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# New measurements of cumulative photofission yields of $^{239}\text{Pu}$ , $^{235}\text{U}$ and $^{238}\text{U}$ with a 17.5 MeV Bremsstrahlung photon beam and progress toward actinide differentiation

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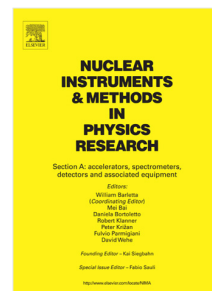
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New measurements of cumulative photofission yields of  $^{239}\text{Pu}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$  with a 17.5 MeV Bremsstrahlung photon beam and progress toward actinide differentiation

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24 zirconium. The contribution of neutron fission in the different samples has also been  
25 estimated by MCNP simulations in order to assess as precisely as possible the photofission  
26 yields. Finally, 26 cumulative photofission product yields are reported for  $^{239}\text{Pu}$ , 28 for  
27  $^{238}\text{U}$  and 26 for  $^{235}\text{U}$ , with half-lives ranging from 14 min to more than 3 days, some of  
28 them being not recorded so far in the literature. Among these reported photofission product  
29 yields, 18 have been measured for all 3 actinides, which can thus be used for their  
30 discrimination. A differentiation criterion based on delayed gamma-ray ratios has been  
31 established to determine the most efficient photofission product couples to estimate the  
32 enrichment of a  $^{235}\text{U}/^{238}\text{U}$  mixture or the fissile fraction ( $^{235}\text{U}+^{239}\text{Pu}$ )/actinide mass in a  
33 mixture of uranium and plutonium.

## 34 1. Introduction

35 The safety related to the management of radioactive waste (transportation, interim storages  
36 and final repositories) is ensured with an accurate non-destructive characterization of their  
37 actinide content in relation with the corresponding specifications. Among active non-  
38 destructive methods that have been studied to address this characterization in the case of  
39 large and dense packages, such as concrete drums [1]-[5], Active Photon Interrogation  
40 based on the photofission phenomenon, is the only one that can bring a sufficient signal  
41 from the nuclear materials inside the package. Specifically, the detection of delayed gamma  
42 radiation emitted by fission products induced by high-energy photons has the potential to  
43 assess the actinide mass present in a package, and possibly to distinguish fissile nuclei (that  
44 can undergo thermal neutron fission, e.g.  $^{235}\text{U}$  and  $^{239}\text{Pu}$ ) and fertile nuclei (that can absorb  
45 a neutron, leading to the formation of a fissile nuclei, e.g.  $^{238}\text{U}$ ). To that extent, photofission  
46 yields of the actinides of interest must be known precisely. Even though nuclear data  
47 related to photofission yields of  $^{238}\text{U}$  exist, they sometimes present significant  
48 discrepancies, even in recent studies [6]-[12]. Photofission yield data are even scarcer for  
49 fissile isotopes such as  $^{235}\text{U}$  [7][12] and  $^{239}\text{Pu}$  [11]-[14], hence the need to perform new  
50 measurements.

51 The potential of analyzing the delayed gamma ray signal following fission to obtain an  
52 actinide identification information has already been demonstrated in the past. Hollas *et al.*

53 [15] and Beddingfield *et al.* [16] have reported the use of delayed gamma-ray ratios for  
54 actinide differentiation, respectively for photofission and thermal neutron fission. Further  
55 experimental work conducted by Gmar *et al.* [17] pointed out variations of the delayed  
56 gamma-ray emissions for uranium samples of different enrichments. Also, Carrel *et al.* [1]  
57 brought information about the delayed gamma emission following photofission in mixed  
58 samples of  $^{235}\text{U}$  and  $^{238}\text{U}$ . Besides, the uranium isotopes differentiation in an 870 L waste  
59 drum by using delayed gamma-ray ratios has already been investigated experimentally with  
60 a mockup package [1] and by using Monte-Carlo simulations in the work of Simon *et al.*  
61 [5]. Furthermore, photofission products emitting several gamma rays can be used as  
62 attenuation indicators to estimate the depth at which nuclear materials are localized inside  
63 the package [18].

64 In the frame of a long-term research program conducted by the Nuclear Measurement  
65 Laboratory of CEA IRESNE Institute in France, this work follows the study recently  
66 reported in [19], which provided cumulative photofission yields of  $^{235}\text{U}$  and  $^{238}\text{U}$  with a  
67 15.8 MeV Bremsstrahlung beam produced by a linear electron accelerator (LINAC) in  
68 CINPHONIE casemate of CHICADE nuclear facility [20]. We present here new  
69 cumulative photofission yields for  $^{239}\text{Pu}$ , and again for  $^{235}\text{U}$  and  $^{238}\text{U}$ , which are measured  
70 with the same setup but with an endpoint electron energy of 17.5 MeV. To this aim, the  
71 characterization of the photon beam is first carried out by photon activation analysis with  
72 different samples of Au, Ni, U, Zn and Zr. Then we estimate the neutron fission rates in  
73 the different samples with MCNP, in view to subtract it from the total fission rate and thus  
74 obtain the photofission yields. We also identify photofission products of interest for the  
75 differentiation between fissile and fertile actinides.

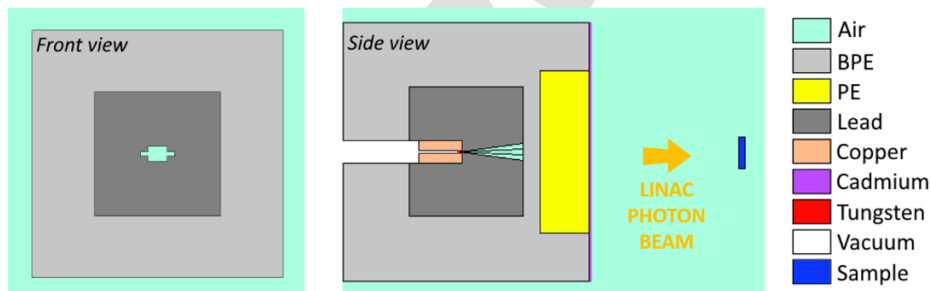
## 76 2. Experimental setup

77 Experiments were performed by using a Bremsstrahlung photon beam produced by a  
78 Saturne LINAC located in the CINPHONIE irradiation cell at CEA Cadarache. In pulse  
79 mode, the LINAC accelerates electrons up to 21 MeV. The electrons strike a 5 mm thick  
80 tungsten target and a part of their kinetic energy is converted into Bremsstrahlung radiation.  
81 The pulse frequency and width are 200 Hz and 4.1  $\mu\text{s}$ , respectively, and the peak current is  
82 100 mA at the target entrance. A 20 cm thick lead collimator allows focusing the beam on

83 the actinide samples, surrounded by a 20 cm thick shield made of borated polyethylene  
 84 (BPE) and polyethylene to limit the photoneutron flux reaching the samples, and thus to  
 85 minimize neutron fissions. A cadmium (Cd) layer of 2 mm was added on the front face to  
 86 complete the thermal neutron absorption occurring in BPE. A picture of the LINAC  
 87 configuration and the corresponding simulated model with a materials description are  
 88 respectively given in Figure 1 and Figure 2. During the experiments, the photon dose rate  
 89 at 1 m from the tungsten target measured by an ionization chamber was 33 Gy/min.



90  
91 *Figure 1: Saturne LINAC with collimator and neutron shielding.*



92  
93 *Figure 2: MCNP model of the irradiation configuration (LINAC, lead collimator, neutron shielding and sample).*

94 The photon flux at the output of the collimator aperture being not known precisely,  
 95 nor the endpoint-energy of the Bremsstrahlung spectrum expected around 16 MeV, a  
 96 characterization of the photon beam produced by the LINAC is performed using photon  
 97 activation of reference materials, following the method described in our previous work  
 98 [19]. The main steps and results of the beam characterization are described in section 3.  
 99 The pellets irradiated to this aim and positioned in the axis of the LINAC photon beam are  
 100 described in Table 1. The other pellets in Figure 3 are made of indium (left) and magnesium  
 101 (right). They were originally designed to be used as neutron activation spectrometers [21]  
 102 and were irradiated to estimate the photoneutron production in the CINPHONIE irradiation  
 103 cell (not reported in this paper but used qualitatively in section 4).

104

Element	Mass (g)	Diameter (mm)	Thickness (mm)	Position on Figure 3
Au	0.045	5	0.05	1
Zn	2.70	19	1.33	2
Ni	10.03	22	3	3

Table 1: Characteristics of the metal pellets irradiated for the photon beam characterization.

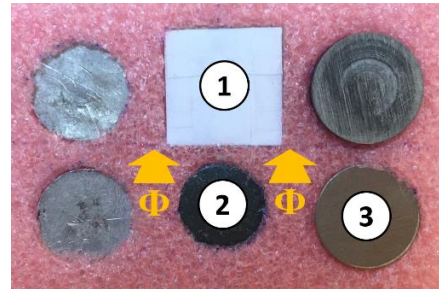


Figure 3: Position of the pellets on their support in view of their common irradiation.

