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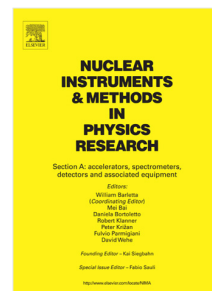
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Measurement of cumulative photofission yields of ^{235}U and ^{238}U with a 16 MeV Bremsstrahlung photon beam

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Keywords

Photofission, uranium, delayed gamma rays, Bremsstrahlung, fission yield, photon activation analysis, MCNP

Abstract

In the frame of a long-term research program on the characterization of large radioactive waste packages by photofission, the Nuclear Measurement Laboratory of CEA IRESNE, France, has measured cumulative photofission yields of ^{235}U and ^{238}U short-lived and long-lived fission products by using a Bremsstrahlung photon beam produced by a 16 MeV electron linear accelerator (LINAC). To this aim, a characterization of the Bremsstrahlung photon beam has been carried out by photon activation analysis with different samples of gold, nickel, uranium and zirconium. The residual neutron flux exiting the LINAC head (lead collimator, borated polyethylene and cadmium shield) has also been characterized by neutron activation analysis with indium samples to estimate the contribution of photoneutron fissions in the uranium samples used to assess the

26 photofission yields. Finally, 49 fission product yields are reported for ^{238}U and 26 for ^{235}U ,
27 with half-lives ranging from 64 s to more than 3 days, some of them not recorded so far in
28 the literature. Some photofission products cumulative yields show significant differences
29 between ^{235}U and ^{238}U , which confirms the possibility of an isotopic discrimination method
30 based on delayed gamma-ray ratios analysis for radioactive waste characterization.

31 1. Introduction

32 The management of radioactive waste (transportation, interim storages and final
33 repositories) needs an accurate characterization of their nuclear material content. Among
34 many non-destructive methods that have been studied to address this characterization,
35 active interrogation methods are necessary for large and dense packages, such as concrete
36 drums [1]-[4]. Specifically, Active Photon Interrogation (API), based on the detection of
37 delayed gamma radiation emitted by fission products induced by high-energy photons, has
38 the potential to assess the nuclear material mass present in a package, and possibly to
39 distinguish fertile and fissile isotopes (e.g. ^{238}U and ^{235}U , respectively) using a
40 discrimination signal based on their delayed gamma rays. The detection of delayed gamma
41 rays with a high-purity germanium (HPGe) detector has the potential to measure the
42 production yields of fission products with an accuracy lower than 5 %, enabling the
43 differentiation from one isotope to the other [1].

44 However, nuclear data related to photofission yields are scarce and the reported fission
45 product cumulative yields show significant discrepancies, even for recent studies [5]-[11].
46 Due to the Giant Dipolar Resonance (GDR), the photofission cross-section of both odd and
47 even mass number actinides (e.g. ^{238}U and ^{235}U) are of the same order of magnitude,
48 contrary to thermal neutron fission for which they differ by several decades. Uranium 235
49 is a “fissile” actinide (in the sense of thermal neutron fission) of first interest in a
50 radioactive waste, but it is most of the time accompanied with a 10 to 100 times larger mass
51 of uranium 238. Even with well-known photofission cross-sections, relative fission product
52 yields of these two uranium isotopes have to be determined with a good accuracy in view
53 of their discrimination.

54 Several research groups have shown the potential of measuring delayed gamma rays
55 following fission for the differentiation of actinides, such as Hollas *et al.* [12] for
56 photofission, Beddingfield [13] for thermal neutron fission, with the use of delayed
57 gamma-ray ratios for actinide identification. Further experimental work was conducted by
58 Gmar *et al.* [14], pointing out variations of the delayed gamma-ray emissions for uranium
59 samples of different enrichments. Carrel *et al.* [1] have investigated delayed gamma
60 emission following photofission in mixed samples of ^{235}U and ^{238}U , and delayed gamma-
61 ray ratios for actinide differentiation with samples positioned in the center of an 870 L
62 mock-up package. Simon *et al.* have also reported the possibility to distinguish uranium
63 235 and 238 in 870 L radioactive waste packages using Monte-Carlo simulations [4], by
64 comparing the intensities of gamma rays from photofission products with close energies,
65 for centered and peripheral sample locations.

66 We present here the first experimental step of the development of a method using
67 photofission as a probe to quantify the fissile mass in large volume heterogeneous
68 radioactive waste packages, which aims at producing cumulative photofission yields for
69 the fission products of ^{235}U and ^{238}U , and identifying fission products of interest for ^{235}U
70 vs. ^{238}U discrimination. Fission products emitting several gamma rays could be used as
71 attenuation indicators to estimate the depth at which nuclear materials are localized inside
72 the package. In this way, attenuation corrections could be applied to avoid using only close-
73 in-energy gamma rays for the discrimination method. The attenuation corrections
74 according to the depth of nuclear materials in the package, also known as matrix and
75 localization effect corrections, are also needed for the final assessment of their mass and
76 constitutes a challenge in large, dense and heterogeneous radioactive waste packages.

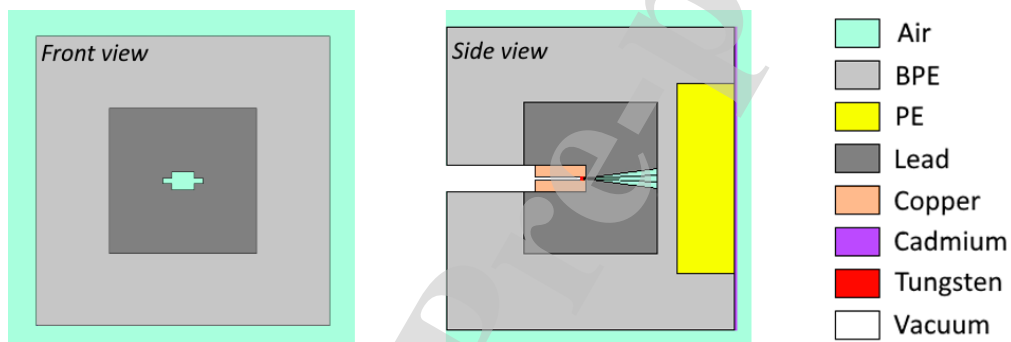
77 This paper presents the experimental setup deployed at CEA Cadarache using a high-
78 energy electron LINAC in the CINPHONIE casemate of the CHICADE facility [15],
79 uranium samples and a high-resolution gamma-ray spectrometer. The experimental and
80 numerical methods used to characterize the interrogating photon beam are then described,
81 as well as the neutron spectrum, by photon and neutron activation analysis, respectively.
82 These data are crucial to finally obtain data on 26 and 49 cumulative photofission yields
83 for respectively ^{235}U and ^{238}U fission products, which are presented at the end of this paper.

84 2. Experimental setup

85 Experiments were performed by using a Bremsstrahlung photon beam produced by a
86 LINAC located in the CINPHONIE irradiation cell at CEA Cadarache. This SATURNE
87 accelerator initially designed for medical treatments has two accelerating cavities. It has
88 been restored in the past few years in order to meet dose rates and Bremsstrahlung
89 endpoint-energies required by high-energy photon imaging and photofission experiments
90 on large and dense radioactive waste packages. At the time of the experiments presented
91 in this paper, the LINAC was operated in degraded mode and the dose rate at 1 m from the
92 conversion target in the beam axis reached only a few Gy/min (since, it has been upgraded
93 up to several dozens of Gy/min). In pulse mode, the LINAC accelerates electrons up to
94 more than 15 MeV. The electrons strike a tungsten target and a part of their kinetic energy
95 is converted into Bremsstrahlung radiation. The pulse frequency is up to 100 Hz with a
96 pulse width of 4.1 μ s and a peak current of 10 mA at the target entrance. Furthermore, a 20
97 cm thick lead collimator with two rectangular apertures, of dimensions 6.5 cm in width and
98 5.5 cm in height for the largest and 12 cm in width and 1.5 cm in height for the narrowest,
99 allows focusing the beam on the uranium samples (see further Figure 1). With this setup,
100 however, the photon flux at the output of the collimator aperture is not known precisely,
101 nor is the endpoint-energy of the Bremsstrahlung spectrum, which is expected to be around
102 15 MeV. Therefore, a characterization of the photon beam produced by the LINAC, using
103 photon activation of reference materials, is detailed in section 3. The tungsten
104 Bremsstrahlung conversion target is a 5 mm thick cylinder with a 4 mm radius, placed
105 perpendicularly to the incident electron beam. The tungsten target and the lead collimator
106 of the LINAC can both produce photoneutrons by (γ ,n) reactions when irradiated by high-
107 energy photons. The (γ ,n) reaction threshold energies range from 6.19 MeV to 8.07 MeV
108 for tungsten isotopes and from 6.74 MeV to 8.09 MeV for lead isotopes [16]. The
109 photoneutron flux expected at the location of the uranium sample could cause undesired
110 neutron reactions in addition to photofissions, such as neutron fissions that can increase the
111 delayed gamma emission and lead to an overestimation of the cumulative photofission
112 yields, or neutron captures forming activation products that can emit delayed gamma rays
113 constituting of an additional background. To limit the number of photoneutrons reaching

114 the uranium sample, a borated polyethylene (BPE) and polyethylene (PE) shield made of
 115 20 cm thick blocks has been implemented all around the lead collimator (see Figure 5).
 116 The photon beam attenuation in 20 cm of polyethylene decreases the photofission rate in
 117 the samples by a factor 1.7. In order to complete the thermal neutron absorption occurring
 118 in the BPE shield, a cadmium layer of 2 mm was added on the front face. The simulated
 119 model of the LINAC with the associated collimator and neutron shielding is shown in
 120 Figure 1.

121



122

123 *Figure 1: MCNP model of the collimator and of the neutron shielding surrounding the LINAC with the collimator*
 124 *aperture (left side) and the accelerating cavity as well as the tungsten target (right side)*

125 A sample of Depleted Uranium (DU) of 99.7 % ^{238}U content, with a mass of several
 126 hundred grams and a sample of Highly Enriched Uranium (HEU), composed of more than
 127 90 % of ^{235}U , with a mass lower than 1 g, were irradiated. The thickness and density of the
 128 DU metallic sample being respectively 1 cm and 18.96 g.cm^{-3} , significant self-attenuation
 129 effects occur both for the interrogating photon flux and the delayed gamma rays emitted
 130 by photofission products. Therefore, we apply correction factors in order to calculate the
 131 photofission product yields (see section 4.2). The second sample consists of a fissile HEU
 132 core, with a thickness well below 1 mm, a density of 18.96 g.cm^{-3} , and zirconium separation
 133 wedges with a density of 6.52 g.cm^{-3} . In addition, the fissile core is held between two
 134 Zircaloy sheets of density 6.56 g.cm^{-3} . In radioactive waste, ^{235}U is generally mixed with
 135 ^{238}U and Figure 2 shows that the photofission cross-sections of these two isotopes are of
 136 the same order of magnitude. They reach only a fraction of barn and the cross-section of
 137 ^{235}U is approximately twice larger than that of ^{238}U , see Figure 3. The photofission cross-

138 section being very low compared to the thermal neutron fission cross-sections of fissile
 139 isotopes like ^{235}U (see Figure 3), we use a LINAC to provide a high interrogating photon
 140 flux. For incoming neutrons in the MeV energy range, however, the fission cross-section
 141 is similar for uranium 235 and 238, and it is only a few barns. As the aim of these
 142 experiments is to assess the photofission product yields as precisely as possible, we will
 143 estimate the residual contribution of neutron fissions in the uranium samples in section 4.1.
 144

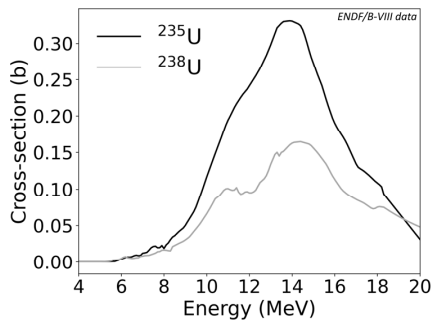


Figure 2: Photofission cross-sections of ^{235}U and ^{238}U [17]

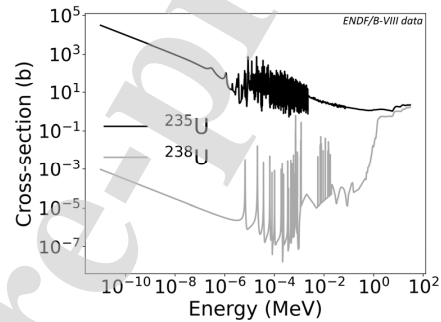


Figure 3: Neutron fission cross-sections of ^{235}U and ^{238}U [17]

145 To this aim, a set of thin metallic activation pellets was irradiated during 1 hour at 100 Hz
 146 at the same position as the uranium samples. These so-called “Neutron Activation
 147 Spectrometers” (French acronym “SNAC”) shown in Figure 4 are pure metallic samples
 148 as described in Table 1. They will also be used to characterize the Bremsstrahlung beam
 149 by photon activation, namely the photon flux and its endpoint energy, in section 3.1.

