport code MCNPX [4] combined with the material evolution program CINDER'90 [5]. The DN and DP data (DN emission probabilities, DP intensities and decay constants of fission fragments) were based on the ENDF/B-VI evaluations [6] from neutron induced fission. Our choice to use the decay data from neutron induced fission is mainly due to the absence of equivalent data for photo-fission. However, by following the systematic approach in terms of the same composite nucleus before fission and comparable excitation energy for photon and neutron induced reactions, we expect our results be justified and valid at least within a factor of two.

We model a typical concrete block, characterized by its dimensions of 120 x 120 x 150 cm^3 and density of ρ = 3 g/cm^3. A volumetric waste source is placed in the center and defined as a ~2 liters cylinder. Within this volume either DNs, or PNs, or DPs start their transport history and outgoing particles are tracked using Monte Carlo techniques within MCNPX. Realistic energy spectra of all particles are taken into account in detail. The main variable in our modeling is the hydrogen level in the concrete matrix. This quantity is allowed to vary from 0 to 2.5 %.

Results

As it was mentioned above, the main disadvantage of DPs is that some of the activation products may produce undesirable background. However, by conditioning detected DPs in terms of their energy and measuring time one can overcome this drawback. From Fig. 2 (on the left) one can see that photon decay curves from fission fragments are defined by their characteristic averaged half-lives (similarity to DN time spectra). For example, for ^{235}U this $<T_{1/2}>$ is of the order of 19 s within the cooling time interval from ~20 s to ~60 s. Decay photons from activation products inside concrete are either very short lived (both high and low energy photons) or very long lived (high energy photons only) as presented in Fig. 2 (on the right). Therefore, by selecting only high energy photons from fission fragments with specific half-lives, high energy photons from activation products would contribute only to the constant background (within a certain period of data taking).

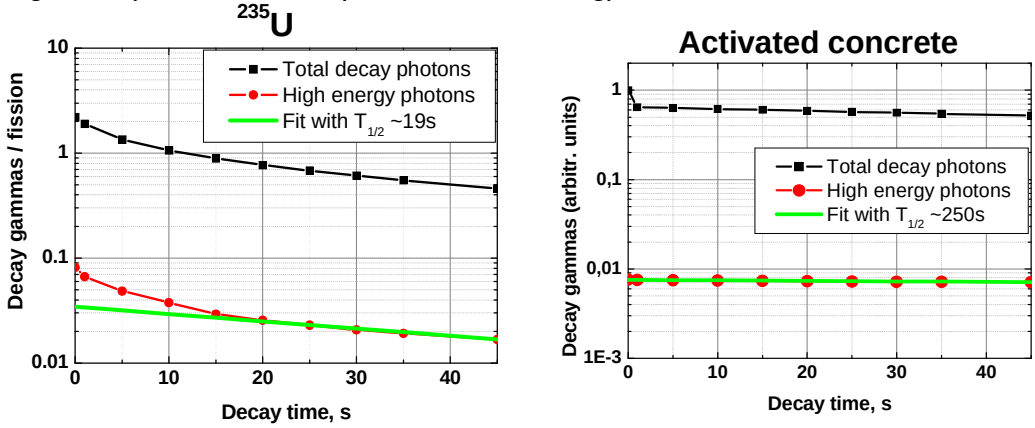

Figure 2. Intensity of total and high energy (E_γ > 2.5 MeV) DPs as a function of cooling time: on the left – from fission fragments of ^{235}U; on the right – from activation products of concrete and its impurities (>200 typical impurity isotopes were taken into account).