107 Three actinide samples were irradiated: a sample of Depleted Uranium (DU) which is  
 108 the same as we used in our previous work [19], a sample of Highly Enriched Uranium  
 109 (HEU) and a sample of plutonium (Pu). The different samples are described in Table 2.

110

Table 2: Description of the actinide samples.

	Uranium		Plutonium	
	DU	HEU	Sample	Pu
Mass	> 100 g and < 1 kg	> 1 g and < 100 g	Mass	~1 g
<sup>235</sup> U content	0.3 %	> 90 %	Isotopic composition	<sup>239</sup> Pu: 83 % <sup>240</sup> Pu: 12 %
Composition	Metallic uranium	Metallic uranium core held between Zircaloy sheets	Chemical form	PuO <sub>2</sub> powder mixed with resin
Dimension	1 cm thickness	< 1 mm thickness	Density	1.98 g.cm <sup>-3</sup>
Density	18.96 g.cm <sup>-3</sup>	Fissile core: 18.96 g.cm <sup>-3</sup> Zircaloy: 6.56 g.cm <sup>-3</sup>		

111

112 Since the DU sample is 1 cm thick and composed of metallic uranium of density  
 113 18.96 g.cm<sup>-3</sup>, significant self-attenuation effects occur both for the interrogating photon  
 114 flux and the delayed gamma rays emitted by photofission products. Therefore, correction  
 115 factors are applied in order to calculate the photofission product yields (see section 4.2).  
 116 The plutonium sample is composed of 83 % of <sup>239</sup>Pu and 12 % of <sup>240</sup>Pu. In this work, we  
 117 will consider that the sample is made of 95 % of <sup>239</sup>Pu since the photofission cross sections  
 118 for these two isotopes are similar, as seen in Figure 4. It can also be noted that the calculated  
 119 photofission rates are the same with the real isotopic composition of the plutonium sample  
 120 and with 100 % of <sup>239</sup>Pu. Based on the work of Bernard *et al.* [22] with the GEF code, we  
 121 can also assume that the cumulative photofission yields of <sup>239</sup>Pu and <sup>240</sup>Pu are very close,  
 122 enabling us to consider our plutonium sample as a <sup>239</sup>Pu sample without introducing a  
 123 significant bias in our analyses.

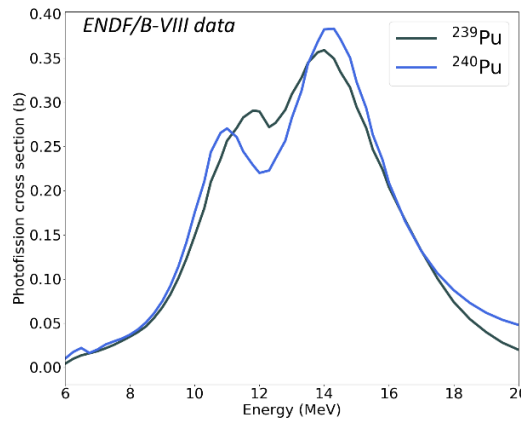


Figure 4: Photofission cross sections of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  [23].

124

125

126 For each actinide sample, a 2 h irradiation with the LINAC is followed by an automatic  
 127 transfer from the irradiation to the counting position, lasting less than a minute and noted  
 128 “cooling time” in further activation analysis equations. The samples used to characterize  
 129 the Bremsstrahlung photon beam were irradiated all together, on the support shown in  
 130 Figure 3, and transferred to a low-background spectrometer located in another  
 131 experimental room. Table 3 summarizes the distance and time parameters related to each  
 132 sample.

133

Table 3: Experimental distances and timings.

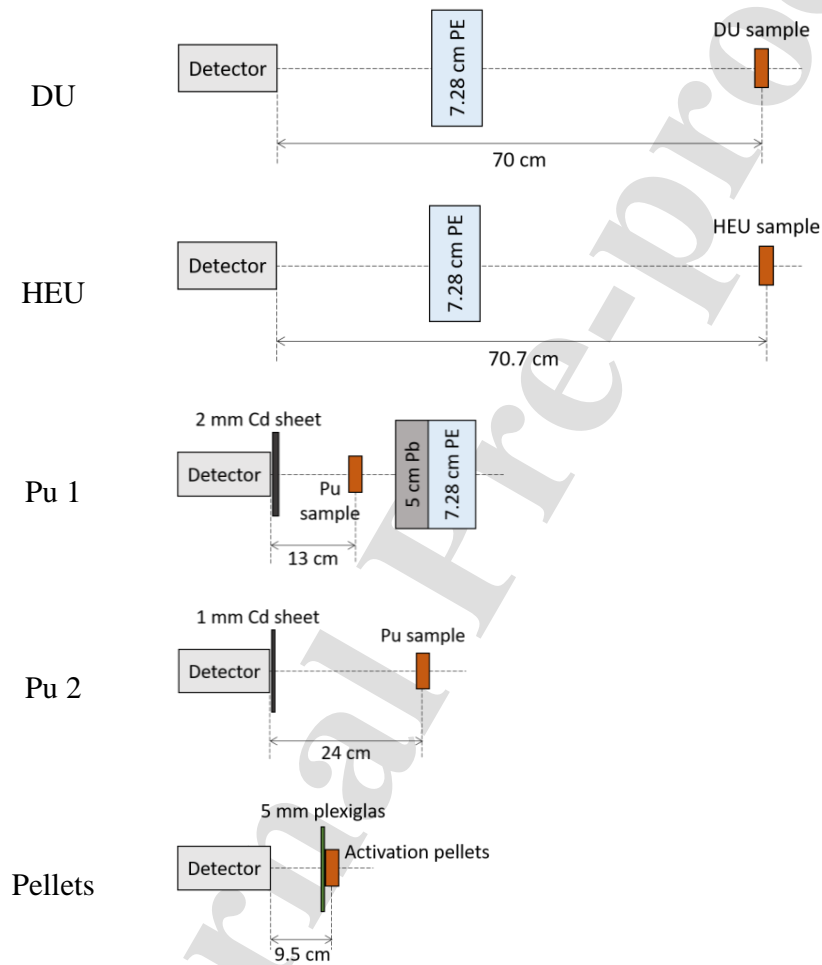
Sample	Denomination	Target-sample distance (cm)	Sample-detector distance (cm)	Irradiation time	Cooling time*	Counting time
Depleted Uranium	DU	102.0	70.0	2 h	19 min	42 h
Highly Enriched Uranium	HEU	102.7	70.7	2 h	42 s	24 h
Plutonium	Pu 1	45.4	13.0	2 h	50 min	24 h
	Pu 2	113.8	24.0	2 h	10 min	21 h
Activation samples (Au, Ni, Zn)	Pellets	88.0	9.5	1 h	390 s	145 h

134 \*See text for the reason of the different cooling times

135 Figure 5 shows the layout for every detection configuration. Different screens were added  
 136 depending on the irradiated samples. For the DU and HEU measurements, only a  
 137 polyethylene screen was inserted in front of the detector to protect the crystal from fast  
 138 photoneutron damage during irradiation (it also allows reducing the count rate). A thin  
 139 cadmium sheet was added in front of the detector to cut the passive emission component  
 140 of the plutonium sample (59.5 keV due to  $^{241}\text{Am}$ ). For the “Pu 1” measurement, a lead



141 shield and a polyethylene screen were used to diminish neutron activation of the detector  
 142 during irradiation and to reduce the dead-time related to the activation of the surroundings  
 143 during the measurement (the plutonium sample is inserted between the detector and these  
 144 shields by a mechanical device). Concerning the metal pellets measurement for the photon  
 145 beam characterization, the 5 mm Plexiglas screen corresponds to the sample holder.