Element	Mass (g)	Diameter (mm)	Thickness (mm)	Position on Figure 4
Au	0.045	5	0.05	1
In	2.711	22	1	2 & 4
Cu	1.731	22	0.5	3
Ni	10.027	22	3	5
Mg	6.599	22	10	6

Table 1: Characteristics of the neutron activation spectrometers

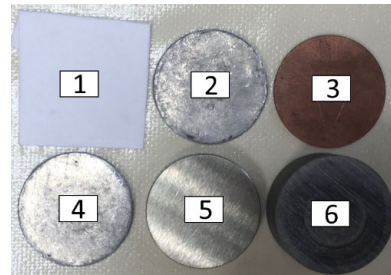


Figure 4: Position of the different materials with respect to the interrogating flux

153 The uranium samples were located at 43 cm from the LINAC tungsten target. They
 154 were fixed on the polyethylene sample holder visible in Figure 5, and irradiated one at a
 155 time. Then, an automatic transfer lasting 30 seconds, noted “cooling time” in further

156 activation analysis equations, was performed from the irradiation position to the counting
 157 position for both uranium samples. The transfer is done with a mechanical bench usually
 158 dedicated to high-energy photon tomography of radioactive waste packages in the
 159 CINPHONIE cell [18]. The samples used to characterize the Bremsstrahlung beam by
 160 photon activation were irradiated in the same position, but the transfer time to a low-
 161 background spectrometer located in another experimental room was longer. Table 2
 162 summarizes the configuration related to each sample. Note that the LINAC frequency is
 163 lowered at 10 Hz (instead of 100 Hz) for the DU sample, resulting in an interrogating
 164 photon flux decreased by a factor 10 (same peak current but fewer pulses), in order to limit
 165 the dead time due to a high photofission delayed gamma-ray emission in this large sample.

166

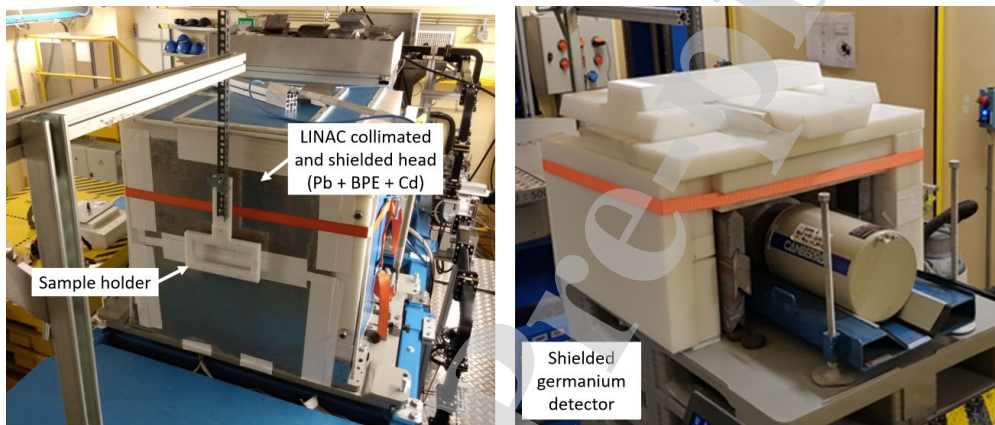
Table 2: Experimental configurations

Sample	LINAC frequency	Irradiation time	Cooling time	Counting time
DU	10 Hz	2 h	30 s	67 h
HEU	100 Hz	2 h	30 s	22 h
Activation samples (Au, In, Cu, Ni, Mg)	100 Hz	1 h	40 min	43 h

167

168 In order to measure the fission and activation products created in the uranium samples,
 169 we use a 30 % relative efficiency n-type coaxial high-purity germanium detector (HPGe,
 170 CANBERRA GR3018) with a transistor-reset preamplifier, coupled to a LYNX Digital
 171 Signal Analyzer (CANBERRA) driven by the Genie2000 software (MIRION
 172 Technologies) also used for gamma spectra analysis. The energy resolution is 1.8 keV
 173 (FWHM) at the 1332.5 keV gamma line of ^{60}Co . Although an n-type HPGe crystal is used
 174 to limit neutron damage, the detector is shielded by lead and polyethylene, as shown in
 175 Figure 5. The uranium samples in their measurement position are located 20 cm away from
 176 the detector entrance surface to keep dead time below 30 %. The dead time compensation
 177 is a live-time correction. For the DU sample measurement, dead time right after the cooling
 178 period of 30 s was 29.7 % and gamma spectra are acquired sequentially every 60 s, with a
 179 spectrum reset at the start of each sequence. For the HEU sample measurement, the initial
 180 dead time is 4.21 % and spectra acquisitions are reset every 180 s. Consequently, it was

181 possible to follow and to correct for dead time according to its time evolution. The non-
 182 nuclear metallic samples used for the beam characterization were transferred inside a low-
 183 background spectrometer with a 10 % relative efficiency HPGe detector (CANBERRA
 184 BE2020) in a lead shield, with walls covered with a copper layer to cut lead X-rays. The
 185 detector is connected to a digital spectrometer (CANBERRA DSP9960) and the resolution
 186 is 1.75 keV (FWHM) at the 1332.5 keV gamma line of ^{60}Co . At the beginning of the
 187 activation pellets measurement, the dead time was very low (0.11 %).



188
 189
 190

Figure 5: Experimental set-up inside the CINPHONIE casemate, with the head of the LINAC (left picture) and the HPGe detector (right picture)

191 3. LINAC beam characterization

192 3.1. Photon beam

193 The thin metallic samples initially intended to characterize the photoneutron flux (Au,
 194 In, Cu, Ni and Mg) finally proved to reveal also photon activation gamma rays. Their
 195 activation spectrum is presented in Figure 6.

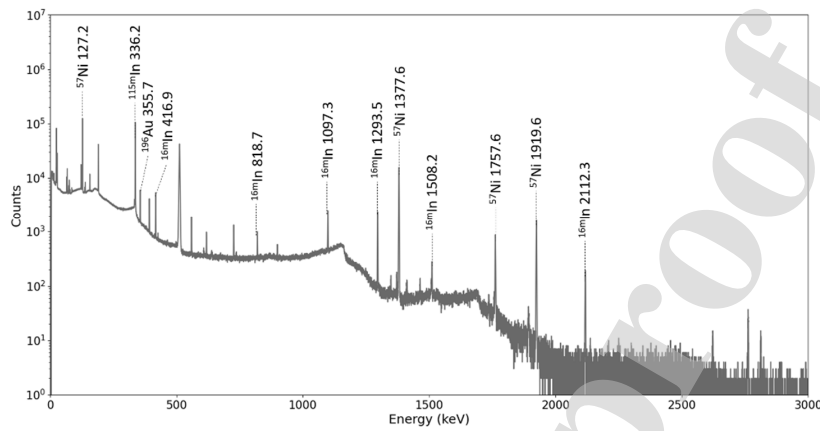


Figure 6: Activation gamma spectrum of the Au, In, Cu, Ni and Mg thin pellets

196
197

198 Especially, gamma rays due to (γ,n) reactions in gold and nickel isotopes were detected
199 (see Table 3). It is worth mentioning that gold is often used as a photon flux monitor [19]-
200 [21]. Since the characteristics of the interrogating photon beam were not known precisely,
201 the photon activation of these materials as well as the samples themselves has been
202 exploited in order to estimate the endpoint-energy of the bremsstrahlung beam and the
203 corresponding photon flux. To this aim, we need to assess the electron energy distribution
204 of the LINAC. The method we used is based on the energy-dependence of photonuclear
205 cross-sections, and on the study of radioactive isotopes produced by these photonuclear
206 reactions [22]. In addition to gold and nickel, ^{238}U of the depleted sample and ^{90}Zr present
207 in both the UZr fissile core and in the Zircaloy frame of the HEU sample were also used in
208 the spectroscopic analysis.

209 The characteristics of the activation products formed by (γ,n) reactions and leading to
210 detectable gamma rays are given in Table 3. Their half-lives and gamma-ray energies are
211 taken from JEFF-3.3 nuclear database [23]. The net areas of the gamma-ray peaks are
212 derived from the spectra analysis with the Genie2000 software. The net area statistical
213 uncertainty is $\sigma(N_{\text{counts}}) = \sqrt{N_{\text{counts}} + 2B}$, where B is the Compton background under
214 the peak.

215

Table 3: Activation gamma rays due to the (γ,n) reaction analyzed to characterize the photon beam

Activated isotope	Activation product	Half-life	γ -ray line analyzed (keV)	Net area N_{counts}
^{197}Au	^{196}Au	6.17 days	355.8	22332 ± 589
^{58}Ni	^{57}Ni	35.9 h	127.2	427713 ± 1454
^{238}U	^{237}U	6.75 days	208.0	551097 ± 1212
	^{89m}Zr	250.8 s	587.8	44363 ± 248
^{90}Zr	^{89}Zr	3.26 days	/	/
	$^{89m}\text{Y}^*$	15.7 s	909.0	182607 ± 438

216

* From the ^{89}Zr and ^{89m}Zr decays

217 Since each material has a different energy threshold and cross-section for the (γ,n) reaction,
 218 we are looking for the incident photon flux on the samples that matches the observed
 219 activation of four materials. At a distance d from the tungsten target, the experimental
 220 photon flux is given by equation (1) for gold, nickel and uranium whose gamma rays are
 221 coming from the decay of the activation product itself:

$$\Phi_{d,exp}(E_\gamma) = \frac{N_{counts}(E_\gamma) \times \lambda}{I(E_\gamma) \times \varepsilon(E_\gamma) \times f} \times \frac{1}{(1 - e^{-\lambda \cdot t_{irr}}) \times e^{-\lambda \cdot t_{cool}} \times (1 - e^{-\lambda \cdot t_{count}})} \times \frac{\Phi_{d,simulated}}{\int_{E_{th}}^{E_{max}} \varphi(E) \times \sigma(E) dE} \quad (1)$$

222 With:

- 223 - $\Phi_{d,exp}(E_\gamma)$ the experimental integral Bremsstrahlung photon flux at a distance d , in
 224 photons.cm⁻².pulse⁻¹ unit, calculated from the net number of counts in the peak of
 225 energy E_γ (i.e., photon flux corresponding to the measured activation in the
 226 samples);
- 227 - N_{counts} the net number of counts (after subtraction of the Compton background
 228 and of potential gamma interferences) in the gamma-ray line of interest;
- 229 - λ the radioactive decay constant of the activation product in s⁻¹ from JEFF-3.3
 230 nuclear database [23];
- 231 - $I(E_\gamma)$ the gamma-ray intensity (number of gamma ray of energy E_γ emitted per
 232 disintegration) taken from JEFF-3.3 nuclear database [23];
- 233 - $\varepsilon(E_\gamma)$ the absolute detection efficiency (number of count in the HPGe detector in
 234 the full energy peak of energy E_γ per gamma ray emitted at this energy in the
 235 sample), obtained via MCNP6.1 [24] Monte-Carlo simulation to account for the

- 236 attenuation effects in the sample, the experimental geometry (such as the distance
 237 to the detector) and the intrinsic efficiency of the detector;
- 238 - f the pulse repetition rate (or frequency) in pulse.s⁻¹ (Hz);
 - 239 - t_{irr} , t_{cool} and t_{count} respectively the irradiation, cooling and counting time in s;
 - 240 - $\Phi_{d,simulated}$ the integral Bremsstrahlung photon flux at distance d , obtained by
 241 MCNP simulations of the LINAC accelerated electrons impacting the tungsten
 242 target, in photons.cm⁻².incident electron⁻¹;
 - 243 - $\int_{E_{th}}^{E_{max}} \varphi(E) \times \sigma(E) dE$ the number of (γ ,n) reactions in the sample per incident
 244 electron, calculated with MCNP, corresponding to the Bremsstrahlung spectrum
 245 $\varphi(E)$ convolved with the reaction cross-section $\sigma(E)$, from the threshold value E_{th}
 246 of $\sigma(E)$ to the endpoint-energy E_{max} of $\varphi(E)$.

247 The case of ⁹⁰Zr is slightly different since ⁸⁹Zr can also be formed in an isomeric state ^{89m}Zr
 248 (587.8 keV gamma ray) following the (γ ,n) reaction, and since we are also interested by
 249 the daughter nucleus ^{89m}Y (909.0 keV gamma ray) formed by both ⁸⁹Zr and ^{89m}Zr decays.
 250 Solving Bateman evolution equations for ^{89m}Zr, ⁸⁹Zr, and ^{89m}Y (simplified owing to its
 251 short half-life) enables to determine the global ⁹⁰Zr(γ ,n) reaction rate and to calculate the
 252 flux corresponding to the observed activation of ⁹⁰Zr, with an equation a bit more complex
 253 than (1) but depending on the same parameters. Note that we do not take into account the
 254 pulsed emission of the LINAC in photon (or further neutron) activation equations, but we
 255 consider a continuous emission during the LINAC irradiation phase, because the half-lives
 256 of the observed nuclides are much longer than the LINAC period (0.01 s to 0.1 s).
 257 Since we do not have the possibility to measure the electron energy distribution of our
 258 LINAC, we assume a semi-Gaussian shape based on simulations of the electron beam
 259 dynamics performed by the LINAC manufacturer [25]. In fact, we are not looking for its
 260 exact shape, but an energy distribution leading to a good agreement between measured and
 261 calculated gamma rays of the four activated materials. The semi-Gaussian function (see
 262 further Figure 7) parametrized with two variables, the endpoint-energy and the width at
 263 half-maximum, which are adjusted together. The endpoint-energy was varied from
 264 15 MeV to 17 MeV with 0.1 MeV steps, and the width at half-maximum from 0 MeV
 265 (mono-energy distribution) to 2 MeV with 0.5 MeV steps. For each of the 105 pairs of
 266 parameters, MCNP simulations were performed by impinging electrons of the considered

267 energy distribution on the LINAC tungsten target to produce the Bremsstrahlung photon
 268 beam. Then, the number of (γ,n) reactions, in each activation sample was numerically
 269 evaluated, corresponding to the convolution of the resulting photon flux on the different
 270 materials with their reaction cross-section as explained above in (1). Finally, an
 271 experimental photon flux is calculated for each activated isotope (^{197}Au , ^{58}Ni , ^{238}U and
 272 ^{90}Zr) by using the net area of the peaks listed in Table 3. As a result, four photon fluxes are
 273 obtained for each couple of parameters. The most probable electron energy distribution is
 274 then identified as the one minimizing the squared differences between the four flux values.
 275 In other words, the most probable distribution among the 105 parametric cases gives the
 276 most consistent flux estimations for the four activation samples. In this case, the electron
 277 energy distribution with an endpoint-energy of 15.8 MeV and a 1 MeV width at half-
 278 maximum provides the more consistent photon fluxes for all the materials, as reported in
 279 Table 4.

