# Delayed gammas detection technique for nuclear waste characterization

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***Abstract:*** Photons can be used to detect small amounts of nuclear material inside massive nuclear waste storage barrels or large cargo containers. The method consists of the irradiation of the waste barrels by photons in order to create photofission reactions on actinides. By detecting the decay gammas associated with fission, it is possible to determine the presence of the actinides and their mass contained in the waste barrel.
The experimental data on delayed gamma emission following photofission is very scarce and insufficient for the above purposes. Therefore, within the INPHO project (**In**terrogation by **Pho**tons) the measurements of delayed gamma spectra have been performed at CEA. The gamma detection device was composed of the BGO detector. $^{238}$U and $^{232}$Th targets were irradiated separately to determine fundamental parameters (decay gamma energy and time spectra). Preliminary data analysis of this experiment will be presented, including model calculations based on systematic from neutron induced fission.

## Introduction

Recently, non-destructive characterization of waste containers and detection of nuclear materials, both based on photofission process, has attracted particular attention. This technique employs intense beam of Bremsstrahlung photons with energy above 6 MeV in order to induce photofission on actinides. Simplicity and attractive costs of low energy electron accelerators, associated with prompt neutron (PN), delayed neutron (DN) and delayed-decay photon (DP) detection devices, make possible to detect small amounts of fissile material present in massive nuclear waste storage barrels. In principle, the same technique could be applied in nuclear material control on the borders [1].
The measurement of DNs and DPs emitted by fission products brings specific information on localization and quantification of the nuclear material. A simultaneous measurement of both of these delayed signals can overcome some important limitations due to matrix effects like heavy shielding and/or the presence of light elements as hydrogen [1, 2].
To determine the total number of DP events, one needs a detector which allows counting of high energy photons in an environment, where photons and neutrons are both present. We have chosen the BGO crystal scintillator the properties of which (efficiency, neutron transparency, etc.) are well suited for this purpose.
The investigated gamma energy range was above 2-3 MeV to maximize the counting of delayed gammas from photofission compared to the active background. Similarly as in the delayed neutron experiments [3], different irradiation-decay cycles were employed to optimize the extraction of “unique” decay parameters of delayed gammas.
This paper presents our first experimental results and preliminary data analysis on photofission induced DP energy and time spectra. Determination of the fundamental parameters at different gamma energy cuts has been performed for $^{238}$U, while for $^{232}$Th the analysis is still in progress. Comparison with model calculations based on systematic from neutron induced fission will also be discussed.

## Description of the experiment

The experiment was performed using the ELSA electron accelerator of CEA/DAM Ile-de-France at Bruyères-le-Châtel [4]. A 15 MeV-electron beam with an energy resolution lower than 100 keV and an average intensity of ~1 μA was employed. All irradiations were done with a frequency of 1 Hz and with the 70 μs pulse duration. The electrons were converted into Bremsstrahlung photons using a 2 mm-thick tantalum target. The distance from the target to

the actinide samples was ~150 cm along the beam axis. The $^{238}$U and $^{232}$Th samples were ~400 g and ~300 g metallic cylinders respectively. A massive lead collimator narrowed the photon beam to reduce the background.
Each measurement consisted of a repetitive cycles of irradiation and counting once the beam is switched off. The irradiation and counting time were adjusted in order to maximize the contribution of different groups with their characteristic decay periods, namely 70μs (single pulse) – 30 s (142 cycles), 10 s – 50 s (60 cycles), 60 s – 300 s (21 cycles), and 300 s – 300 s (15 cycles).
Each gamma event was recorded with two parameters, its energy and the elapsed time since the end of the irradiation. For each irradiation time, a run without any sample was also performed in order to estimate the active gamma background.
A 5"– diameter × 3"– height BGO scintillator (127S76/5 BGO + preamplifier AS16, SAINT GOBAIN) was used to detect high-energy gammas. The BGO was placed at the angle of 90° with respect to the beam axis and embedded in a 30 × 30 × 40 cm polyethylene block as shown in Fig. 1. The distance from the uranium sample to the detector was ~100 cm.