146 *Figure 5: Experimental configuration layout for detection.*

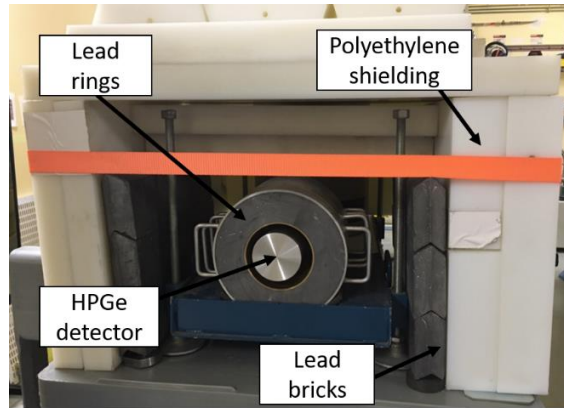
147 The gamma rays of fission and activation products are measured with a 50 % relative  
 148 efficiency n-type coaxial high-purity germanium detector (HPGe, ORTEC GMX50-83-1-  
 149 PL) equipped with a transistor-reset preamplifier and coupled to a LYNX Digital Signal  
 150 Analyzer (CANBERRA) driven by Genie2000 software (MIRION Technologies). The rise  
 151 time and flat top parameters are respectively set at 2  $\mu$ s and 0.5  $\mu$ s following an  
 152 optimization. The energy resolution is 2.0 keV (FWHM) at the 1332.5 keV gamma line of

153  $^{60}\text{Co}$ . Although an n-type HPGe crystal is used to limit neutron damage, the detector is  
154 shielded by lead and polyethylene, as shown in Figure 6. The analysis of actinide delayed  
155 gamma spectra is performed with the MAGIX software developed by CEA LIST, in  
156 collaboration with CEA IRESNE Nuclear Measurement Laboratory, to analyze complex  
157 gamma- and X-ray spectra measured with HPGe detectors. This automatic software, based  
158 on CEA LIST know-how in complex spectrum processing [24][25], performs a complete  
159 analysis including energy calibration, identification of radionuclides, peak deconvolution,  
160 determination of a relative detection efficiency as a function of energy, activity calculation  
161 for each radionuclide if the absolute efficiency is provided by the end-user, and otherwise  
162 activity ratios using the relative efficiency. One of its main features is to include iterative  
163 steps to identify the radionuclides likely to be associated with each peak of the spectrum,  
164 based on the gamma- and X-rays given in JEFF-3.3 database [26] and on a list of possible  
165 radionuclides provided by the user.

166 In order to monitor and correct for the varying dead time during the counting period  
167 (due to the rapidly decreasing total count rate), the delayed photofission gamma spectra of  
168 the actinide samples were acquired sequentially every 60 s with a spectrum reset. The dead  
169 time compensation is a live-time correction, which was assessed to be reliable for dead  
170 times below 50 % with the two-source method ( $^{88}\text{Y} + ^{137}\text{Cs}$  as the reference, and  $^{152}\text{Eu}$  as  
171 the perturbing source responsible of an increasing count rate), prior to LINAC acquisitions.  
172 The initial dead times after irradiation were 80 % and 39 %, respectively, for DU and HEU  
173 uranium samples. Therefore, the DU sample spectrum was analyzed only after a 19 min  
174 cooling time, hence the impossibility to measure short half-life photofission products. As  
175 the first measurement of the plutonium sample (Pu 1, cf. Table 3) had an initial dead time  
176 of 94 % (dead time fell below 50 % only after 50 min), another irradiation further from the  
177 LINAC tungsten target was performed (Pu 2, cf. Table 3), leading to an initial dead time  
178 of 61 % that fell below 50 % after 10 min of cooling.

179 The **non-actinide** activated metallic samples used for the beam characterization were  
180 transferred inside a low-background spectrometer with a 9 % relative efficiency HPGe  
181 detector (CANBERRA BEGe 2020) in a lead shield, with inner walls covered by a copper  
182 layer to cut lead X-rays. The detector is connected to a digital spectrometer (CANBERRA  
183 DSP9660) and the resolution is 1.75 keV (FWHM) at the 1332.5 keV gamma line of  $^{60}\text{Co}$ .

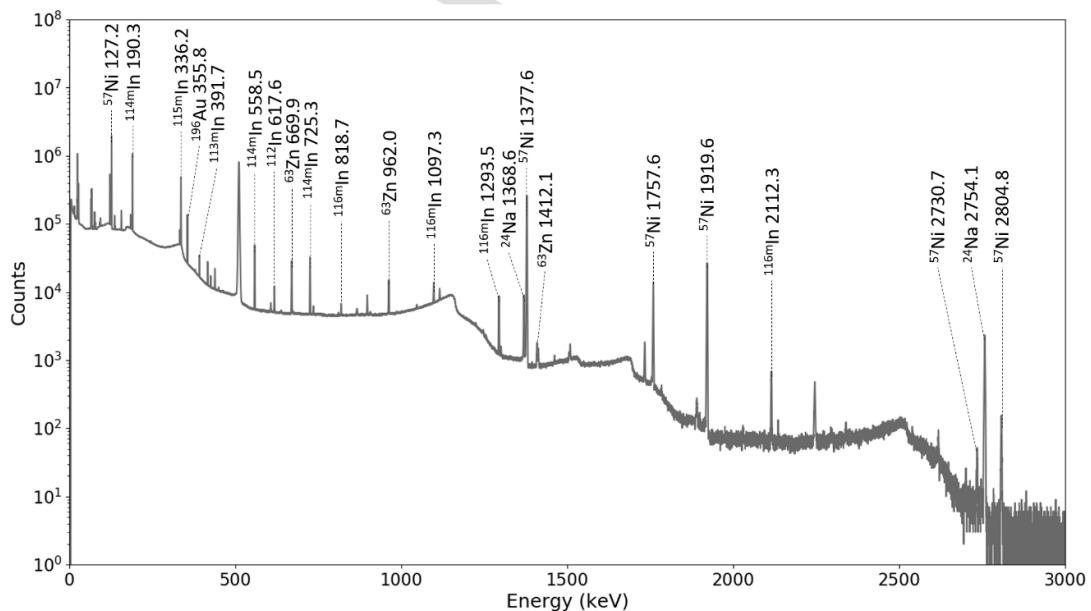
184 The gamma spectrum from the activation pellets was recorded during 6 days after  
 185 irradiation. Regular resets of the spectrum acquisition were also undertaken to properly  
 186 correct for dead time. These gamma spectra were analyzed with Genie2000 software  
 187 (MIRION Technologies).



188  
 189 *Figure 6: Shielded germanium detector.*

### 190 3. LINAC photon beam characterization

191 The photon and neutron activation spectrum of the thin metallic pellets irradiated to  
 192 characterize the photon beam of the LINAC is given in Figure 7.



193  
 194 *Figure 7: Activation gamma spectrum of the metallic pellets shown in Figure 3.*

195 The detected gamma rays due to ( $\gamma,n$ ) activation reactions of gold, zinc and nickel isotopes  
 196 are reported in Table 4, as well as those of  $^{238}\text{U}$  activation in the DU sample and  $^{90}\text{Zr}$   
 197 activation in the HEU sample (Zircaloy frame, see Table 2). Half-lives and gamma-ray  
 198 energies are taken from JEFF-3.3 nuclear database [26]. The net areas of the gamma-ray  
 199 peaks are from the spectra analysis with Genie2000 software. The net area statistical  
 200 uncertainty is  $\sigma(N_{counts}) = \sqrt{N_{counts} + 2B}$ , where  $B$  is the Compton background under  
 201 the total absorption peak.

202 *Table 4: Activation gamma rays due to the ( $\gamma,n$ ) reaction analyzed to characterize the LINAC photon beam.*

Activated isotope	Activation product	Half-life	$\gamma$ -ray line analyzed (keV)	Net area $N_{counts}$
$^{197}\text{Au}$	$^{196}\text{Au}$	6.17 days	355.8	$549721 \pm 741$
$^{58}\text{Ni}$	$^{57}\text{Ni}$	35.9 h	1377.6	$2411040 \pm 1553$
$^{238}\text{U}$	$^{237}\text{U}$	6.75 days	208.0	$1310540 \pm 1145$
$^{64}\text{Zn}$	$^{63}\text{Zn}$	38.3 min	669.9	$157242 \pm 397$
$^{90}\text{Zr}$	$^{89m}\text{Zr}$	250.8 s	587.8	$37223 \pm 193$
	$^{89}\text{Zr}$	3.26 days	/	/
	$^{89m}\text{Y}^*$	15.7 s	909.0	$22248 \pm 472$

203 *\* From the  $^{89}\text{Zr}$  and  $^{89m}\text{Zr}$  decays*

204 Since the characteristics of the interrogating photon beam were not known precisely,  
 205 photon activation of these materials is used to estimate the endpoint-energy of the  
 206 bremsstrahlung beam and the photon flux, as described in our previous work [19]. The  
 207 method is based on the differences in the photonuclear cross-sections [27], since each  
 208 material has a different energy threshold and cross-section for the ( $\gamma,n$ ) reaction. Therefore,  
 209 we are looking for the incident photon flux characteristics that best matches the observed  
 210 activation of five materials. To this aim, we assume a semi-Gaussian shape of the electron  
 211 energy distribution, see further Figure 8, of which we are looking for the optimal endpoint-  
 212 energy and width at half-maximum.