280 *Table 4: Experimental photon fluxes calculated for the four materials with the most probable electron energy*
 281 *distribution*

Activated isotope	$\Phi_{43\text{cm},\text{exp}}$ (photons.cm ⁻² .pulse ⁻¹)
^{197}Au	$(9.02 \pm 1.26) \times 10^7$
^{58}Ni	$(10.05 \pm 1.33) \times 10^7$
^{238}U	$(10.21 \pm 1.36) \times 10^7$
^{90}Zr	$(9.43 \pm 1.25) \times 10^7$
Mean photon flux $\bar{\phi} = (9.68 \pm 1.30) \times 10^7$ photons.cm ⁻² .pulse ⁻¹	

282 The uncertainty on the experimental photon flux for each material is calculated as a
 283 quadratic combination of the main following sources of uncertainty:

- 284 - a relative uncertainty estimated to 10 % on the (γ,n) cross-section of the activated
 285 isotopes, according to the EXFOR cross-section library [26]. As an example, the
 286 $^{197}\text{Au}(\gamma,n)^{196}\text{Au}$ reaction cross-section uncertainty is about 10 % in the work of
 287 Plaisir *et al.* [27];
- 288 - a relative uncertainty of 7 % on the detection efficiency to take into account the
 289 detector intrinsic efficiency (less than 5 % according to the detector model
 290 optimization with reference sources) and the modeling of the experimental set-up
 291 (uncertainties on samples and on equipment dimensions, arbitrarily set at 5 %

- 292 based on our experience of such simulations). Intrinsic and geometric efficiency
 293 uncertainties are combined in quadratic sum, leading to 7 % relative uncertainty
 294 on detection efficiency;
- 295 - the statistical uncertainty related to MCNP simulation results, which is lower than
 - 296 5 % for all activation samples calculations;
 - 297 - the uncertainty associated to the most probable semi-Gaussian model of the
 - 298 electron energy distribution is estimated as the standard deviation of the photon
 - 299 fluxes obtained with the four materials:

$$300 \quad \frac{\sigma_{distribution}}{\bar{\phi}} = \frac{1}{\bar{\phi}} \sqrt{\frac{(\phi_{Au}-\bar{\phi})^2 + (\phi_{Ni}-\bar{\phi})^2 + (\phi_U-\bar{\phi})^2 + (\phi_{Zr}-\bar{\phi})^2}{4}} = 5 \%;$$

- 301 - the uncertainties on the radioactive decay constants of the activation products and
- 302 their gamma-ray intensities, provided by JEFF-3.3 database [23], which are lower
- 303 than 2 %;
- 304 - the uncertainty on the net area of the gamma rays reported in Table 3, which is at
- 305 most 2.6 %.

306

307 The Bremsstrahlung beam characteristics for this most probable electron energy
 308 distribution are given in Table 5.

309

Table 5: Characteristics of the LINAC beam

End point energy (electron beam)	15.8 MeV
Width at half maximum (electron beam)	1 MeV
Bremsstrahlung photon flux at 43 cm from the tungsten target (0°)	$(9.68 \pm 1.30) 10^7 \text{ photons.cm}^{-2}.\text{pulse}^{-1}$

310

311 This electron distribution is shown in Figure 7 and the corresponding Bremsstrahlung
 312 photon distribution calculated with MCNP is given in Figure 8.

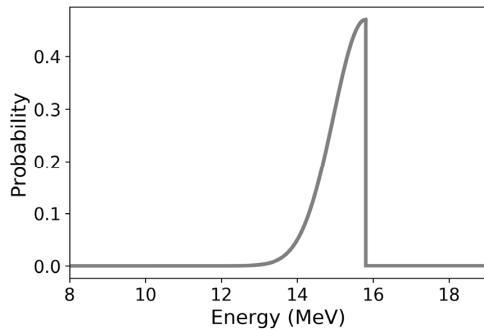


Figure 7: Most probable electron energy distribution (source of MCNP flux and reaction rate calculations)

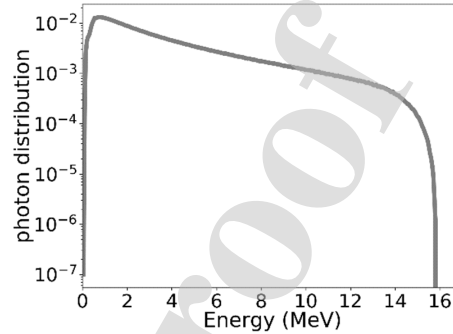


Figure 8: Bremsstrahlung photon energy distribution calculated with MCNP and the electron energy distribution of Figure 7

313

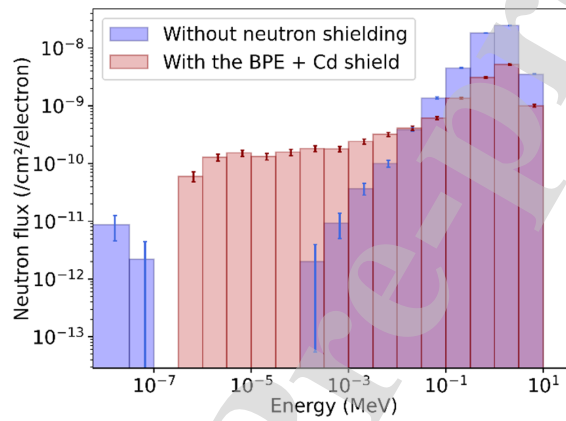
314 The mean experimental photon flux, at the center of the beam and 43 cm away from the
 315 tungsten target, is used to normalize MCNP simulation results that are given per electron
 316 impinging on the tungsten target.

317

318 3.2. Neutron spectrum

319 As mentioned in section 2, a photoneutron flux resulting from photonuclear (γ, n)
 320 reactions of high-energy Bremsstrahlung photons with high-Z materials (W and Pb) is
 321 expected at the location of the samples. The analysis of the pellets activation gamma
 322 spectrum shown in Figure 6 spotted the presence of fast neutrons through the 336.2 keV
 323 gamma-ray line of ^{115m}In , created by the $^{115}\text{In}(n, n')$ reaction with a threshold around 0.5
 324 MeV [28]. However, this contribution is merged with that of the $^{115}\text{In}(\gamma, \gamma')$ reaction with a
 325 poorly known cross-section. The observation of ^{116m}In gamma rays highlights the presence
 326 of epithermal neutrons, i.e., with an energy larger than 0.5 MeV, which are not absorbed
 327 by the cadmium foil in front of the LINAC head. On the other hand, no thermal neutrons
 328 are observed through the activation of the gold foil. Indeed we do not observe the
 329 411.8 keV gamma rays of ^{198}Au expected from the $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$ reaction. To illustrate
 330 the neutron-absorption efficiency of the LINAC head shielding, MCNP simulations of the
 331 photoneutron spectrum with and without the borated polyethylene blocks and the cadmium
 332 sheet in front of the collimator are shown in Figure 9. Polyethylene attenuates the fast
 333 neutron flux created by photonuclear reactions on the LINAC Bremsstrahlung target and
 334 on the lead collimator (76 % of photoneutrons being produced in lead), while boron and

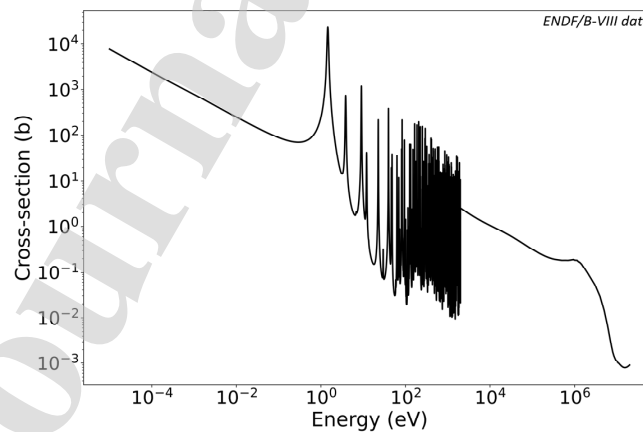
335 cadmium capture thermal neutrons. However, simulations showed that cadmium leads to
 336 the production of photoneutrons in the MeV range. Altogether, the benefit of cadmium to
 337 absorb thermal neutrons causing parasitic fissions in ^{235}U is far more important than the
 338 drawback relative to the production of photoneutrons in cadmium, which are much less
 339 numerous than fast neutrons produced inside the uranium sample (see section 4.1). Finally,
 340 the global shielding decreases the neutron flux reaching the sample by a factor 4.



341

342 *Figure 9: Simulation of the neutron energy spectrum with and without the borated polyethylene and cadmium neutron*
 343 *shielding around the LINAC head (tungsten target and lead collimator)*

344 In order to validate the MCNP model, the neutron activation of indium, through the
 345 $^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$ reaction, whose cross-section is given in Figure 10, was compared to
 346 *experimental results.*



347

348

Figure 10: $^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$ reaction cross-section, from ENDF/B-VIII.0 [17]

349 The net areas (number of counts) of the six ^{116m}In gamma rays visible in the spectrum of
 350 Figure 6 are compared with those calculated with MCNP6.1. The neutron spectrum of
 351 Figure 9 with the BPE and Cd shielding was simulated as a MCNP neutron source in front
 352 of the neutron Au, In, Cu, Ni and Mg activation samples. The resulting $^{115}\text{In}(n,\gamma)^{116m}\text{In}$
 353 reaction rate in the two indium pellets enables to estimate the net surface of ^{116m}In gamma
 354 rays:

$$N(E_i) = \frac{R(^{115}\text{In}(n,\gamma)^{116m}\text{In}) I(E_i) \varepsilon(E_i)}{\lambda} (1 - e^{-\lambda t_{\text{irr}}}) e^{-\lambda t_{\text{cool}}} (1 - e^{-\lambda t_{\text{count}}}) \quad (2)$$

355

356 With the same notation as in (1) and with $R(^{115}\text{In}(n,\gamma)^{116m}\text{In})$ the reaction rate in s^{-1} . The
 357 gamma-ray energies and intensities are found in JEFF-3.3 nuclear database [23]. The
 358 comparison between the simulated and the experimental numbers of counts is given in
 359 Table 6. The experimental number of counts corresponds to the peak surface calculated
 360 with Genie2000 software. The simulated number of counts is calculated with equation (2)
 361 and given with an uncertainty of 18 % corresponding to the quadratic combination of the
 362 13.4 % uncertainty of the photon flux used to normalize the MCNP simulation result, the
 363 7 % detection efficiency uncertainty and the 10 % uncertainty on the $^{115}\text{In}(n,\gamma)^{116m}\text{In}$
 364 reaction cross-section. The other uncertainty sources are considered negligible.

365

Table 6: Experimental vs. calculated areas in the full-energy peaks of ^{116m}In gamma rays

Energy (keV)	Intensity (%)	Experiment (counts)	Simulation (counts)	Relative gap (%)
1294.5	84.7	20250 (± 287)	19101 (± 3438)	- 6
1097.3	57.6	16937 (± 373)	15463 (± 2783)	- 9
416.9	28.0	25069 (± 429)	23465 (± 4224)	- 6
2112.3	15.3	2086 (± 87)	2013 (± 362)	- 4
818.7	11.6	4756 (± 85)	4266 (± 768)	- 10
1508.2	10.4	1942 (± 52)	1994 (± 359)	+ 3

366

367 The calculated numbers of counts are globally slightly lower than the experimental ones,
 368 but the agreement is quite satisfactory taking into account the uncertainty on the calculated
 369 thermal neutron flux exiting the neutron shielding. The latter is mainly due to the
 370 uncertainty on photoneutron production (itself coming from the uncertainty on the
 371 Bremsstrahlung beam characteristics and on the (γ,n) cross-sections of W and Pb) and on
 372 the very high neutron absorption in the BPE and Cd shielding.

373 4. Cumulative yields of ^{235}U and ^{238}U photofission products

374 4.1. Estimation of neutron vs. photofission proportions

375 MCNP simulations were conducted to characterize neutron production inside the
 376 uranium samples via (γ,n) , (n,f) and (γ,f) reactions, and their contribution to fast neutron
 377 fission (as a reminder of section 3.2, the presence of thermal neutrons was not observed),
 378 compared to photofission, which depends on sample geometry and composition of the
 379 samples, as shown in Table 7.