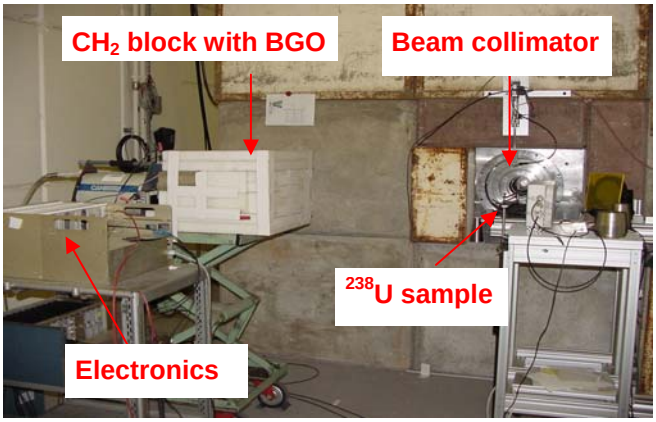


***Figure 1:*** Experimental set up for DP measurements at the ELSA electron accelerator

Both the time and energy spectra were recorded event by event. In order to determine with a good accuracy the "start" of the gamma decay, the last 2 s of the beam were recorded for each cycle. The dead time of the whole system was estimated to be about 20 %.

## Preliminary results of data analysis

Fig. 2 presents both energy and time spectra of DPs obtained in this experiment. By choosing the DP energies higher than 2 MeV we analyzed the raw DP time spectra as a sum of exponentials in the case of $^{238}$U, namely

$$F(t) = \sum_i A_i \exp(-\lambda_i t) \times (1 - \exp(-\lambda_i T_{irr})) + C \,. \qquad \text{(Eq. 1)}$$

As in the approach well known in the case of delayed neutrons, we wanted to extract "unique parameters" $\{A_i; \lambda_i\}$, which could be used to predict the DP activity for variable irradiation ($T_{irr}$) and decay (t) times. Note that the above equation is rather good approximation once $T_{irr}$ is much shorter than t or both of them are "infinitely" long. The constant *C* was added in order to take into account the active background, which was assessed at 10 % of the raw signal. The same approach can be applied for $^{232}$Th but by choosing the DP energies higher than 3 MeV due to the "naturally" present the 2.614 MeV gamma line of $^{208}$Tl (see Fig. 2).
The following fitting procedure was employed: starting from the 300 s / 300 s (irradiation / counting) cycles, the specific values for group 1 were determined. Then in the 60 s / 300 s irradiation the first group parameters were fixed to determine the other group parameters, and so on until the single pulse irradiation. The measured average decay half-lives for $^{238}$U are shown in Table 1.

***Table 1:*** Experimental results on DP averaged half-lives $T^{i}_{1/2}$ for $^{238}$U.

| DP group | $i$ = 1 | $i$ = 2 | $i$ = 3 |
|---|---|---|---|
| $T^{i}_{1/2}$ **(s)** | 158 ± 8 | 26 ± 1 | 1.89 ± 0.13 |

At the 2nd stage, these values were fixed and tested by fitting all the time spectra once again. The results are shown in Figure 3 for the representation of the "unique global fit" in the case of 4 different irradiation periods. One can clearly see that all spectra are well reproduced. Finally, using these "unique" time constants, relative group contributions $A_i$ will be extracted for each irradiation time. This work is still in progress.