213 The endpoint-energy was varied from 15 MeV to 18.5 MeV with 0.5 MeV steps, and the  
 214 width at half-maximum from 0 MeV (mono-energy distribution) to 2 MeV with 0.5 MeV  
 215 steps. For each of the 40 pairs of parameters, MCNP [28] simulations were performed by  
 216 impinging electrons of the considered energy distribution on the LINAC tungsten target to  
 217 produce the Bremsstrahlung photon beam. Then, the number of ( $\gamma,n$ ) reactions in each  
 218 activated sample was numerically evaluated, corresponding to the convolution of the

219 resulting photon flux on the different materials with their reaction cross-sections. Finally,  
 220 an experimental photon flux is calculated for each activated isotope ( $^{197}\text{Au}$ ,  $^{58}\text{Ni}$ ,  $^{238}\text{U}$ ,  $^{64}\text{Zn}$   
 221 and  $^{90}\text{Zr}$ ) by using the net area of the peaks listed in Table 4. As a result, five photon fluxes  
 222 are obtained for each couple of beam parameters. The most probable electron energy  
 223 distribution is then identified as the one minimizing the squared differences between these  
 224 five flux values. In our case, the electron energy distribution with an endpoint-energy of  
 225 17.5 MeV and a 0.5 MeV width at half-maximum provides the most consistent photon  
 226 fluxes for all the materials, as reported in Table 5.

227 *Table 5: Experimental photon fluxes calculated for the five materials with the most probable electron energy*  
 228 *distribution.*

Activated isotope	$\Phi_{88\text{cm},\text{exp}}$ (photons.cm <sup>-2</sup> .s <sup>-1</sup> )
$^{197}\text{Au}$	$(6.82 \pm 0.87) \times 10^{10}$
$^{58}\text{Ni}$	$(7.31 \pm 0.89) \times 10^{10}$
$^{238}\text{U}$	$(6.07 \pm 0.75) \times 10^{10}$
$^{64}\text{Zn}$	$(6.57 \pm 0.84) \times 10^{10}$
$^{90}\text{Zr}$	$(5.97 \pm 0.75) \times 10^{10}$
Mean photon flux $\bar{\phi} = (6.55 \pm 0.94) \times 10^{10}$ photons.cm <sup>-2</sup> .s <sup>-1</sup>	

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

- 231 - a relative uncertainty estimated to 10 % on the ( $\gamma,n$ ) cross-section of the activated  
 232 isotopes, according to the EXFOR cross-section library [29]. As an example, the  
 233  $^{197}\text{Au}(\gamma,n)^{196}\text{Au}$  reaction cross-section uncertainty is about 10 % in the work of  
 234 Plaisir *et al.* [30];
- 235 - a relative uncertainty of 7 % on the detection efficiency to take into account both  
 236 the detector intrinsic efficiency (less than 5 % thanks to a fine detector model,  
 237 optimized using reference measurements of standard sources) and the modeling of  
 238 the experimental set-up (uncertainties on samples and on equipment dimensions,  
 239 set at 5 % based on our experience of such simulations). Intrinsic and geometric  
 240 efficiency uncertainties are combined in quadratic sum, leading to a 7 % relative  
 241 uncertainty on detection efficiency;

- 242 - the statistical uncertainty related to MCNP simulation results, which is lower than  
 243 2 % for all activation calculations;
- 244 - the uncertainties on the radioactive decay constants of the activation products and  
 245 their gamma-ray intensities, provided by JEFF-3.3 database [26], which are lower  
 246 than 4 %;
- 247 - the uncertainty on the net area of the gamma rays reported in Table 4, which is at  
 248 most 2.1 %. It is provided by the Genie2000 software and takes into account the  
 249 uncertainty related to the counting statistics as well as that related to the fitting  
 250 procedure.

251 The uncertainty associated to the mean photon flux is here estimated, conservatively, as  
 252 the quadratic combination of the mean uncertainty of the five calculated photon fluxes  
 253 (around 12 %) and the standard deviation of the photon fluxes obtained with the five  
 254 materials:

$$255 \quad \frac{\sigma_{distribution}}{\bar{\phi}} = \frac{1}{\bar{\phi}} \sqrt{\frac{(\phi_{Au}-\bar{\phi})^2 + (\phi_{Ni}-\bar{\phi})^2 + (\phi_U-\bar{\phi})^2 + (\phi_{Zn}-\bar{\phi})^2 + (\phi_{Zr}-\bar{\phi})^2}{5}} = 7 \%;$$

256

257 The electron distribution corresponding to the most likely, 17.5 MeV endpoint energy and  
 258 0.5 MeV width at half maximum, is shown in Figure 8, and the corresponding  
 259 Bremsstrahlung photon distribution calculated with MCNP is given in Figure 9.

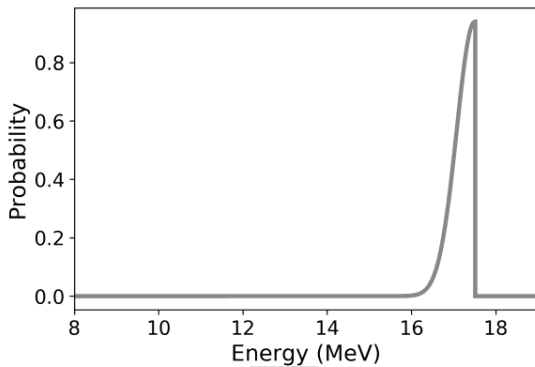


Figure 8: Most probable electron energy distribution (source of MCNP flux and reaction rate calculations).

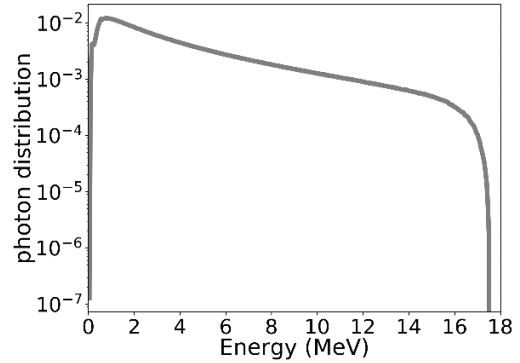


Figure 9: Bremsstrahlung photon energy distribution (normalized) calculated with MCNP and the electron energy distribution of Figure 8.

260 The mean experimental photon flux of  $(6.55 \pm 0.94) \times 10^{10} \text{ photons.cm}^{-2}.\text{s}^{-1}$ , at the  
 261 center of the beam and 88 cm away from the tungsten target, will be used to normalize  
 262 MCNP simulation results that are given per electron impinging on the tungsten target.

#### 263 4. Comparison of simulated photofission and neutron fission rates

264 The photofission rate in the samples is defined by (1) for a mixture of  $n$  actinides.

$$\tau_p = \frac{N_A}{\mathcal{M}} \left( \sum_{i=1}^n m_i \int_{E_{threshold}}^{E_{max}} \varphi(E) \sigma_{i(\gamma,f)}(E) dE \right) \quad (1)$$

265 Where:

- 266 -  $\tau_p$  is the photofission rate in  $s^{-1}$ ;
- 267 -  $N_A$  is the Avogadro constant, equals to  $6.02 \times 10^{23} \text{ mol}^{-1}$ ;
- 268 -  $\mathcal{M}$  is the molar mass of the actinide mixture, expressed in  $\text{g.mol}^{-1}$ ;
- 269 -  $m_i$  is the mass of actinide  $i$  in the sample, in g;
- 270 -  $E_{threshold}$  and  $E_{max}$  are respectively the threshold energy of the photofission  
271 reactions, around 6 MeV, and the Bremsstrahlung endpoint-energy, 17.5 MeV in  
272 our case;
- 273 -  $\varphi(E)$  is the Bremsstrahlung photon flux at the energy  $E$  in the sample, in  
274  $\text{photons.cm}^{-2}.\text{s}^{-1}$ ;
- 275 -  $\sigma_{i(\gamma,f)}(E)$  is the photofission reaction cross-sections at the energy  $E$  for actinide  $i$ ,  
276 in  $\text{cm}^2$ .