380

381 *Table 7: Fraction of neutrons created inside the uranium samples in the total neutron flux (including photoneutrons*
 382 *produced outside the samples) and part of neutron fissions (in all fissions including photofissions)*

	DU	HEU
Part of neutrons produced inside the sample (%)	99	66
Part of neutron fissions among total fissions (%)	5.5	0.5

383

384 For the thick Depleted Uranium (DU) sample, simulations show that neutrons produced
 385 inside the sample itself represent 99 % of the total neutron flux in the sample. They are
 386 mainly produced via (γ,n) and $(\gamma,2n)$ reactions, or during photofissions, and they induce
 387 fast neutron fissions representing 5.5 % of total fissions. For the Highly Enriched Uranium
 388 (HEU) sample, 66 % of the neutrons are created in the sample itself. Among them, 30 %
 389 are produced by (γ,xn) reactions on zirconium isotopes of the UZr fissile core and its
 390 Zircaloy support. For this HEU sample, fast neutron fissions only represent 0.5 % of total
 391 fissions because of the small thickness of the sample (neutron escape without interaction).
 392 It can be noted that photoneutron production outside uranium samples (mainly in the
 393 LINAC tungsten target and lead collimator) has no significant impact on the parasitic
 394 neutron fission rate for this experimental configuration.

395 For the purpose of calculating the photofission product yields, the photofission rate
 396 in the samples is calculated with MCNP. It significantly depends on the sample geometry
 397 and density because of self-shielding, i.e. attenuation of the Bremsstrahlung photon flux
 398 inside the sample. The validity of the DU sample numerical model was first verified by
 399 comparing the experimental and simulated (γ,n) reaction rates on ^{238}U , forming ^{237}U that
 400 emits a 208.0 keV gamma ray following its β^- decay. Given the relatively long half-life of

401 ^{237}U (almost 7 days) with respect to the irradiation time (2 hours), the experimental reaction
 402 rate can be calculated with (3) using the net area of the 208.0 keV peak.

$$R_{exp}[^{238}\text{U}(\gamma, n)^{237}\text{U}] = \frac{N_{counts}(208 \text{ keV})}{I(208 \text{ keV}) \times \varepsilon(208 \text{ keV}) \times e^{-\lambda t_{cool}} \times (1 - e^{-\lambda t_{count}}) \times t_{irr} \times f} \quad (3)$$

403

404 With the same notation as in (1) and with $R_{exp}[^{238}\text{U}(\gamma, n)^{237}\text{U}]$ the experimental reaction rate
 405 per LINAC pulse. The uncertainty on $R_{exp}[^{238}\text{U}(\gamma, n)^{237}\text{U}]$ is estimated as the quadratic
 406 combination of 7 % relative uncertainty on the detection efficiency (quadratic combination
 407 of 5 % uncertainties on both the detector intrinsic efficiency and geometric efficiency),
 408 0.3 % uncertainty on the 208.0 keV gamma-ray intensity (JEFF-3.3 database [23]), and
 409 0.22 % statistical uncertainty on the 208.0 keV gamma-ray net area (see Table 3).

410 Finally, we obtain $R_{exp}[^{238}\text{U}(\gamma, n)^{237}\text{U}] = (7.51 \pm 0.53) \times 10^5 \text{ reactions.pulse}^{-1}$.

411 For comparison, the reaction rate can be calculated with MCNP model as in (4):

$$R_{simul}[^{238}\text{U}(\gamma, n)^{237}\text{U}] = \int_{E_{th}}^{E_{max}} \varphi(E) \times \sigma_{(\gamma, n)}(E) dE \times \frac{\Phi_{exp}}{\Phi_{simulated}} \quad (4)$$

412 With:

- 413 - $R_{simul}[^{238}\text{U}(\gamma, n)^{237}\text{U}]$ the simulated reaction rate per LINAC pulse;
- 414 - $\int_{E_{th}}^{E_{max}} \varphi(E) \times \sigma_{(\gamma, n)}(E) dE$ the number of (γ, n) reactions in the DU sample per source
 415 electron, calculated with MCNP, corresponding to the Bremsstrahlung spectrum
 416 $\varphi(E)$ convolved with the (γ, n) reaction cross-section $\sigma_{(\gamma, n)}(E)$, from the threshold
 417 value E_{th} of $\sigma_{(\gamma, n)}(E)$ to the endpoint-energy E_{max} of $\varphi(E)$;
- 418 - Φ_{exp} the integral Bremsstrahlung photon flux in front of the DU sample reported
 419 in section 3.1 (photon activation measurements with four material targets):
 420 $(9.68 \pm 1.30) \times 10^7 \text{ photons.cm}^{-2}.\text{pulse}^{-1}$;
- 421 - $\Phi_{simulated}$ the integral Bremsstrahlung photon flux in front of the DU sample,
 422 obtained by MCNP simulations of LINAC accelerated electrons impacting the
 423 tungsten target, in $\text{photons.cm}^{-2}.\text{source electron}^{-1}$.
- 424 - Note that the term $\frac{\Phi_{exp}}{\Phi_{simulated}}$ provides the number of source electrons per LINAC
 425 pulse, necessary to normalize the MCNP simulation result of the reaction rate
 426 $\int_{E_{min}}^{E_{max}} \varphi(E) \times \sigma_{(\gamma, n)}(E) dE$ (in reactions per source electron).

427 The uncertainty on $R_{simul}[{}^{238}\text{U}(\gamma, n){}^{237}\text{U}]$ is calculated as the quadratic combination of the
 428 13.4 % relative uncertainty on Φ_{exp} as reported in section 3.1, the 10 % uncertainty on the
 429 ${}^{238}\text{U}(\gamma, n){}^{237}\text{U}$ cross-section in accordance with the EXFOR library [26], and the 0.4 %
 430 statistical uncertainty on the Monte-Carlo calculation for the ${}^{238}\text{U}(\gamma, n){}^{237}\text{U}$ reaction rate.
 431 Finally, we obtain $R_{simul}[{}^{238}\text{U}(\gamma, n){}^{237}\text{U}] = (7.15 \pm 1.19) \times 10^5 \text{ reactions.pulse}^{-1}$.
 432 We obtain a satisfactory agreement between the experimental and simulated ${}^{238}\text{U}(\gamma, n){}^{237}\text{U}$
 433 reaction rates per LINAC pulse, which worth respectively $(7.51 \pm 0.53) \times 10^5$ and
 434 $(7.15 \pm 1.19) \times 10^5$, reinforcing our confidence in the numerical model of the DU sample
 435 and in the Bremsstrahlung photon beam determined in section 3.1. Consequently, we can
 436 simulate the photofission rate in the samples, defined by (5) for a mixture of ${}^{235}\text{U}$ and ${}^{238}\text{U}$.

$$\tau_p = \frac{N_A}{\mathcal{M}} \left(m_5 \int_{E_{th}}^{E_{max}} \varphi(E) \sigma_{5(\gamma, f)}(E) dE + m_8 \int_{E_{th}}^{E_{max}} \varphi(E) \sigma_{8(\gamma, f)}(E) dE \right) \quad (5)$$

437 Where:

- 438 - τ_p is the photofission rate in s^{-1} ;
- 439 - N_A is the Avogadro constant, equal to $6.02 \times 10^{23} \text{ mol}^{-1}$;
- 440 - \mathcal{M} is the molar mass of the uranium mixture, expressed in g.mol^{-1} ;
- 441 - m_5 and m_8 are respectively the ${}^{235}\text{U}$ and ${}^{238}\text{U}$ masses in the sample, in g;
- 442 - E_{th} and E_{max} are respectively the threshold energy of the photofission reactions,
 443 around 6 MeV, and the Bremsstrahlung endpoint-energy, 15.8 MeV in this case;
- 444 - $\varphi(E)$ is the Bremsstrahlung photon flux at the energy E in the sample;
- 445 - $\sigma_{5(\gamma, f)}(E)$ and $\sigma_{8(\gamma, f)}(E)$ are the photofission reaction cross-sections at the energy
 446 E for ${}^{235}\text{U}$ and ${}^{238}\text{U}$, respectively.

447
 448 The uncertainty on the photofission rate calculated with MCNP is the quadratic
 449 combination of the following uncertainties:

- 450 - 13.4 % relative uncertainty on the bremsstrahlung photon flux, determined in
 451 section 3.1, $(9.68 \pm 1.30) \times 10^7 \text{ photons.cm}^{-2}.\text{pulse}^{-1}$ (see Table 5);
- 452 - 0.4 % and 0.8 % relative statistical uncertainties on MCNP calculations for the
 453 photofission rate in the DU and HEU samples, respectively;
- 454 - 2 % uncertainty on the photofission cross-section, according to datasets available
 455 in the EXFOR library [26];

456 - 2 % uncertainty on the uranium mass of the HEU sample, and a negligible
 457 uncertainty (smaller than 1 %) on the uranium mass of the DU sample.

458 Finally, using the characteristics of the beam (electron energy distribution and photon flux)
 459 and the experimental position of the samples with respect to the tungsten target, the
 460 photofission rates in the samples calculated with MCNP are:

$$461 \quad \tau_{p,DU} = (2.80 \pm 0.38) \cdot 10^6 \text{ s}^{-1}$$

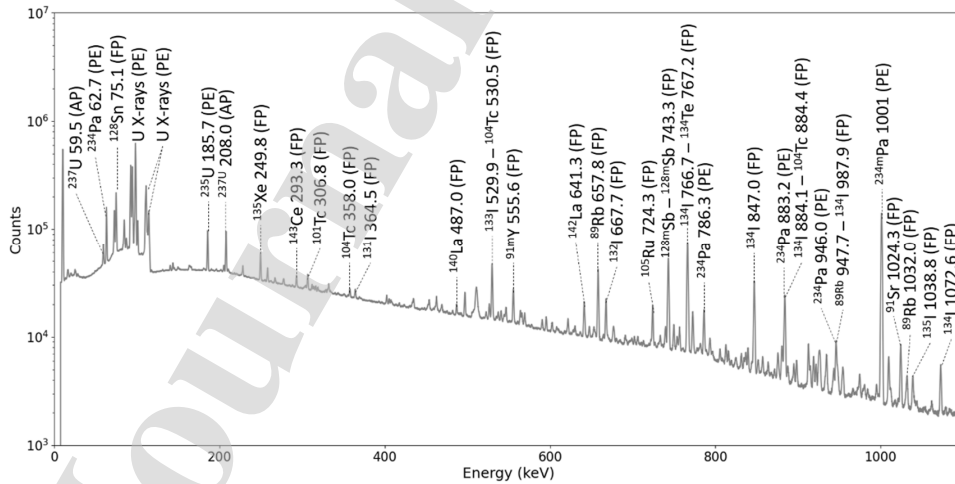
$$462 \quad \tau_{p,HEU} = (1.04 \pm 0.14) \cdot 10^5 \text{ s}^{-1}$$

463 Given the 0.3 % ^{235}U content of the DU sample, we consider that all fissions occur on ^{238}U .
 464 As a result, the delayed gamma rays measured with the DU sample are used to directly
 465 calculate the ^{238}U photofission products cumulative yields. In the HEU sample, however,
 466 3.7 % of photofissions occur on ^{238}U nuclei and 96.3 % on ^{235}U nuclei. Therefore, the ^{238}U
 467 contribution to photofission will be subtracted to calculate the ^{235}U photofission products
 468 cumulative yields.

469

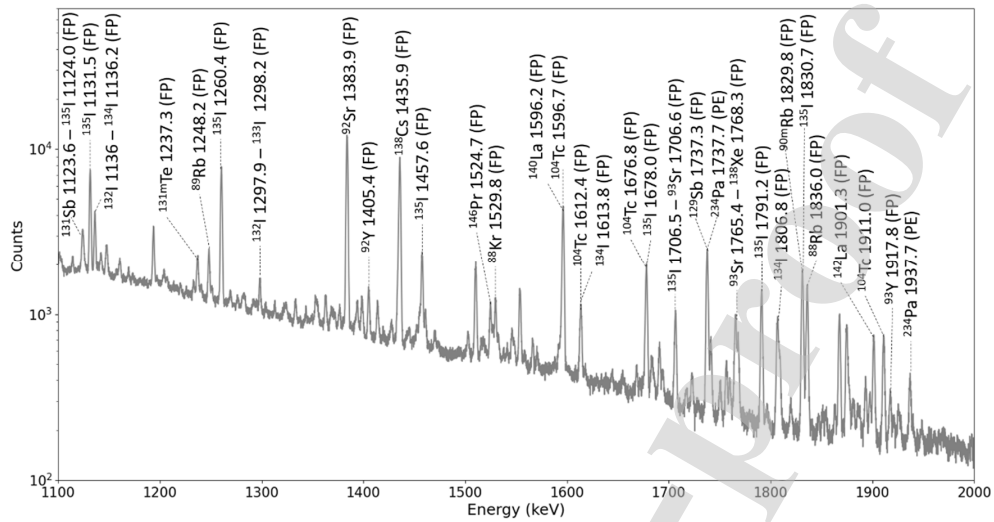
470 4.2. Photofission product yields computation

471 Figures 11 to 13 show the delayed gamma spectrum of the DU sample recorded during
 472 67 h, after a 2 h irradiation and a 30 s cooling time. The notations used are PE for Passive
 473 Emission, AP for Activation Product and FP for Fission Product.