How these DPs compare with DNs or PNs escaped from the concrete matrix and detected afterwards? In Fig. 3 we present a number of escaped particles as a function of hydrogen level in the concrete. Note that in the case of DPs only high energy photons are taken into account and the escape signal is normalized per one fission event in ^{235}U. From the very preliminary analysis of these calculations we can conclude that one of the ways to increase the actinide detection sensitivity would be to measure the relatively high intensity of β-delayed γ rays with energies higher than ~2.5 MeV that are emitted from short lived fission products. These β-delayed γ rays have yields that are nearly an order of magnitude higher than the corresponding β-delayed neutrons. In addition, these high energy gammas are likely to be transmitted through the hydrogenous material at least one order of magnitude better than β-delayed neutrons. As a result, the detection sensitivity of nuclear material could be increased by 2 or 3 orders of magnitude (compare green and blue bars). Another important result is that, as expected, the DPs escape is not dependent at all on hydrogen level of the waste matrix. Finally, we note that at hydrogen content as high as 2.5 %, DP signal becomes equivalent even to the PN signal.

We continued our analysis with delayed photons even further. In Ref. [7] the authors argued that isotopic identification of actinides is experimentally feasible by measuring delayed neutron time spectra of composite material and by knowing with a good precision the base lines of separate isotopes (e.g., identification of ^{235}U and ^{239}Pu in the Pu-U mixture). In brief, this result was possible because of difference in: 1) absolute yields of DNs, namely DN(^{235}U)/DN(^{239}Pu)~2.6; and 2) averaged half-lives of DN decay curves, namely $<T_{1/2}>$(^{235}U)~8.8 s and $<T_{1/2}>$(^{239}Pu)~10.2 s. We

performed similar analysis in the case of ^{235}U and ^{238}U mixture but using delayed high energy photons from fission as observable. It seems that also with this method isotopic identification of actinides would be feasible, again because of difference in 1) absolute yields of DPs, namely DP(^{235}U)/DN(^{238}U)~2.0; and 2) averaged half lives of DP decay curves, namely $<T_{1/2}>(^{235}U)$~19 s and $<T_{1/2}>(^{238}U)$~15 s within the cooling-detection time interval from ~20 s to ~60 s. Due to the lack of the data on delayed photon yields and their time spectra from photo-fission an experimental program should be carried out to confirm our results.

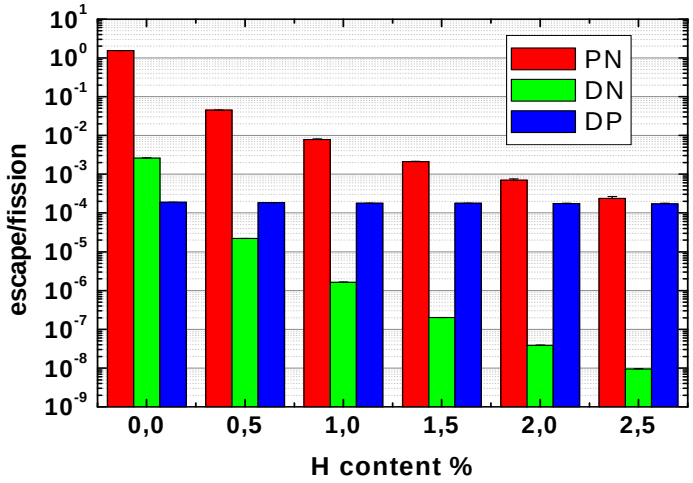

Figure 3. Expected escape particles over 4π (PNs, DNs and high energy DPs) as a function of hydrogen level in the concrete block. The normalization is per one fission event. In all cases the same detection efficiency of 100 % is assumed.

Summary

Actinide waste detection by photon interrogation and expected delayed photon counting was examined and quantitatively compared with counting of delayed neutrons and prompt neutrons following the photo-fission event. We conclude that with DPs the detection sensitivity of nuclear material could be increased by 2-3 orders of magnitude if compared to DNs. In addition, contrary to neutrons, the DP escape is not dependent at all on hydrogen level or neutron absorbers in the waste matrix. Finally, our preliminary calculations show that DPs also could be used for isotopic identification of actinides in different sample mixtures in the matrix-free environment. In order to confirm the above findings some test measurements will be performed at SAPHIR electron accelerator of CEA Saclay already in 2005.

References

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