277 The uncertainty on the photofission rate calculated with MCNP is the quadratic  
278 combination of the following uncertainties:

- 279 - a 14.3 % relative uncertainty on the Bremsstrahlung photon flux, determined in  
280 section 3,  $(6.55 \pm 0.94) \times 10^{10} \text{ photons.cm}^{-2}.\text{s}^{-1}$  (see Table 5);
- 281 - a 0.1 % relative statistical uncertainties on MCNP calculations for the photofission  
282 rate in the plutonium, DU and HEU samples;
- 283 - a 2 % uncertainty on the photofission cross-section, according to datasets available  
284 in the EXFOR library [29];

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

$$288 \tau_{p,\text{Pu } 1} = (1.32 \pm 0.19) \cdot 10^7 \text{ s}^{-1}$$

$$289 \tau_{p,\text{Pu } 2} = (2.33 \pm 0.34) \cdot 10^6 \text{ s}^{-1}$$

$$\tau_{p,DU} = (2.66 \pm 0.38) \cdot 10^8 \text{ s}^{-1}$$

$$\tau_{p,HEU} = (3.38 \pm 0.48) \cdot 10^7 \text{ s}^{-1}$$

It was shown in our previous work [19] that neutron fissions in the actinides are mainly due to fast neutrons produced in the samples themselves. Indeed, the neutron shielding around the LINAC head (tungsten target and lead collimator), composed of borated polyethylene and cadmium, was proven to be efficient since the presence of thermal neutrons was not observed from neutron activation of the metallic pellets. Indeed, although radiative capture gamma rays of  $^{116m}\text{In}$ , due to the  $^{115}\text{In}(n,\gamma)^{116m}\text{In}$  activation reaction, highlight the presence of epithermal neutrons with an energy larger than 0.5 MeV (not absorbed by the cadmium foil in front of the LINAC head), thermal neutrons are not detected through the activation of the gold foil. Indeed, we do not observe in the gamma spectrum the 411.8 keV line of  $^{198}\text{Au}$ , which was expected from the  $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$  reaction, despite a high cross section for thermal neutrons of 100 b ( $1 \text{ b} = 10^{-28} \text{ m}^2$ ) at 0.025 eV according to ENDF/B-VIII.0 library [23].

MCNP simulations were conducted to characterize the origin of the fissions occurring in the samples, i.e. photofission vs. neutron fissions are reported in Table 6, in order to calculate the photofission yields as precisely as possible for the three actinides.

Table 6: Origin of the fissions in the different actinide samples.

Plutonium		Uranium		
Sample	Pu	Sample	DU	HEU
Photofissions	98.3 %	Photofissions of $^{238}\text{U}$	94.2 %	3.6 %
Neutron fissions	1.7 %	Photofissions of $^{235}\text{U}$	-	93.1 %
		Neutron fissions of $^{238}\text{U}$	5.8 %	-
		Neutron fissions of $^{235}\text{U}$	-	3.3 %

In the plutonium sample, the only 1.7 % of neutron fissions will be subtracted to obtain the photofission yields of  $^{239}\text{Pu}$ . Besides, given the similarity between the photofission cross sections of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  (cf. Figure 4), we assume that all photofissions occur on  $^{239}\text{Pu}$ . Concerning the DU sample with a  $^{235}\text{U}$  content of 0.3 %, we consider that all fissions occur on  $^{238}\text{U}$ . Among them, 5.8 % are  $^{238}\text{U}$  fissions caused by fast neutrons. As a result, the delayed gamma rays measured with the DU sample are used to directly calculate the  $^{238}\text{U}$

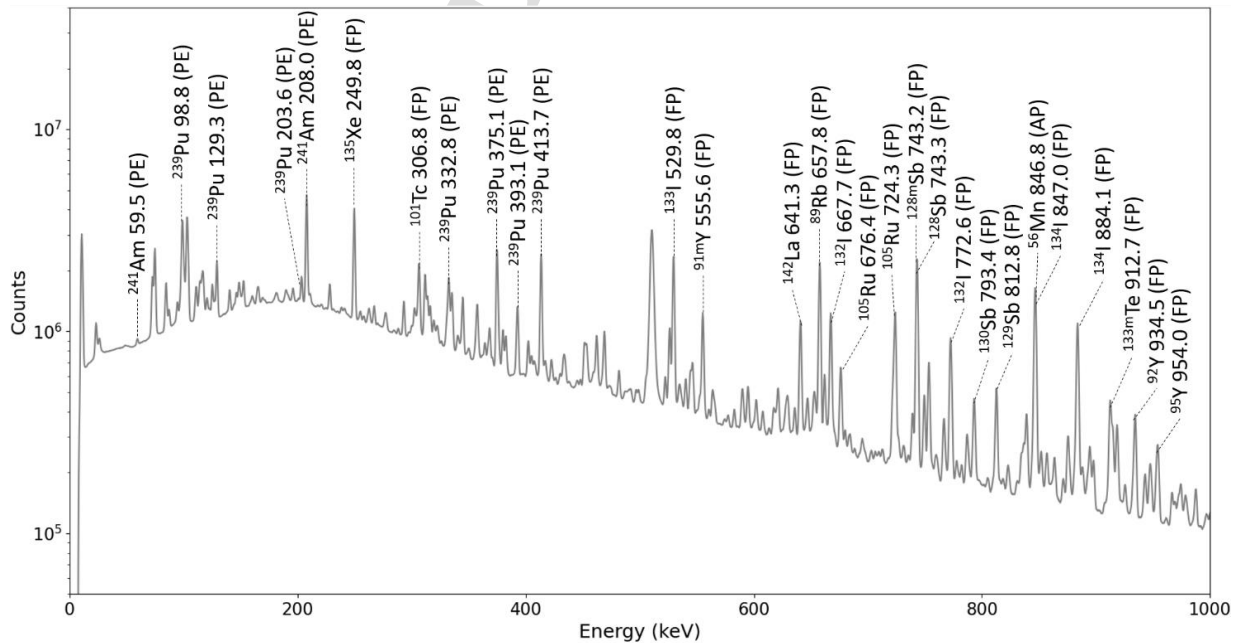


315 photofission products cumulative yields, after subtraction of the neutron fission  
 316 contribution. In the HEU sample, however, 3.6 % of photofissions occur on  $^{238}\text{U}$  and  
 317 93.1 % on  $^{235}\text{U}$ . Moreover, 3.3 % of fissions are fast neutron fissions on  $^{235}\text{U}$ . Therefore,  
 318 the  $^{238}\text{U}$  contribution to photofission and the  $^{235}\text{U}$  contribution to neutron fission will be  
 319 subtracted to calculate the  $^{235}\text{U}$  photofission products cumulative yields.

## 320 5. Cumulative yields of $^{239}\text{Pu}$ , $^{235}\text{U}$ and $^{238}\text{U}$ photofission products

321 Figures 10 to 12 show the delayed gamma spectrum of the plutonium sample (denoted  
 322 as the Pu 1 measurement in Table 3) recorded during 24 h, after a 2 h irradiation with a  
 323 17.5 MeV endpoint energy Bremsstrahlung photon beam and a 35 s cooling time. The  
 324 notations used are PE for Passive Emission, AP for Activation Product and FP for Fission  
 325 Product. Note that delayed gamma spectra for DU and HEU photofission products have  
 326 already been presented in our previous work [19].