474

475 Figure 11: Delayed gamma spectrum of the DU sample (0-1100 keV) recorded during 67 h, after a 2 h irradiation and a
 476 30 s cooling time

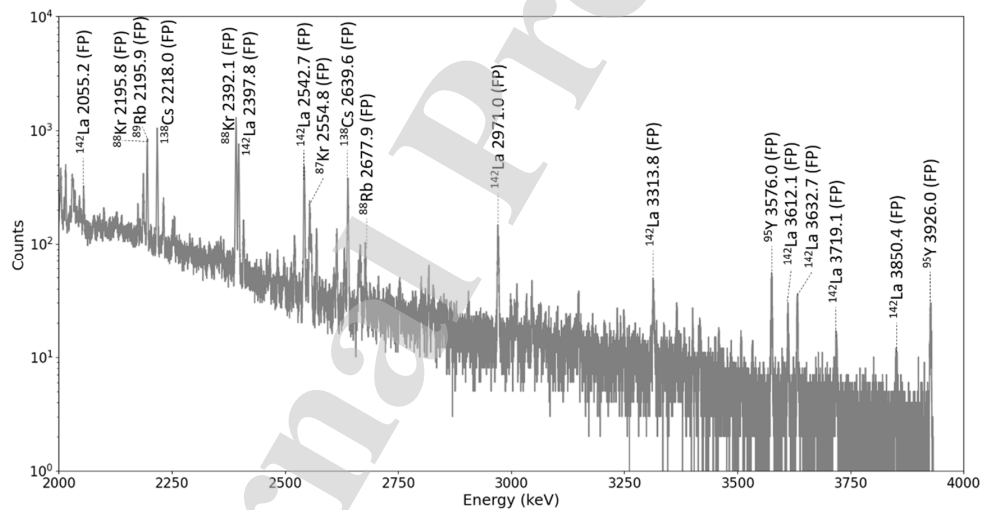


477

478

479

Figure 12: Delayed gamma spectrum of the DU sample (1100-2000 keV) recorded during 67 h, after a 2 h irradiation and a 30 s cooling time



480

481

482

483

Figure 13: Delayed gamma spectrum of the DU sample (2000-4000 keV) recorded during 67 h, after a 2 h irradiation and a 30 s cooling time

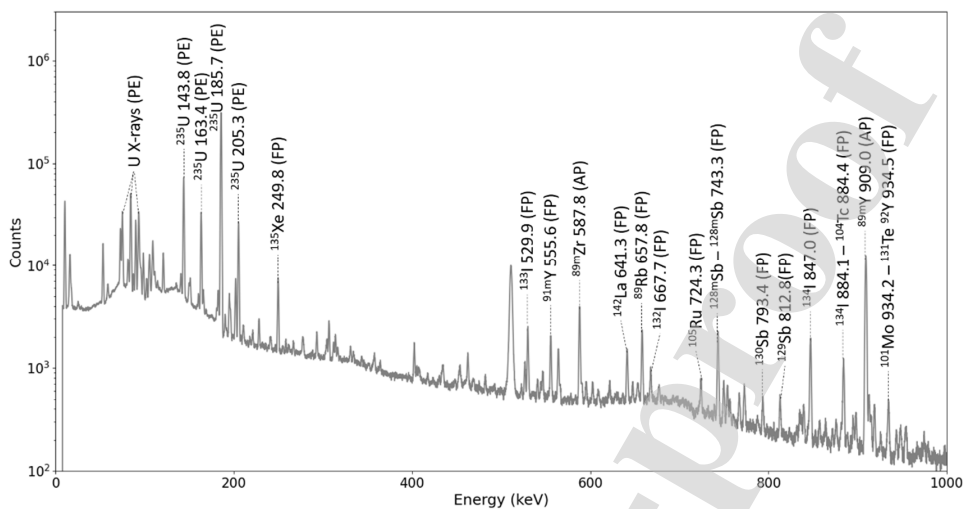
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Figures 14 to 16 show the delayed gamma spectrum of the HEU sample recorded during 22 h, after a 2 h irradiation and a 30 s cooling time. The notations are the same as those for Figures 11 to 13.

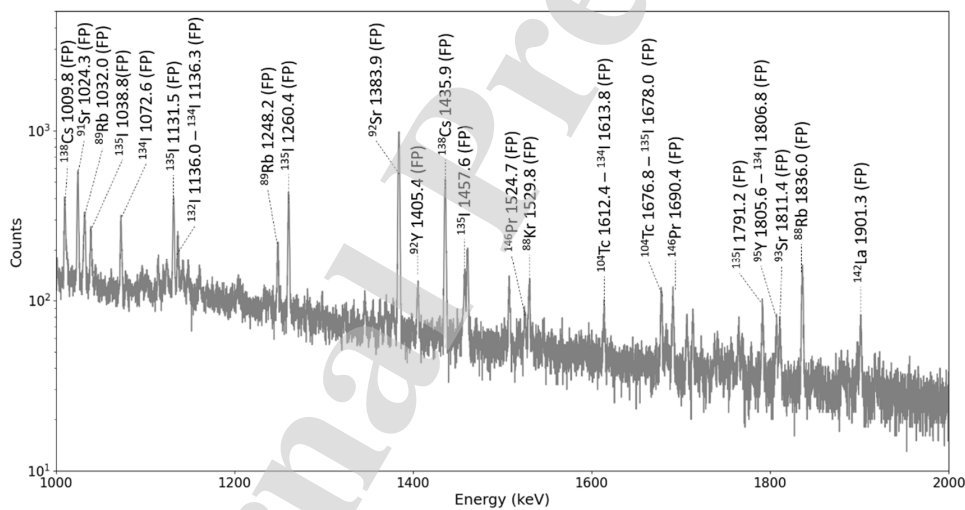


488

489

490

Figure 14: Delayed gamma spectrum of the HEU sample (0-1000 keV) recorded during 22 h, after a 2 h irradiation and a 30 s cooling time

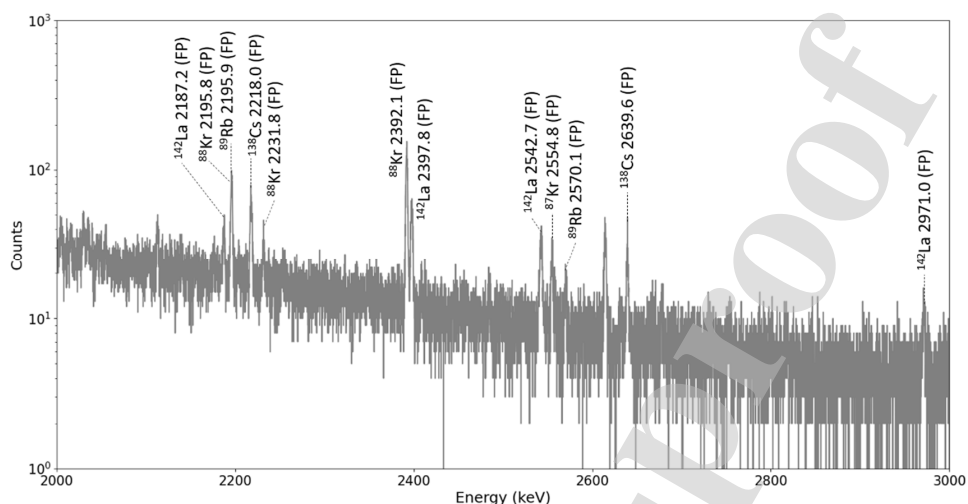


491

492

493

Figure 15: Delayed gamma spectrum of the HEU sample (1000-2000 keV) recorded during 22 h, after a 2 h irradiation and a 30 s cooling time



494

495

496

Figure 16: Delayed gamma spectrum of the HEU sample (2000-3000 keV) recorded during 22 h, after a 2 h irradiation and a 30 s cooling time

497

498 For the DU sample, the delayed gamma-ray spectra have been recorded by sequences of
 499 60 s during the almost three days of acquisition, which allows a spectrum analysis with
 500 different cooling and measurement times to limit some interferences between gamma rays
 501 close in energy, by exploiting the period differences of their emitting isotopes. For the HEU
 502 sample, the acquisition sequences lasted 180 s and the total counting time almost one day.
 503 The spectra analysis is performed with the MAGIX code developed by CEA LIST, in
 504 collaboration with CEA DES to analyze complex gamma- and X-ray spectra measured with
 505 HPGe detectors. This automatic software, based on CEA LIST know-how in complex
 506 spectrum processing [29][30], performs a complete analysis of the spectrum: energy
 507 calibration, identification of radionuclides, peak deconvolution, determination of a relative
 508 detection efficiency, activity calculation for each radionuclide if the absolute efficiency is
 509 provided by the end-user, and otherwise activity ratios. One of its main features is to
 510 include iterative steps with the automatic analysis of gamma- and X-rays JEFF-3.3
 511 database [23] in order to identify all the peaks associated to the radionuclides potentially
 512 present in the list provided by the user.

513

514 Theoretically speaking, the fission products created during the irradiation are part of
 515 radioactive decay chains and their activities can be calculated by solving Bateman

516 equations [31]. In general, these equations can be simplified by considering only the
 517 activation of the photofission product emitting the delayed gamma rays of interest, as
 518 below in (6). However, as explained further to introduce (7), we sometimes also need to
 519 consider the direct precursor of the photofission product of interest which are respectively
 520 called the father and daughter nuclides, like in the work of Kahane *et al.* [5] and Carrel *et*
 521 *al.* [6].

522 When the delayed gamma rays are emitted by a nucleus with a much longer half-life than
 523 its precursors and the cooling time, the net area $N(E_i)$ of its gamma rays of energy E_i is
 524 directly related to ^{235}U and ^{238}U cumulative photofission yields $Y_{c5,p}$ and $Y_{c8,p}$ through
 525 equation (6):

$$N(E_i) = \frac{I(E_i) \varepsilon(E_i)}{\lambda_j} (1 - e^{-\lambda t_{irr}}) e^{-\lambda t_{cool}} (1 - e^{-\lambda t_{count}}) [\tau_p (\eta_{8,p} Y_{c8,p} + \eta_{5,p} Y_{c5,p}) + \tau_n (\eta_{8,n} Y_{c8,n} + \eta_{5,n} Y_{c5,n})] \quad (6)$$

526 With the same notations as in (1) and (2), and:

- 527 - $\varepsilon(E_i)$ the absolute detection efficiency taking into account, in addition to the
 528 abovementioned intrinsic detector and geometric efficiencies, the interrogating
 529 photon flux self-shielding and delayed gamma self-attenuation in the uranium
 530 sample, both estimated with MCNP. For example, regarding the self-shielding in
 531 the DU sample, the photofission rate is 2,5 times higher on the entrance surface of
 532 the sample, with respect to the LINAC photon beam, than on its rear exit surface.
 533 Concerning self-attenuation, for instance, less than 60 % of 1 MeV delayed gamma
 534 rays emitted in the DU sample manage to escape from it;
- 535 - λ_j the radioactive decay constant of the fission product j in s^{-1} ;
- 536 - τ_p and τ_n the photofission and neutron fission rates in the sample, in s^{-1} ;
- 537 - $Y_{c5,p}$ and $Y_{c8,p}$ the cumulative photofission yields of photofission product j ,
 538 respectively for ^{235}U and ^{238}U ;
- 539 - $Y_{c5,n}$ and $Y_{c8,n}$ the cumulative fast neutron fission product yields of fission product
 540 j , respectively for ^{235}U and ^{238}U , taken from JENDL database [32];
- 541 - $\eta_{5,p}$ and $\eta_{8,p}$ are the fractions of photofissions occurring respectively in ^{235}U and
 542 ^{238}U , determined via MCNP simulation;
- 543 - $\eta_{5,n}$ and $\eta_{8,n}$ are the fractions of neutron fissions occurring respectively in ^{235}U
 544 and ^{238}U , determined via MCNP simulations.

545 However, when the photofission product of interest is a daughter nuclide in a decay chain
 546 with a father having a similar half-life (for instance ^{134}I and ^{138}Cs , which are respectively
 547 the daughters of ^{134}Te and ^{138}Xe , see radioactive periods in Table 8), its net peak areas are
 548 given by (7).

$$N(E_i) = I(E_i)\varepsilon(E_i)(\tau_p[\eta_{8,p}(Y_{i8,p}^d f_d + Y_{c8,p}^f f_f) + \eta_{5,p}(Y_{i5,p}^d f_d + Y_{c5,p}^f f_f)] \dots \dots + \tau_n[\eta_{8,n}(Y_{i8,n}^d f_d + Y_{c8,n}^f f_f) + \eta_{5,n}(Y_{i5,n}^d f_d + Y_{c5,n}^f f_f)]) \quad (7)$$

549 Where:

- 550 - $I(E_i)$, $\varepsilon(E_i)$, τ_p , τ_n , $\eta_{8,p}$, $\eta_{5,p}$, $\eta_{8,n}$ and $\eta_{5,n}$ keep the same meaning as in (6);
- 551 - $Y_{c5,p}^f$ and $Y_{c8,p}^f$ are the cumulative yields of the father nuclide, respectively for the
 552 photofission of ^{235}U and ^{238}U ;
- 553 - $Y_{i5,p}^d$ and $Y_{i8,p}^d$ are the independent yields of the daughter nuclide, respectively for
 554 the photofission of ^{235}U and ^{238}U . The independent yield (in %) of a photofission
 555 product corresponds to the number of nuclei created per 100 photofissions of the
 556 considered actinide, right after the prompt neutron emission but before the delayed
 557 neutron emission. These values will be calculated to determine the cumulative
 558 photofission yield of the daughter nuclide, which is the sum of its independent
 559 yield together with that of its precursor. Note that as in Kahane *et al.* [5] and Carrel
 560 *et al.* [6], we only consider one precursor since the precursors of the father nuclide
 561 have a relatively short half-life compared to the cooling time of these experiments.
 562 In this case, we can write $Y_{c,p}^d = Y_{i,p}^d + Y_{c,p}^f$
- 563 - $Y_{c5,n}^f$ and $Y_{c8,n}^f$ are the cumulative yields of the father nuclide, respectively for the
 564 neutron fission of ^{235}U and ^{238}U . These values are taken from JENDL database
 565 [32];
- 566 - $Y_{i5,n}^d$ and $Y_{i8,n}^d$ are the independent yields of the daughter nuclide, respectively for
 567 the neutron fission of ^{235}U and ^{238}U . These values are taken from JENDL database
 568 [32];
- 569 - The terms f_d and f_f describe the evolution of the number of nuclei over time, and
 570 are given by:

$$f_d = \frac{1}{\lambda_d} (1 - e^{-\lambda_d t_{irr}}) e^{-\lambda_d t_{cool}} (1 - e^{-\lambda_d t_{count}})$$

$$f_f = \frac{1}{\lambda_d - \lambda_f} \left[\frac{\lambda_d}{\lambda_f} (1 - e^{-\lambda_f t_{irr}}) e^{-\lambda_f t_{cool}} (1 - e^{-\lambda_f t_{count}}) \dots \right. \\ \left. \dots - \frac{\lambda_f}{\lambda_d} (1 - e^{-\lambda_d t_{irr}}) e^{-\lambda_d t_{cool}} (1 - e^{-\lambda_d t_{count}}) \right]$$

571 With λ_d and λ_f the radioactive decay constants of the daughter and father nuclides
 572 (in s^{-1}), respectively, and t_{irr} , t_{cool} , t_{count} the irradiation, cooling and counting
 573 times (in s).