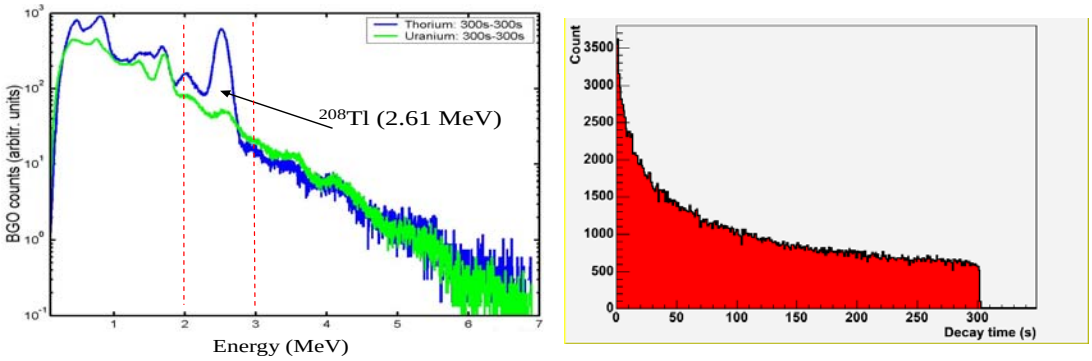


***Figure 2:*** On the left – DP energy spectra from photofission of $^{238}$U (in green) and $^{232}$Th (in blue); on the right – DP time spectra for $^{238}$U ($E\gamma$ > 2MeV and 300s-300s irradiation-decay).

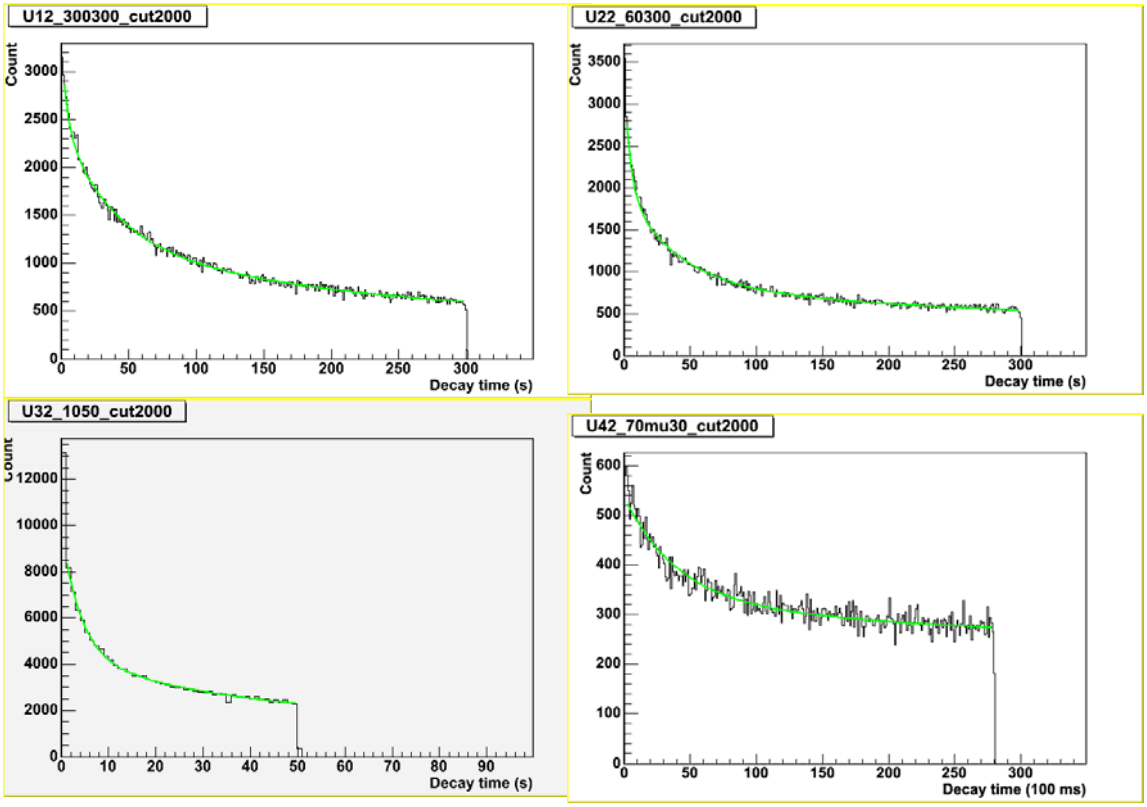


***Figure 3:*** $^{238}$U relevant unique fit with time constants fixed according to Table 1 and a 2 MeV gamma energy cut. See the legend for irradiation-decay periods.