327

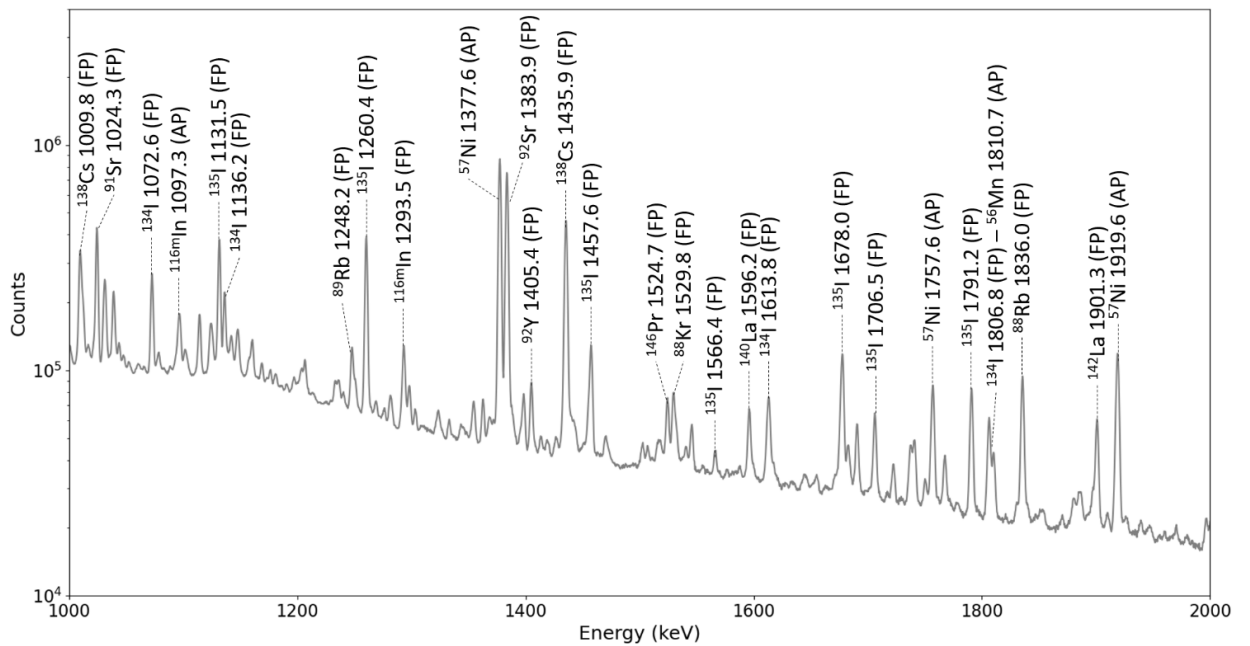


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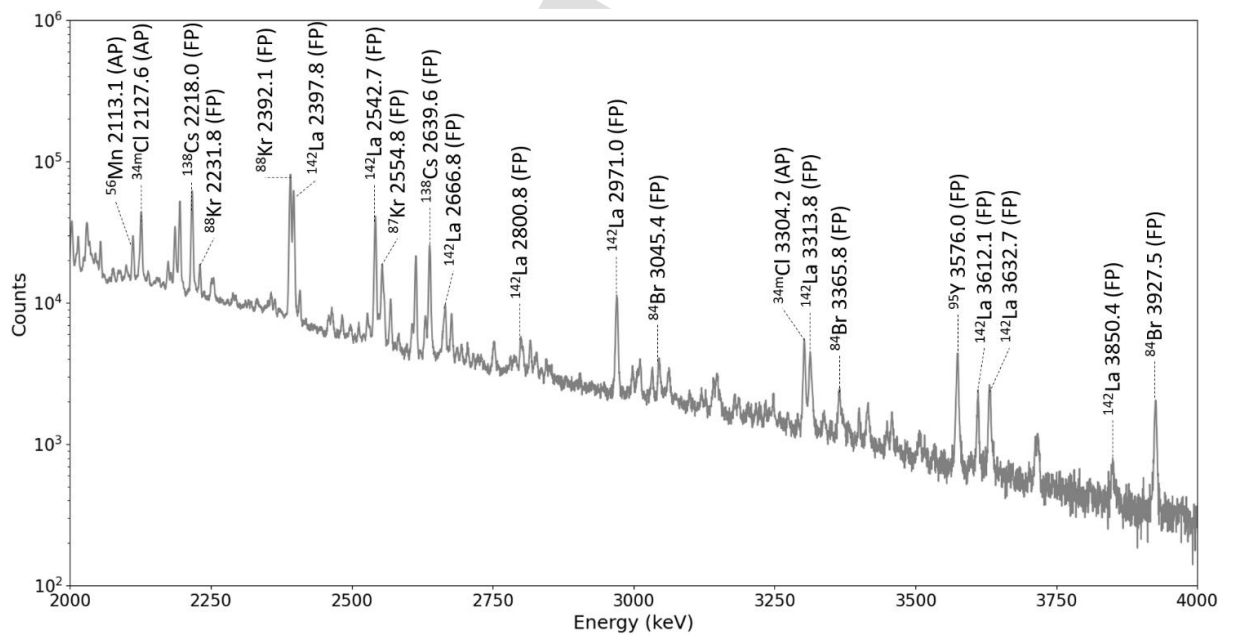
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Figure 10: Delayed gamma spectrum of the plutonium sample (0-1000 keV) recorded during 24 h, after a 2 h irradiation and a 35 s cooling time.



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Figure 11: Delayed gamma spectrum of the plutonium sample (1000-2000 keV) recorded during 24 h, after a 2 h irradiation and a 35 s cooling time.



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335  
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Figure 12: Delayed gamma spectrum of the plutonium sample (2000-4000 keV) recorded during 24 h, after a 2 h irradiation and a 35 s cooling time.

337 For all actinide samples, the delayed gamma-ray spectra have been recorded by  
338 sequences of 60 s during several dozens of hours, which allows a spectrum analysis with  
339 different cooling and measurement times to limit some interferences between close-in-  
340 energy gamma rays, by exploiting the differences in the radioactive periods of their

341 emitting isotopes (we can enhance short-lived isotopes by summing the spectra acquired  
 342 shortly after irradiation, and long-lived ones latter). The spectra analysis is performed with  
 343 the MAGIX software (see description in Section 2).

344 Theoretically speaking, the fission products created during irradiation are part of  
 345 radioactive decay chains and their activities can be calculated by solving Bateman  
 346 equations [31]. In general, these equations can be simplified by considering only the  
 347 activation of the photofission product emitting the delayed gamma rays of interest, as  
 348 below in (2). However, as explained further to introduce (3), it is sometimes needed to  
 349 consider the direct precursor of the photofission product of interest, which are respectively  
 350 called the father and daughter nuclides, like in the work of Kahane *et al.* [6] and Carrel *et*  
 351 *al.* [7].

352 For a mixture of two actinides  $k$  and  $l$ , when the delayed gamma rays are emitted by a  
 353 nucleus with a much longer half-life than its precursors and the cooling time, the net area  
 354  $N(E_i)$  of its gamma rays of energy  $E_i$  is directly related to cumulative photofission yields  
 355  $Y_{ck,p}$  and  $Y_{cl,p}$  through equation (2):

$$N(E_i) = \frac{I(E_i) \varepsilon(E_i)}{\lambda_j} (1 - e^{-\lambda_j t_{irr}}) e^{-\lambda_j t_{cool}} (1 - e^{-\lambda_j t_{count}}) [\tau_p (\eta_{k,p} Y_{ck,p} + \eta_{l,p} Y_{cl,p}) + \tau_n (\eta_{k,n} Y_{ck,n} + \eta_{l,n} Y_{cl,n})] \quad (2)$$

356 With:

- 357 -  $I(E_i)$  the gamma-ray intensity taken from JEFF-3.3 database [26];
- 358 -  $\varepsilon(E_i)$  the absolute detection efficiency taking into account, in addition to the  
 359 abovementioned intrinsic detector and geometric efficiencies, the interrogating  
 360 photon flux self-shielding and delayed gamma self-attenuation in the uranium  
 361 sample, both estimated with MCNP. For example, regarding the self-shielding in  
 362 the DU sample, the photofission rate is 2.5 times higher on the entrance surface of  
 363 the sample, with respect to the LINAC photon beam, than on its rear exit surface.  
 364 Concerning self-attenuation, for instance, less than 60 % of 1 MeV delayed gamma  
 365 rays emitted in the DU sample manage to escape from it;
- 366 -  $\lambda_j$  the radioactive decay constant of the fission product  $j$  in  $s^{-1}$ ;
- 367 -  $t_{irr}$ ,  $t_{cool}$  and  $t_{count}$  respectively the irradiation, cooling and counting time, in s;
- 368 -  $\tau_p$  and  $\tau_n$  the photofission and neutron fission rates in the sample, in  $s^{-1}$ ;

- 369 -  $Y_{ck,p}$  and  $Y_{cl,p}$  the cumulative photofission yields of photofission product  $j$ ,  
 370 respectively for actinides  $k$  and  $l$  (for example  $^{235}\text{U}$  and  $^{238}\text{U}$ );  
 371 -  $Y_{ck,n}$  and  $Y_{cl,n}$  the cumulative fast neutron fission product yields of fission product  
 372  $j$ , respectively for actinides  $k$  and  $l$ , taken from ENDF/B-VIII.0 database [23];  
 373 -  $\eta_{k,p}$  and  $\eta_{l,p}$  are the fractions of photofissions occurring respectively in actinides  
 374  $k$  and  $l$ , determined via MCNP simulations;  
 375 -  $\eta_{k,n}$  and  $\eta_{l,n}$  are the fractions of neutron fissions occurring respectively in actinides  
 376  $k$  and  $l$ , determined via MCNP simulations.

377 However, when the photofission product of interest is a daughter nuclide in a decay chain  
 378 with a father having a similar half-life (for instance  $^{134}\text{I}$  and  $^{138}\text{Cs}$ , which are respectively  
 379 the daughters of  $^{134}\text{Te}$  and  $^{138}\text{Xe}$ , with respective radioactive periods of 41.8 and 33.4 min),  
 380 net peak areas are given by (3).

$$N(E_i) = I(E_i)\varepsilon(E_i)(\tau_p[\eta_{k,p}(Y_{ik,p}^d f_d + Y_{ck,p}^f f_f) + \eta_{l,p}(Y_{il,p}^d f_d + Y_{cl,p}^f f_f)] \dots \quad (3)$$

$$\dots + \tau_n[\eta_{k,n}(Y_{ik,n}^d f_d + Y_{ck,n}^f f_f) + \eta_{l,n}(Y_{il,n}^d f_d + Y_{cl,n}^f f_f)])$$

381 Where:

- 382 -  $I(E_i)$ ,  $\varepsilon(E_i)$ ,  $\tau_p$ ,  $\tau_n$ ,  $\eta_{k,p}$ ,  $\eta_{l,p}$ ,  $\eta_{k,n}$  and  $\eta_{l,n}$  keep the same meaning as in (2);  
 383 -  $Y_{ck,p}^f$  and  $Y_{cl,p}^f$  are the cumulative yields of the father nuclide, respectively for the  
 384 photofission of actinides  $k$  and  $l$ ;  
 385 -  $Y_{ik,p}^d$  and  $Y_{il,p}^d$  are the independent yields of the daughter nuclide, respectively for  
 386 the photofission of actinides  $k$  and  $l$ . The independent yield (in %) of a  
 387 photofission product corresponds to the number of nuclei created per 100  
 388 photofissions of the considered actinide, right after the prompt neutron emission  
 389 but before the delayed neutron emission. These values will be calculated to  
 390 determine the cumulative photofission yield of the daughter nuclide, which is the  
 391 sum of its independent yield with that of its precursor. Note that, as in Kahane *et al.*  
 392 [6] and Carrel *et al.* [7], we only consider only one precursor, since the  
 393 precursors of the father nuclide have a relatively short half-life compared to the  
 394 cooling time of these experiments.