574

575 Note that when the father nuclide half-life is much shorter than that of the daughter nuclide,
 576 the cumulative yield of the daughter nuclide can be estimated with (6) by analyzing the
 577 delayed gamma spectrum after a cooling time equal to six times the half-life of the father
 578 nuclide (corresponding to the decay of 98.5 % of father nuclei). This approach is used in
 579 practice for the majority of fission products, the precursors of which having very short half-
 580 lives. However, in some instances, the half-life of the precursor is a few minutes, or dozen
 581 minutes, and this approach requires that a sufficient number of daughter nuclei are still
 582 present after the abovementioned cooling time, so that their delayed gamma rays can be
 583 counted with sufficient statistics. On the contrary, when the daughter nuclide half-life is
 584 very short compared to that of the father nuclide, (6) can be used to determine the
 585 independent yield of the daughter nuclide instead of its cumulative yield.

586

587 Concerning the DU sample enrichment of only 0.3 % (^{238}U), the measured delayed gamma
 588 rays directly lead to the photofission products cumulative yields of ^{238}U . However,
 589 according to Table 7, neutron fissions on ^{238}U represent 5.5 % of total fissions in the
 590 sample. Therefore, we use the ^{238}U fast neutron fission yields available in JENDL database
 591 [32] to subtract the neutron fission contribution to the net areas of the gamma rays emitted
 592 by the fission products created in this sample. For the HEU sample, the neutron fissions
 593 represent only 0.5 % among total fissions, so we decided to neglect this contribution, but
 594 we subtract the 3.7 % of photofissions in ^{238}U nuclei present in the HEU sample to calculate
 595 the ^{235}U photofission products cumulative yields. For this purpose, we use the photofission
 596 yields obtained in this work for ^{238}U to evaluate the ^{235}U yields.

597 Finally, 26 and 49 photofission products have been identified in the spectra from their
 598 delayed gamma rays, respectively for ^{235}U and ^{238}U , and their cumulative photofission
 599 yields calculated from (6) or (7). The cumulative photofission yields measured for ^{238}U and
 600 ^{235}U are given respectively in Table 8 and Table 10 under the denomination “This work”.
 601 Note that when several delayed gamma rays are measured for a photofission product, we
 602 calculate a weighted average of the cumulative yields obtained with all the peaks that are
 603 correctly processed by MAGIX software (outliers of the net areas, for instance due to
 604 interferences, are discarded). The weighting coefficient for each gamma ray is defined as
 605 the inverse of the squared relative uncertainty on the net peak area, as described in (8):

$$\bar{y}_c = \frac{\sum_{i=1}^N \frac{y_{c,i}(E_i)}{\left(\frac{\sigma(N(E_i))}{N(E_i)}\right)^2}}{\sum_{i=1}^N \frac{1}{\left(\frac{\sigma(N(E_i))}{N(E_i)}\right)^2}} \quad (8)$$

606 Where $y_{c,i}(E_i)$ is the cumulative photofission yield calculated with the net peak area $N(E_i)$
 607 of the gamma ray of energy E_i , and $\sigma(N(E_i))$ is the statistical uncertainty calculated by
 608 $\sigma(N(E_i)) = \sqrt{N(E_i) + 2B}$, B being the Compton background under this peak.

609

610 The uncertainty associated to the average cumulative yield is calculated with a quadratic
 611 propagation of the main uncertainties listed below:

- 612 - the uncertainty on the photofission rate in the samples, respectively 13.6 % for the
 613 DU sample and 13.4 % for the HEU sample (refer to section 4.1). This uncertainty
 614 is the largest one and mainly comes from the uncertainty on the interrogating
 615 Bremsstrahlung photon flux. It could be reduced by accurately measuring the (γ,n)
 616 cross-sections of the activation materials used to characterize the photon beam,
 617 since their current uncertainties are around 10 % in EXFOR library [26];
- 618 - the uncertainty on the absolute detection efficiency of the gamma ray of energy E_i ,
 619 which is estimated to 7 %;

620 - the relative statistical uncertainty on the weighted average (8), $\frac{\sigma_{stat}(\bar{y}_c)}{\bar{y}_c}$, with

$$621 \quad \sigma_{stat}(\bar{y}_c) = \sqrt{\frac{1}{\sum_{i=1}^N \left(\frac{N(E_i)}{\sigma(N(E_i))} \right)^2}}.$$

622 We provide in Table 9 and Table 11 all the details of the delayed gamma-ray analysis for
623 ^{238}U and ^{235}U , respectively: cooling and counting times for each delayed gamma ray,
624 energy and intensity coming from JEFF-3.3 database [23] **Error! Reference source not**
625 **found.**, net peak area with its associated uncertainty and cumulative yield computed for
626 each line.

627 Our work is then compared to the values of photofission products cumulative yields
628 previously published. Note that we report a simple average of the yields when several
629 gamma rays are given in the other publications. The characteristics of the interrogating
630 photon beams are indicated in the first two lines of Table 8 and Table 10. For
631 Bremsstrahlung photon beams, the energy indicated corresponds to the endpoint energy.
632 The nuclides half-lives are from JEFF-3.3 database [23], except for ^{92}Sr because it is not
633 consistent in the different databases, and therefore its half-life is taken from Leconte *et al.*
634 [33].

635 *Table 8: Cumulative yields (number of photofission product per 100 fissions) for the photofission of ^{238}U and*
636 *comparison with published data*

Photon beam type	Bremsstrahlung	Neutron-capture gamma rays	Bremsstrahlung	Bremsstrahlung	Bremsstrahlung	Bremsstrahlung	Bremsstrahlung	Monoenergetic	
Energy (MeV)	15.8	7.8	16.3	10.0	14.987	8.0	22.0	13.0	
Fission Product	T _{1/2}	This work	Kahane 1985 [5]	Carrel 2011 [6]	Naik 2011 [7]	Naik 2013 [8]	Naik 2014 [9]	Wen 2016 [10]	Krishichayan 2019 [11]
^{84}Br	31.8 min	1.37 ± 0.20	-	-	-	0.90 ± 0.06	0.30 ± 0.06	-	2.49 ± 0.19
^{87}Kr	1.3 h	1.96 ± 0.31	1.82 ± 0.21	-	1.61 ± 0.20	1.86 ± 0.30	1.11 ± 0.13	1.0 ± 0.06	2.29 ± 0.16
^{88}Kr	2.8 h	2.24 ± 0.34	-	2.52 ± 0.23	2.77 ± 0.53	2.58 ± 0.19	2.77 ± 0.38	1.0 ± 0.03	2.92 ± 0.17
^{88}Rb	17.8 min	2.68 ± 0.79	-	2.82 ± 0.19	-	-	-	-	-
^{89}Rb	15.4 min	3.50 ± 0.55	2.51 ± 0.40	3.30 ± 0.20	-	3.12 ± 0.16	3.42 ± 0.34	1.4 ± 0.13	-
^{90}Rb	2.6 min	1.67 ± 0.27	-	-	-	-	-	-	-
^{90m}Rb	4.3 min	1.06 ± 0.17	-	-	-	-	-	-	-
^{91}Sr	9.7 h	3.86 ± 0.60	3.81 ± 0.45	4.53 ± 0.22	3.82 ± 0.17	3.69 ± 0.23	4.75 ± 0.48	-	5.27 ± 0.18
^{91m}Y	49.7 min	2.35 ± 0.49	3.84 ± 0.44	4.43 ± 0.21	-	-	-	-	-
^{92}Sr	2.6 h	4.04 ± 0.63	-	4.77 ± 0.22	3.83 ± 0.45	4.26 ± 0.13	4.59 ± 0.47	1.7 ± 0.02	4.82 ± 0.15
^{92}Y	3.5 h	4.79 ± 0.81	4.16 ± 0.21	-	-	-	-	-	8.48 ± 0.96
^{93}Sr	7.4 min	4.44 ± 0.84	-	4.90 ± 0.23	-	-	-	-	-
^{93}Y	10.2 h	4.70 ± 0.94	-	-	5.47 ± 0.76	4.00 ± 0.21	3.91 ± 0.40	-	9.53 ± 1.45
^{94}Sr	1.3 min	4.35 ± 0.68	-	4.97 ± 0.23	-	-	-	-	-

⁹⁴ Y	18.7 min	4.48 ± 0.77	-	5.06 ± 0.24	-	4.47 ± 0.25	4.25 ± 0.47	-	7.50 ± 0.46
⁹⁵ Y	10.3 min	4.85 ± 0.72	-	5.84 ± 0.39	-	-	6.20 ± 0.69	-	-
⁹⁷ Zr	16.7 h	5.95 ± 0.93	5.89 ± 0.66	-	5.43 ± 0.19	5.78 ± 0.17	6.00 ± 0.64	-	6.20 ± 0.19
⁹⁹ Mo	2.7 d	4.65 ± 0.77	-	-	4.84 ± 0.44	5.11 ± 0.15	4.75 ± 0.50	-	6.57 ± 0.22
^{99m} Tc	6 h	4.36 ± 0.70	-	-	-	-	-	-	-
¹⁰¹ Mo	14.6 min	5.37 ± 0.86	-	6.78 ± 0.32	-	7.13 ± 0.30	7.56 ± 0.77	-	-
¹⁰¹ Tc	14.2 min	5.61 ± 1.20	6.98 ± 0.82	-	-	-	-	-	-
¹⁰⁴ Tc	18.3 min	3.60 ± 0.56	4.13 ± 0.50	-	-	3.65 ± 0.28	4.06 ± 0.42	-	4.44 ± 0.64
¹⁰⁵ Ru	4.4 h	2.76 ± 0.44	2.95 ± 0.45	-	2.57 ± 0.21	2.55 ± 0.06	2.91 ± 0.38	-	4.06 ± 0.20
¹⁰⁵ Rh	1.5 d	2.78 ± 0.49	2.85 ± 0.38	-	2.63 ± 0.28	2.58 ± 0.05	2.98 ± 0.40	-	3.42 ± 0.11
¹²⁸ Sn	59.1 min	0.57 ± 0.09	-	-	-	0.85 ± 0.04	0.21 ± 0.03	-	1.22 ± 0.22
¹²⁸ Sb	9 h	0.21 ± 0.03	-	0.16 ± 0.01	-	-	-	-	-
^{128m} Sb	10.4 min	1.73 ± 0.25	-	-	-	-	-	-	-
¹²⁹ Sb	4.4 h	1.09 ± 0.17	0.54 ± 0.10	1.33 ± 0.06	1.48 ± 0.33	1.35 ± 0.16	0.63 ± 0.06	-	2.59 ± 0.09
¹³⁰ Sn	3.7 min	0.47 ± 0.10	-	-	-	-	-	-	-
¹³⁰ Sb	39.5 min	0.79 ± 0.12	-	1.08 ± 0.05	-	-	-	-	1.61 ± 0.11
¹³¹ Sb	23 min	2.51 ± 0.37	-	3.94 ± 0.19	-	4.18 ± 0.18	2.40 ± 0.27	-	5.01 ± 0.33
^{131m} Te	1.3 d	0.70 ± 0.10	3.12 ± 0.40	-	-	-	-	-	-
¹³² Te	3.2 d	4.71 ± 0.73	2.43 ± 0.50	-	4.84 ± 0.46	5.48 ± 0.14	6.15 ± 0.65	-	5.44 ± 0.25
¹³² I	2.3 h	4.87 ± 0.76	3.74 ± 0.46	-	-	-	-	-	-
¹³³ Sb	2.5 min	2.59 ± 0.42	-	-	-	-	-	-	-
^{133m} Te	55.4 min	3.23 ± 0.44	2.35 ± 0.39	4.43 ± 0.21	-	-	-	3.8 ± 0.42	-
¹³⁴ Te	41.8 min	5.29 ± 0.84	6.25 ± 0.89	6.34 ± 0.30	8.27 ± 0.26	7.23 ± 0.33	7.21 ± 0.74	-	7.43 ± 0.51
¹³⁴ I	52.5 min	7.30 ± 1.16	6.29 ± 0.94	-	-	8.06 ± 0.34	8.63 ± 0.87	3.1 ± 0.19	-
¹³⁵ I	6.6 h	5.85 ± 0.92	5.91 ± 0.68	6.66 ± 0.42	5.88 ± 0.57	5.57 ± 0.12	6.55 ± 0.67	2.6 ± 0.16	6.13 ± 0.29
^{135m} Xe	15.3 min	1.32 ± 0.43	-	-	-	-	-	-	-
¹³⁸ Xe	14.1 min	3.75 ± 0.59	5.38 ± 0.90	6.60 ± 0.58	-	-	5.91 ± 0.63	1.8 ± 0.41	-
¹³⁸ Cs	33.4 min	5.91 ± 0.86	6.10 ± 0.71	-	8.00 ± 0.48	6.84 ± 0.25	6.44 ± 0.68	2.6 ± 0.10	-
¹⁴⁰ Cs	64 s	5.65 ± 0.92	-	-	-	-	-	-	-
¹⁴⁰ La ^(a)	1.7 d	5.72 ± 0.92	-	-	-	-	-	-	-
¹⁴¹ Ba	18.3 min	4.75 ± 0.73	-	-	-	4.51 ± 0.23	5.44 ± 0.56	-	-
¹⁴² Ba	10.6 min	4.38 ± 0.78	-	4.66 ± 0.22	-	4.38 ± 0.29	4.68 ± 0.50	-	-
¹⁴² La	1.5 h	4.89 ± 0.88	3.69 ± 0.43	5.01 ± 0.24	5.26 ± 0.52	4.69 ± 0.20	4.88 ± 0.52	1.7 ± 0.13	5.58 ± 0.18
¹⁴³ Ce	1.4 d	4.39 ± 0.68	5.38 ± 0.62	-	4.27 ± 0.29	4.74 ± 0.14	4.73 ± 0.53	-	4.92 ± 0.15
¹⁴⁶ Pr	24.2 min	5.89 ± 0.91	3.24 ± 0.44	-	-	3.08 ± 0.17	3.27 ± 0.36	-	-