**Total number of DP per photofission**

Theoretically, an infinite time irradiation allows to determine the total number of delayed gammas emitted per fission. Let us assume that the 300 s / 300 s cycles correspond to this particular case, i.e. the major DP contributors have reached their production equilibrium. The total number of high energy DPs per fission at a given time can be obtained by:

$$\nu_{DP} = \frac{N_\gamma(t)}{N_{fiss}} = \frac{N_{BGO}(t)}{N_{fiss}\varepsilon_{BGO}}, \qquad \text{(Eq. 2)}$$

where $N_{BGO}$ is the number of DP detected per second at time $t$ (see Fig. 3), $N_{fiss}$ - the number of fissions per second, $\varepsilon_{BGO}$ - the total BGO efficiency, which includes the intrinsic detector efficiency, solid angle and attenuation both in uranium target and in shielding.
The number of fission events and the BGO efficiency were evaluated using the MCNPX code [5] calculations taking into account in detail the experimental set-up geometry and the information related to the primary electron beam intensity. The calculated value for the number of fissions was $N_{fiss} = 1.75 \times 10^6$ fissions.$s^{-1}$.$\mu A^{-1}$ for $^{238}U$. By taking into account the theoretical energy spectra of DP from neutron induced fission [6] and using MCNPX we estimated that $\varepsilon_{BGO} = 2.46 \times 10^{-4}$ events per incident DP with $E_\gamma > 2$MeV.
In order to check these simulations the absolute efficiency calibration measurements were done with low energy gamma sources such as $^{137}Cs$ ($E\gamma$ = 661 keV) and $^{60}Co$ ($E\gamma$ = 1333 keV/1173 keV) and identical experimental geometry. The experimental and simulated efficiencies agreed within 13 %.
The total DP emission of $^{238}U$ was also estimated using the CINDER code [6], based on neutron induced fission process. The experimental irradiation conditions were taken into account in these simulations. If one assumes that both n-fission and $\gamma$-fission of $^{238}U$ result in similar fission products, the predicted DP value is in a good agreement with our experimental result as shown in Table 2. More detailed analysis on these comparisons is in progress.

***Table 2:*** Results on $\upsilon_{DP}$ for the $^{238}U$ target with with $E_\gamma > 2$ MeV.

| Decay time t = 1s | Exp. $^{238}U(\gamma,fiss)$ | Eval. $^{237}U(n,fiss)$ | Eval. $^{238}U(n,fiss)$ |
|---|---|---|---|
| $\upsilon_{DP}$ ($\gamma$/fission) | 0.462 ± 0.070 | 0.555 | 0.607 |

## Conclusions

The DP emission following photofission was measured in the case of $^{238}U$ and $^{232}Th$ targets. 15 MeV end-point Bremssthralung photons were employed to induce photofission and BGO was used to detect high energy gammas.
The DP decay curves were studied with different energy cuts from 2 MeV to 3.5 MeV. The decay time parameters for $^{238}U$ were determined for variable irradiation times and photon energies higher than 2 MeV, while data analysis for $^{232}Th$ is still in progress. It seems that all irradiations can be reproduced with only 3 averaged half lives, namely ~158 s, ~26 s and ~2 s. The obtained time constants are compatible with known major photofission products (high energy gamma emitters) determined by gamma off line spectroscopy at CEA [7]. The experimental total high energy (above 2 MeV) DP emission per photofission for $^{238}U$ is ~0.46, what is very close to the value of ~0.61 known from neutron induced fission on the same nucleus. Further investigations have to be undertaken including the extraction of relative group contributions in particular.
The future experiments will be done using $^{235}U$, $^{237}Np$ and $^{239}Pu$ targets.

## Acknowledgements

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