395 In this case, we can write  $Y_{c,p}^d = Y_{i,p}^d + Y_{c,p}^f$

- 396 -  $Y_{ck,n}^f$  and  $Y_{cl,n}^f$  are the cumulative yields of the father nuclide, respectively for the  
 397 neutron fission of actinides  $k$  and  $l$ . These values are taken from ENDF/B-VIII.0  
 398 database [23];
- 399 -  $Y_{ik,n}^d$  and  $Y_{il,n}^d$  are the independent yields of the daughter nuclide, respectively for  
 400 the neutron fission of actinides  $k$  and  $l$ . These values are taken from ENDF/B-  
 401 VIII.0 database [23];
- 402 - The terms  $f_d$  and  $f_f$  describe the evolution of the respective numbers of daughter  
 403 and father nuclei over time, and are given by:

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

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

404 With  $\lambda_d$  and  $\lambda_f$  the radioactive decay constants of the daughter and father nuclides  
 405 (in  $s^{-1}$ ), respectively, and  $t_{irr}$ ,  $t_{cool}$ ,  $t_{count}$  the irradiation, cooling and counting  
 406 times (in s).

407 Note that when the half-life of the father nuclide is much shorter than that of the daughter  
 408 nuclide, the cumulative yield of the daughter nuclide can be estimated with (2) by analyzing  
 409 the delayed gamma spectrum after a cooling time equal to six times the half-life of the  
 410 father nuclide (corresponding to the decay of 98.5 % of father nuclei). This approach is  
 411 used in practice for the majority of fission products, the precursors of which having very  
 412 short half-lives.

413 For the plutonium sample, the 1.7 % of neutron fissions (see Table 6) are subtracted to  
 414 obtain the cumulative photofission yields of  $^{239}\text{Pu}$ . Concerning the DU sample, with a  $^{235}\text{U}$   
 415 enrichment of 0.3 %, the measured delayed gamma rays directly lead to the cumulative  
 416 yields of  $^{238}\text{U}$  photofission products, after subtraction of neutron fissions on  $^{238}\text{U}$  that  
 417 represent 5.8 % of total fissions in the sample. For the HEU sample with more than 90 %  
 418 of  $^{235}\text{U}$ , the 3.3 % of neutron fissions are subtracted to obtain the photofission rate, and the  
 419 3.6 % of photofissions in  $^{238}\text{U}$  are subtracted to calculate the cumulative yields of  $^{235}\text{U}$   
 420 photofission products. For this purpose, we use the photofission yields obtained in this  
 421 work for  $^{238}\text{U}$  to evaluate the  $^{235}\text{U}$  yields. For each case, the fast neutron fission yields

422 available in ENDF/B-VIII.0 database [23] are used to subtract the neutron fission  
 423 contribution to the net areas of the gamma rays emitted by the fission products created in  
 424 the actinide samples.

425 Finally, 26 and 28 photofission products have been identified in the spectra from their  
 426 delayed gamma rays, respectively for  $^{239}\text{Pu}$  as well as  $^{235}\text{U}$  and  $^{238}\text{U}$ , and their cumulative  
 427 photofission yields calculated from (2) or (3). The cumulative photofission yields measured  
 428 for  $^{239}\text{Pu}$ ,  $^{238}\text{U}$  and  $^{235}\text{U}$  are given respectively in Table 7, Table 9 and Table 11. Note that  
 429 when several delayed gamma rays are measured for a photofission product, we calculate a  
 430 weighted average of the cumulative yields obtained with all the peaks that are correctly  
 431 processed by MAGIX software (outliers of the net areas, for instance due to interferences,  
 432 are discarded). The weighting coefficient for each gamma ray is defined as the inverse of  
 433 the squared relative uncertainty on the net peak area, as described in (4). Note that the  
 434 weighted mean does not take into account the accuracy of the emission intensity of the  
 435 considered gamma rays.

$$\bar{y}_c = \frac{\sum_{i=1}^N \frac{y_{c,i}(E_i)}{\left(\frac{\sigma(N(E_i))}{N(E_i)}\right)^2}}{\sum_{i=1}^N \frac{1}{\left(\frac{\sigma(N(E_i))}{N(E_i)}\right)^2}} \quad (4)$$

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

439

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

- 442 - the 14.3 % uncertainty on the photofission rate in the samples (refer to section 4)
- 443 mainly due to the uncertainty on the interrogating Bremsstrahlung photon flux
- 444 (section 3). This is the largest part of the overall uncertainty, and it could be
- 445 reduced in the future by accurately measuring the  $(\gamma,n)$  cross-sections of the
- 446 activation materials used to characterize the photon beam, since their uncertainties
- 447 are around 10 % in EXFOR library [29];

448 - the uncertainty on the absolute detection efficiency of the gamma ray of energy  $E_i$ ,  
 449 which is estimated to 7 %;

450 - the relative statistical uncertainty on the weighted average (5),  $\frac{\sigma_{stat}(\bar{y}_c)}{\bar{y}_c}$ , with

$$451 \quad \sigma_{stat}(\bar{y}_c) = \sqrt{\frac{1}{\sum_{i=1}^N \left( \frac{N(E_i)}{\sigma(N(E_i))} \right)^2}}.$$

452 The  $^{239}\text{Pu}$  cumulative photofission yields measured with the plutonium sample are given  
 453 in Table 7 and we provide in Table 8 all the details of the delayed gamma-ray analysis:  
 454 cooling and counting times for each delayed gamma ray, energy and intensity coming from  
 455 JEFF-3.3 database [26], net peak area with its associated uncertainty, and cumulative yield  
 456 computed for each line. Our data (“this work”) are then compared to the values of  
 457 photofission products cumulative yields previously published. Note that we report a simple  
 458 average of the yields when several gamma rays are given in the other publications. The  
 459 characteristics of the interrogating photon beams are indicated in the first two lines of Table  
 460 7. For Bremsstrahlung photon beams, the energy indicated corresponds to the endpoint  
 461 energy. The nuclides half-lives are from JEFF-3.3 database [26], except for  $^{92}\text{Sr}$  because it  
 462 is not consistent in the different databases, its half-life being taken from Leconte *et al.* [32].

463

464 *Table 7 : Cumulative yields (number of photofission products per 100 fissions) for the photofission of  $^{239}\text{Pu}$  and*  
 465 *comparison with published data*

<i>Photon beam type</i>		Bremsstrahlung	Bremsstrahlung	Bremsstrahlung	Monoenergetic	Monoenergetic
<i>Energy (MeV)</i>		17.5	28.0	22.0	11.0	13.0
<b>Fission product</b>	<b>T<sub>1/2</sub></b>	<b>This work</b>	<b>Kondrat'ko 1981 [13]</b>	<b>Wen 2016 [11]</b>	<b>Bhike 2017 [14]</b>	<b>Krishichayan 2019 [12]</b>
$^{84}\text{Br}$	31.8 min	$1.19 \pm 0.19$	-	-	-	-
$^{87}\text{Kr}$	1.3 h	$2.03 \pm 0.32$	-	-	-	$1.45 \pm 0.12$
$^{88}\text{Kr}$	2.8 h	$2.36 \pm 0.36$	$1.62 \pm 0.21$	-	-	$2.08 \pm 0.15$
$^{89}\text{Rb}$	<sup>(a)</sup> 15.4 min	$3.99 \pm 0.61$	-	$3.45 \pm 0.43$	-	-
$^{91}\text{Sr}$	9.7 h	$4.63 \pm 0.72$	$2.89 \pm 0.23$	-	$4.15 \pm 0.51$	$3.91 \pm 0.24$
$^{91\text{m}}\text{Y}$	<sup>(a)</sup> 49.7 min	$3.19 \pm 0.56$	-	-	-	-
$^{92}\text{Sr}$	2.6 h	$4.73 \pm 0.76$	-	$4.00 \pm 0.20$	$4.21 \pm 0.49$	$4.19 \pm 0.66$
$^{92}\text{Y}$	3.5 h	$5.42 \pm 0.93$	$3.20 \pm 0.16$	-	-	-
$^{93}\text{Y}$	10.2 h	$4.45 \pm 0.58$	$4.02 \pm 0.20$	-	-	-
$^{94}\text{Y}$	<sup>(a)</sup> 18.7 min	$7.05 \pm 1.13$	-	$4.60 \pm 0.32$	-	$4.71 \pm 0.33$