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(a) The cumulative yield of ¹⁴⁰Ba could not have been calculated considering its long half-life of almost 13 days (compared to the counting time of less than 3 days). We assume that the independent photofission yield of ¹⁴⁰Ba is negligible, as it is the case for neutron fission ($Y_{i,s}({}^{140}\text{Ba}) = 0.027\%$ according to JENDL database [32]). Since the half-life of ¹⁴⁰La is much shorter than that of its father nuclide ¹⁴⁰Ba (1.7 days vs. 13 days), the independent yield of ¹⁴⁰La can be calculated by analyzing the delayed gamma spectrum. We assume that the sum of the ¹⁴⁰Cs cumulative yield and the ¹⁴⁰La independent yield provide a relevant estimation of the ¹⁴⁰La cumulative yield.

Table 9: Detailed data of the delayed gamma-ray analysis for each ^{238}U photofission product

Fission Product	Cooling time (s)	Counting time (s)	Gamma-ray energy (keV)	Relative intensity (%)	Net peak area (counts)	Photofission cumulative yield (%)
^{84}Br	1185	11429	881.60	41.60	9965 ± 174	1.37
^{87}Kr	334	27474	402.59	49.60	42830 ± 403	1.99
			2554.80	9.23	3882 ± 70	1.87
			2558.10	3.92	1646 ± 52	1.87
			2011.88	2.88	1497 ± 78	2.01
^{88}Kr	30	61362	2392.11	34.60	24501 ± 164	2.18
			834.83	12.97	18760 ± 287	2.34
			196.30	25.98	17385 ± 582	2.34
			1529.77	10.93	11463 ± 156	2.27
			2029.84	4.53	3860 ± 98	2.46
			2035.41	3.74	3441 ± 96	2.46
^{88}Rb	30	67713	2231.77	3.39	2658 ± 78	2.26
			1836.02	22.40	23215 ± 173	2.68
^{89}Rb	1003	6626	1032.00	63.60	14361 ± 159	3.55
			1248.20	45.60	9090 ± 129	3.42
			657.80	11.00	2723 ± 185	3.42
^{90}Rb	30	912	831.68	39.90	1665 ± 103	1.62
			3383.4	6.66	109 ± 15	1.90
$^{90\text{m}}\text{Rb}$	213	1519	831.69	94.09	2228 ± 114	1.06
^{91}Sr	334	208128	1024.30	33.50	88784 ± 381	3.89
			749.80	23.68	67494 ± 485	3.77
			652.90	8.04	24203 ± 478	3.89
$^{91\text{m}}\text{Y}$	30	226302	555.57	95.00	179440 ± 688	2.35
^{92}Sr	30	61362	1383.90	93.00	185418 ± 450	4.05
					6065 ± 195	3.86
^{92}Y	30	134990	561.10	2.39	9197 ± 466	4.79
^{93}Sr	30	3829	1387.11	3.47	843 ± 87	4.44
^{93}Y	30	222534	266.90	7.42	18045 ± 765	4.70
^{94}Sr	30	425	1427.70	94.42	2913 ± 68	4.35
^{94}Y	30	7173	918.74	56.00	45081 ± 257	4.48
^{95}Y	30	3829	954.00	15.80	6711 ± 145	4.65
			1324.00	4.91	2215 ± 98	5.81
			2175.60	7.00	1926 ± 63	5.08
			2632.40	4.76	944 ± 40	4.32
			3576.00	6.38	1023 ± 34	4.89
^{97}Zr	577	242902	1149.97	2.62	9894 ± 240	5.95
			355.40	2.09	7902 ± 687	5.90
^{99}Mo	577	242902	777.92	4.28	8320 ± 428	4.65
$^{99\text{m}}\text{Tc}$	577	242902	140.51	89.00	39715 ± 850	4.36
^{101}Mo	30	5528	191.92	18.21	5925 ± 326	5.38
			1304.00	2.71	1646 ± 103	5.36
			1599.26	1.75	919 ± 80	5.38
^{101}Tc	30	10335	306.83	88.70	105166 ± 462	5.61
^{104}Tc	334	7963	358.00	89.00	41397 ± 341	3.62
			1612.40	5.79	1929 ± 85	3.22
			1596.70	4.18	1414 ± 82	3.25
^{105}Ru	2765	95852	316.44	11.12	15466 ± 510	2.77
			262.83	6.58	7300 ± 536	2.63
			413.53	2.27	4035 ± 441	2.98
^{105}Rh	577	242902	318.90	19.10	25015 ± 728	2.78
^{128}Sn	30	21214	482.30	59.00	14261 ± 336	0.57
			557.30	16.52	4119 ± 303	0.58
^{128}Sb	5562	237917	754.00	100.00	13089 ± 417	0.20
			314.10	61.00	6821 ± 685	0.22
$^{128\text{m}}\text{Sb}$	30	3829	753.90	96.40	14579 ± 200	1.61

			314.00	88.69	13222 ± 283	2.03
¹²⁹ Sb	30	94940	812.80	47.60	35168 ± 345	1.09
			683.50	5.66	4447 ± 358	1.11
¹³⁰ Sn	30	1336	780.50	56.43	875 ± 119	0.47
¹³⁰ Sb	3616	14224	793.40	100.00	8827 ± 176	0.74
			839.52	100.00	9622 ± 169	0.83
			182.33	65.00	2549 ± 306	0.82
			732.00	22.00	2201 ± 172	0.83
¹³¹ Sb	334	7963	943.40	46.20	19388 ± 190	2.38
			933.10	25.87	12628 ± 177	2.79
			1207.4	3.88	1463 ± 121	2.38
			854.6	3.23	1465 ± 158	2.50
^{131m} Te	30	243449	793.75	14.10	6057 ± 412	0.70
¹³² Te	2400	241079	228.33	88.12	85431 ± 849	4.71
¹³² I	30	243449	954.55	17.57	29802 ± 331	4.88
			630.19	13.32	24867 ± 523	4.86
			727.20	3.16	5752 ± 456	4.85
¹³³ Sb	30	1336	1096.22	43.00	2165 ± 96	2.59
^{133m} Te	30	28933	912.67	55.27	70144 ± 325	3.17
			647.51	19.40	30434 ± 341	3.63
			863.96	15.64	19427 ± 249	3.04
			914.77	10.94	13995 ± 222	3.20
			978.30	4.86	6903 ± 193	3.66
			1683.23	4.15	3837 ± 108	3.23
¹³⁴ Te	30	15015	565.99	18.60	36590 ± 333	5.38
			435.06	18.90	32642 ± 355	5.00
			210.46	22.40	22979 ± 428	5.84
			461.00	10.60	19492 ± 330	5.24
			464.64	5.03	9270 ± 313	5.24
¹³⁴ I	30	33918	847.03	95.70	386358 ± 659	7.28
			884.09	65.08	262124 ± 551	7.38
			1072.55	14.93	53751 ± 284	6.97
			595.36	11.10	47620 ± 380	7.24
			1136.16	9.09	33235 ± 246	7.40
			857.29	6.70	26628 ± 270	7.15
			947.86	4.01	16365 ± 221	7.77
			1741.49	2.57	7312 ± 120	7.62
¹³⁵ I	30	142104	1260.41	28.70	103434 ± 363	5.93
			1131.51	22.59	84064 ± 355	5.80
			1457.56	8.67	26695 ± 209	5.50
			1678.03	9.56	28593 ± 197	5.98
			1791.20	7.72	21157 ± 169	5.74
^{135m} Xe	30	147578	526.57	80.84	64938 ± 553	1.32
¹³⁸ Xe	30	5228	2015.82	12.25	3750 ± 84	3.75
¹³⁸ Cs	30	17081	1435.86	76.30	130311 ± 375	5.95
			1009.78	29.83	61012 ± 293	5.96
			2218.00	15.18	17644 ± 144	5.54
			871.8	5.11	10134 ± 210	5.32
¹⁴⁰ Cs	30	425	602.36	52.80	2335 ± 106	5.65
¹⁴⁰ La	577	242902	1596.20	95.40	68089 ± 285	5.72
			487.02	46.10	47606 ± 621	5.72
			815.78	23.72	25183 ± 403	5.72
			328.76	20.8	14948 ± 714	5.71
¹⁴¹ Ba	152	6443	190.33	46.00	14922 ± 347	4.69
			343.67	14.44	9776 ± 292	4.97
			457.80	5.01	3560 ± 249	4.45
			462.30	4.88	3471 ± 248	4.44
¹⁴² Ba	30	3829	255.30	20.50	6593 ± 291	5.00
			895.20	13.86	6072 ± 152	4.59
			1204.30	14.23	5082 ± 129	4.21

			1078.70	11.46	4190 ± 128	4.09
			949.10	10.60	4120 ± 134	4.13
			1001.20	9.72	4019 ± 134	4.52
			231.61	12.12	3410 ± 293	4.96
			425.04	5.72	2478 ± 232	4.52
			1202.40	5.54	1979 ± 114	4.21
¹⁴² La	30	36592	641.28	47.40	139288 ± 483	4.89
			1363.00	2.13	4832 ± 140	4.95
¹⁴³ Ce	5075	238404	293.27	42.80	82517 ± 760	4.41
			664.57	5.69	15052 ± 453	4.36
			721.93	5.39	14039 ± 431	4.36
¹⁴⁶ Pr	4893	8693	453.88	48.00	5991 ± 197	5.90
			1524.73	15.60	1435 ± 65	5.89
			735.72	7.49	951 ± 134	5.87

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646 It is worth noting that we provide cumulative yields for some ²³⁸U photofission products
647 that are not available in prior publications, namely ^{90m}Rb, ⁹⁰Rb, ^{99m}Tc, ^{128m}Sb, ¹³⁰Sn, ¹³³Sb,
648 ¹⁴⁰Cs and ¹⁴⁰La. Most photofission product yields obtained in this work are in good
649 agreement with existing data, but they are in average 25 % lower than those provided by
650 Carrel *et al.* [6], in spite of a similar experimental configuration in terms of LINAC
651 characteristics. The observed discrepancies in the published cumulative photofission yields
652 can be due to several factors. In our case, analyzing several gamma-ray lines for a same
653 photofission product, when possible, and observing a good consistency in the obtained
654 yields enhances the reliability on the weighted average reported in the above tables (for
655 example ⁸⁷Kr, ⁸⁸Kr, ¹³⁴I, ¹³⁵I, ¹³⁸Xe, ¹³⁸Cs, ¹⁴²La). In addition, correcting the fraction of
656 neutron fissions with respect to photofissions thanks to MCNP improves the estimation of
657 the absolute photofission yields. Other causes of discrepancy between published data are
658 certainly due to differences in experimental configurations, such as the geometry of the
659 samples, energy spectrum of the interrogating photon beam, photoneutron production,
660 measurement sequence (irradiation, cooling and counting times), etc.

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662 The ²³⁵U cumulative photofission yields measured with the HEU sample are given in Table
663 10. The HEU sample mass being approximately two orders of magnitude smaller than that
664 of the DU sample, counting statistics is not as good and fewer photofission products
665 cumulative yields could be determined for ²³⁵U.

Table 10: Cumulative yields (per 100 fissions) for the photofission of ^{235}U and comparison with published data

Fission product	$T_{1/2}$	Photon beam type	Bremsstrahlung	Bremsstrahlung	Monoenergetic
		Energy (MeV)	15.8	16.3	13.0
		This work	Carrel 2011 [6]	Krishichayan 2019 [11]	
^{87}Kr	1.3 h	4.64 ± 0.74	-	3.45 ± 0.28	
^{88}Kr	2.8 h	5.34 ± 0.84	3.63 ± 0.32	3.87 ± 0.22	
^{89}Rb	15.4 min	6.89 ± 1.08	4.69 ± 0.28	-	
^{91}Sr	9.7 h	7.71 ± 1.18	5.37 ± 0.26	6.08 ± 0.26	
$^{91\text{m}}\text{Y}$	49.7 min	4.80 ± 1.72	5.38 ± 0.25	-	
^{92}Sr	2.6 h	7.92 ± 1.24	5.59 ± 0.26	6.52 ± 0.21	
^{92}Y	3.5 h	9.01 ± 2.44	-	-	
^{93}Sr	7.4 min	8.02 ± 1.53	5.81 ± 0.27	-	
^{93}Y	10.2 h	8.19 ± 2.53	-	6.14 ± 0.93	
^{104}Tc	18.3 min	1.52 ± 0.24	-	2.37 ± 0.14	
^{105}Ru	4.4 h	1.86 ± 0.29	-	1.90 ± 0.11	
^{128}Sn	59.1 min	1.38 ± 0.22	-	1.22 ± 0.16	
^{128}Sb	9 h	0.90 ± 0.14	0.39 ± 0.02	-	
^{129}Sb	4.4 h	2.46 ± 0.36	1.60 ± 0.08	2.47 ± 0.12	
^{130}Sb	39.5 min	1.49 ± 0.25	1.12 ± 0.05	0.82 ± 0.06	
^{131}Sb	23 min	3.42 ± 0.53	2.75 ± 0.13	1.59 ± 0.13	
$^{131\text{m}}\text{Te}$	1.3 d	1.98 ± 0.33	-	-	
^{134}Te	41.8 min	5.33 ± 0.84	3.16 ± 0.15	5.37 ± 0.39	
^{134}I	52.5 min	7.52 ± 2.22	-	-	
^{135}I	6.6 h	7.34 ± 1.14	5.06 ± 0.32	4.72 ± 0.21	
^{138}Xe	14.1 min	7.36 ± 1.18	4.62 ± 0.41	-	
^{138}Cs	33.4 min	7.79 ± 2.61	-	7.74 ± 0.33	
^{141}Ba	18.3 min	6.92 ± 1.06	-	4.43 ± 0.27	
^{142}Ba	10.6 min	5.98 ± 1.06	4.41 ± 0.21	-	
^{142}La	1.5 h	6.39 ± 2.18	5.15 ± 0.24	5.98 ± 0.19	
^{143}Ce	1.4 d	6.41 ± 0.99	-	4.78 ± 0.30	

Table 11: Detailed data of the delayed gamma-ray analysis for each ^{235}U photofission product

Fission Product	Cooling time (s)	Counting time (s)	Gamma-ray energy (keV)	Relative intensity (%)	Net peak area (counts)	Photofission cumulative yield (%)
^{87}Kr	392	27479	402.59	49.60	10621 ± 147	4.66
			845.44	7.34	731 ± 69	4.15
			2554.80	9.23	370 ± 25	4.45
^{88}Kr	30	61289	2392.11	34.60	2535 ± 55	5.54
			834.83	12.97	2234 ± 88	5.16
			165.98	3.10	1575 ± 246	5.15
			1529.77	10.93	1124 ± 53	4.47
			2035.41	3.74	298 ± 36	4.53
^{89}Rb	934	6689	1248.20	45.60	880 ± 43	6.89
^{91}Sr	392	79187	1024.30	33.50	6820 ± 101	7.62
			749.80	23.68	6493 ± 121	7.92
			652.90	8.04	2296 ± 120	7.34
			652.30	2.98	851 ± 113	7.34
$^{91\text{m}}\text{Y}$	30	79549	555.57	95.00	20125 ± 181	4.80
^{92}Sr	30	61289	1383.90	93.00	15870 ± 134	7.92
			430.48	3.35	1506 ± 128	7.65
^{92}Y	30	79549	934.50	13.90	4358 ± 92	9.01
^{93}Sr	30	3797	168.50	18.43	1614 ± 118	8.08
			888.13	22.10	610 ± 50	7.95
^{93}Y	30	79549	266.90	7.42	4946 ± 188	8.19
^{104}Tc	392	7954	358.00	89.00	2219 ± 98	1.52
^{105}Ru	2742	79837	724.30	47.30	3206 ± 102	1.83
			469.37	17.55	1742 ± 115	1.83
			316.44	11.12	1866 ± 152	2.20
			676.36	15.66	1042 ± 105	1.69
^{128}Sn	30	21152	482.30	59.00	2967 ± 111	1.38
			557.30	16.52	705 ± 95	1.34
^{128}Sb	5635	73944	314.10	61.00	3968 ± 150	0.99
			743.30	100.00	2788 ± 91	0.91
			754.00	100.00	2540 ± 88	0.84
			526.50	45.00	1627 ± 101	0.87
^{129}Sb	30	79549	812.80	47.60	4025 ± 101	2.31
			544.70	18.09	2746 ± 126	2.95
			1030.00	13.33	960 ± 70	2.40
^{130}Sb	3646	14282	182.33	65.00	1963 ± 123	1.66
			330.91	78.00	1713 ± 94	1.60
			793.40	100.00	777 ± 54	1.22
			839.52	100.00	864 ± 52	1.42
^{131}Sb	392	7954	943.40	46.20	1404 ± 58	3.57
			933.10	25.87	632 ± 52	2.82
$^{131\text{m}}\text{Te}$	30	79549	773.67	39.51	1416 ± 95	2.08
			852.21	21.40	572 ± 79	1.45
^{134}Te	30	15005	277.95	21.30	5444 ± 144	5.31
			767.20	29.60	3249 ± 89	5.51
			201.24	8.90	2502 ± 150	4.97
			565.99	18.60	2509 ± 98	5.18
^{134}I	30	33988	884.09	65.08	13413 ± 133	7.45
			595.36	11.10	3487 ± 112	8.38
			405.45	7.37	3180 ± 128	8.09
			857.29	6.70	1526 ± 78	8.28
^{135}I	30	79549	1260.41	28.70	5217 ± 89	7.23
			1131.51	22.59	4657 ± 91	7.49
			288.45	3.10	2153 ± 176	7.81
			836.80	6.69	1704 ± 86	7.21

			1457.56	8.67	1496 ± 58	7.84
			1678.03	9.56	1316 ± 53	7.05
			1791.20	7.72	997 ± 47	6.95
¹³⁸ Xe	30	5243	258.41	31.50	4479 ± 121	7.48
			1768.26	16.73	390 ± 33	6.27
			2015.82	12.25	293 ± 28	7.32
¹³⁸ Cs	30	16994	1435.86	76.30	7856 ± 96	7.79
			2218.00	15.18	1060 ± 40	7.78
			2639.59	7.63	448 ± 26	7.85
¹⁴¹ Ba	211	6327	304.19	25.44	3441 ± 109	6.92
¹⁴² Ba	30	3797	985.20	13.86	411 ± 49	5.98
¹⁴² La	30	36519	894.90	8.34	1499 ± 76	6.40
			2397.80	13.27	998 ± 38	6.38
¹⁴³ Ce	5092	74487	293.27	42.80	9883 ± 170	6.41

669

670 Despite the abovementioned limitations in terms of counting statistics, we provide here
671 cumulative yields for some ²³⁵U photofission products that are not reported in prior
672 publications, namely ⁹²Y, ^{131m}Te and ¹³⁴I. For the others, the cumulative yields obtained in
673 this work are quite consistent with existing data, but contrary to ²³⁸U for which our yields
674 were a bit smaller than in previous references, they are now a bit larger. For instance, we
675 observe an average difference of 28 % with Carrel *et al.* [6], despite a similar LINAC
676 configuration as in our experiments as already reported. The origin of possible biases in
677 our experiments with DU and HEU samples will be further investigated by using another
678 HEU sample with a larger mass, which will also allow measuring more ²³⁵U yields.

679

680 4.3. ²³⁵U / ²³⁸U discrimination from gamma-ray lines ratios

681 When photofission occurs, the formation of two asymmetric fission fragments is the
682 most likely to happen, resulting in a mass distribution curve of the fission products
683 comporting two bumps: one for a heavy nucleus centered around mass number 140, and
684 one for a light nucleus centered around 95. Examples of this theoretical mass distribution
685 for photofission products are given in ref. [34] for several actinides. A discrimination
686 information can be obtained between uranium isotopes thanks to their different
687 photofission product yields, and thus the intensity difference in their delayed gamma
688 spectra [1][14]. The ²³⁵U enrichment can thus be computed from photofission **delayed**
689 **gamma ratios of the net areas** and from the respective ²³⁵U and ²³⁸U photofission yields
690 ratios of specific photofission product pairs. The efficiency of this discrimination method
691 can be assessed by defining a discrimination criterion based on the difference of
692 photofission yields ratios from any photofission product to any other. By maximizing this

693 discrimination criterion $\delta = \left[\frac{Y_{5,PF1}}{Y_{8,PF1}} - \frac{Y_{5,PF2}}{Y_{8,PF2}} \right]$, the most appropriate photofission product
 694 couples can be identified based on the cumulative photofission product yields we reported
 695 in previous section. These pairs of photofission products with the largest values of δ are
 696 given in Table 12.

697 *Table 12: Photofission products offering the best capability for ^{235}U vs. ^{238}U discrimination (see the text for the*
 698 *definition of δ)*

Fission product couple	δ
$^{128}\text{Sb}/^{104}\text{Tc}$	3.86
$^{128}\text{Sb}/^{105}\text{Ru}$	3.61
$^{128}\text{Sb}/^{134}\text{Te}$	3.28
$^{128}\text{Sb}/^{134}\text{I}$	3.26
$^{128}\text{Sb}/^{135}\text{I}$	3.03
$^{128}\text{Sb}/^{142}\text{La}$	2.98
$^{128}\text{Sb}/^{138}\text{Cs}$	2.97
$^{128}\text{Sb}/^{131}\text{Sb}$	2.92
$^{128}\text{Sb}/^{142}\text{Ba}$	2.92
$^{128}\text{Sb}/^{141}\text{Ba}$	2.83

699
 700 It appears that ^{128}Sb is part of all fission product couples because of its particularly high
 701 ratio between its cumulative yields for ^{235}U and ^{238}U . However, the value of its cumulative
 702 photofission yield measured in this work for ^{235}U is more than two times higher than the
 703 only value previously published by Carrel *et al.* [6]. Improved counting statistics for ^{235}U
 704 will enable to verify this particular yield and eventually to update the most discriminating
 705 pairs of photofission products in future work. Besides, it should be noted that the fission
 706 product couples listed above are not equally applicable to practical measurements. Indeed,
 707 the energy dependence of detection efficiency and gamma-ray attenuation, due to the waste
 708 matrix, for instance, as well as gamma-ray interferences and the experimental sequence
 709 (irradiation, cooling, counting times), have to be taken into account to select the relevant
 710 fission product couples for a given application. This selection will be refined in future work
 711 aiming at developing a practical method to characterize nuclear materials in large
 712 radioactive waste packages using photofission.

713 5. Conclusion

714 New measurements of cumulative photofission yields with a 15.8 MeV endpoint
715 Bremsstrahlung photon beam produced by a SATURNE electron LINAC have been
716 performed in CINPHONIE facility, at CEA Cadarache, France. To this aim, a
717 characterization of the Bremsstrahlung photon beam has been first carried out by photon
718 activation analysis with different samples of Au, Ni, U and Zr. Then, the residual neutron
719 flux exiting the LINAC head (lead collimator, borated polyethylene and cadmium shield)
720 has been characterized by neutron activation analysis with Au, In, Cu, Ni and Mg samples
721 (mostly In activation gamma rays), so as to estimate the contribution of photoneutron
722 fissions in the DU and HEU samples used to assess the photofission yields. The impact of
723 photoneutrons produced in the LINAC and its surroundings (tungsten target, lead
724 collimator, cadmium sheet) is limited for this type of experiment, most of neutron fissions
725 arising from fast neutrons produced in the sample itself. Finally, the cumulative production
726 yields of 49 and 26 photofission products have been measured for ^{238}U and ^{235}U ,
727 respectively, from which 8 and 3 yields were not reported so far in the literature. Some
728 photofission products, showing large discrepancies between both uranium isotopes appear
729 as good candidates for an isotope discrimination method based on gamma-ray lines ratios
730 measurement.

731 The preparation of new experimental campaigns is underway, firstly to assess photofission
732 products cumulative yields for other actinides of interest, like plutonium, and secondly to
733 test the differentiation of actinides isotopes with a strong attenuation of photofission
734 delayed gamma rays by a dense waste matrix, like concrete.

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Highlights

- Photofission is a promising technique to assay nuclear waste packages
- Photon activation analysis is used to characterize a 16 MeV Bremsstrahlung beam
- New photofission products cumulative yields have been determined for ^{235}U and ^{238}U
- Special attention has been drawn to distinguish neutron fission from photofission
- Photofission product yield difference makes actinides differentiation practicable

CRediT author statement:

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