<sup>97</sup> Zr	16.7 h	8.81 ± 1.41	4.63 ± 0.17	-	6.63 ± 0.73	6.78 ± 0.38
<sup>104</sup> Tc <sup>(a)</sup>	18.3 min	6.91 ± 1.12	-	-	-	1.96 ± 0.14
<sup>105</sup> Ru	4.4 h	6.29 ± 1.01	-	-	-	6.16 ± 0.39
<sup>105</sup> Rh	34.5 h	7.08 ± 1.11	3.96 ± 0.20	-	-	-
<sup>128</sup> Sn	59.1 min	1.14 ± 0.18	-	-	-	1.18 ± 0.16
<sup>128</sup> Sb	9 h	1.14 ± 0.19	-	-	-	-
<sup>129</sup> Sb	4.4 h	2.60 ± 0.42	-	-	-	2.82 ± 0.15
<sup>130</sup> Sb	39.5 min	1.64 ± 0.26	-	1.25 ± 0.12	-	1.36 ± 0.11
<sup>131</sup> Sb <sup>(a)</sup>	23 min	2.84 ± 0.45	-	2.10 ± 0.34	-	-
<sup>134</sup> Te	41.8 min	4.88 ± 0.78	-	11.6 ± 0.81	-	-
<sup>134</sup> I	52.5 min	7.02 ± 1.16	-	5.90 ± 0.15	7.39 ± 0.89	-
<sup>135</sup> I	6.6 h	5.67 ± 0.94	-	-	-	-
<sup>138</sup> Cs	33.4 min	8.24 ± 1.33	-	5.50 ± 0.29	6.18 ± 0.75	6.45 ± 0.37
<sup>141</sup> Ba <sup>(a)</sup>	18.3 min	7.28 ± 1.20	-	-	-	4.22 ± 0.31
<sup>142</sup> La	1.5 h	6.27 ± 1.00	-	4.90 ± 0.29	-	5.87 ± 0.31
<sup>143</sup> Ce	1.4 d	4.48 ± 0.76	3.26 ± 0.13	-	4.41 ± 0.51	3.88 ± 0.20

<sup>(a)</sup>Results obtained with the measurement referred to as Pu 2 in Table 3 and Figure 5. The other cumulative photofission yields of <sup>239</sup>Pu are obtained with the Pu 1 measurement.

466

467

Table 8: Detailed data of the delayed gamma-ray analysis for each <sup>239</sup>Pu photofission product

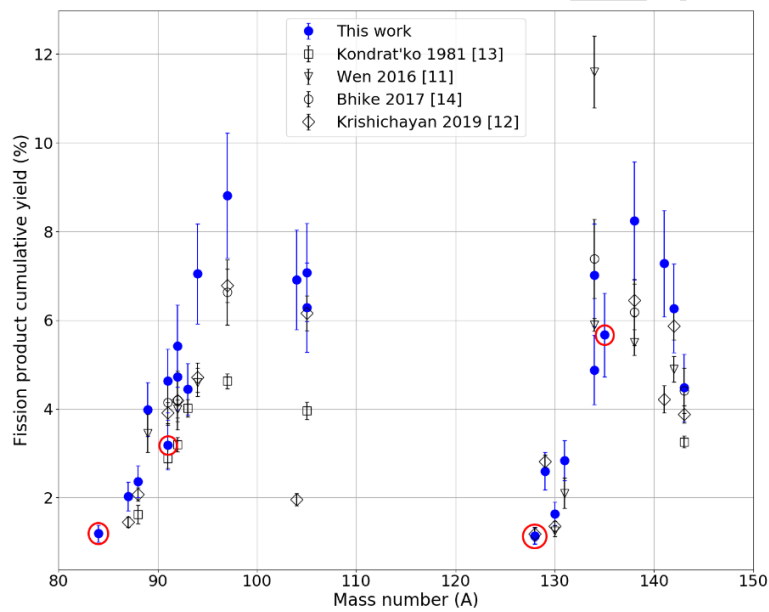
Fission Product	Cooling time	Counting time	Gamma-ray energy (keV)	Relative intensity (%)	Net peak area (counts)	Photofission cumulative yield (%)
<sup>84</sup> Br	50.1 min	2.3 h	1897.60	14.56	24858 ± 312	1.23
			1015.90	6.16	18493 ± 565	1.46
			2484.10	6.66	7682 ± 164	1.01
			3927.50	6.78	5977 ± 79	1.16
			1463.80	1.96	3631 ± 357	1.13
<sup>87</sup> Kr	50.1 min	6.8 h	402.59	49.60	958383 ± 1801	2.00
			2554.80	9.23	74263 ± 308	2.17
			2558.10	3.92	31501 ± 229	2.17
<sup>88</sup> Kr	50.1 min	16.2 h	196.30	25.98	547139 ± 2605	2.23
			2392.11	34.60	525449 ± 752	2.31
			834.83	12.97	426400 ± 1122	2.64
			1529.77	10.93	224744 ± 660	2.28
			2195.84	13.18	222652 ± 544	2.40
			2029.84	4.53	80749 ± 413	2.39
			2035.41	3.74	66486 ± 394	2.39
			2231.77	3.39	51489 ± 342	2.19
1518.39	2.15	44384 ± 508	2.28			
<sup>89</sup> Rb	9.5 min	1.7 h	1032.00	63.60	30790 ± 352	3.80
			1248.20	45.60	21750 ± 277	4.24
			657.80	11.00	7528 ± 523	4.24
			2570.10	10.18	2629 ± 82	3.81
<sup>91</sup> Sr	1.8 h	22.9 h	1024.30	33.50	1799550 ± 1512	4.50
			749.80	23.68	1580910 ± 1540	4.86
			652.90	8.04	510024 ± 1294	4.33



			652.30	2.98	189136 ± 1163	4.33
<sup>91m</sup> Y	9.5 min	20.4 h	555.57	95.00	302322 ± 923	3.19
<sup>92</sup> Sr	1.8 h	22.9 h	1383.90	93.00	3168600 ± 1831	4.74
			953.30	3.62	148282 ± 809	4.64
			1142.39	2.86	108084 ± 785	4.70
<sup>92</sup> Y	1.8 h	22.9 h	934.50	13.90	1140370 ± 1291	5.42
<sup>93</sup> Y	1.8 h	22.9 h	266.90	7.42	406743 ± 2240	3.64
			947.10	2.12	137801 ± 806	5.35
			680.20	0.67	45512 ± 1029	4.79
<sup>94</sup> Y	9.5 min	1.8 h	918.74	56.00	70359 ± 1167	7.05
<sup>97</sup> Zr	1.8 h	22.9 h	1147.97	2.62	211786 ± 796	8.81
<sup>104</sup> Tc	9.5 min	2.1 h	535.10	14.69	22763 ± 609	7.00
			884.40	10.95	12685 ± 394	6.44
			893.10	10.23	13116 ± 394	7.18
			1676.80	7.83	6592 ± 196	7.12
			1612.40	5.79	4632 ± 199	6.59
<sup>105</sup> Ru	1.8 h	22.9 h	724.30	47.30	3994660 ± 2209	6.30
			469.37	17.55	1727880 ± 2019	6.22
			676.36	15.66	1365110 ± 1557	6.30
			316.44	11.12	1076530 ± 2253	6.06
			393.36	3.78	383858 ± 1781	6.22
			875.85	2.50	211630 ± 895	6.93
<sup>105</sup> Rh	1.8 h	22.9 h	969.44	2.11	164478 ± 813	6.66
			318.90	19.10	1045508 ± 2234	7.07
<sup>105</sup> Rh	1.8 h	22.9 h	306.10	5.10	285966 ± 2103	7.28
<sup>128</sup> Sn	50.1 min	5.1 h	482.30	59.00	483730 ± 1472	1.14
<sup>128</sup> Sb	1.8 h	22.9 h	314.10	61.00	1233351 ± 2298	1.20
			754.00	100.00	1639830 ± 1557	1.18
			636.20	36.00	457603 ± 1144	0.86
			628.70	31.00	415906 ± 2162	0.91
<sup>129</sup> Sb	1.8 h	22.9 h	812.80	47.60	1586460 ± 1505	2.66
			914.50	20.94	584657 ± 1065	2.36
			760.80	3.33	114663 ± 934	2.66
			772.80	3.05	104078 ± 911	2.66
			876.00	2.86	81428 ± 819	2.36
<sup>130</sup> Sb	50.1 min	3.1 h	793.40	100.00	549107 ± 1050	1.63
			330.91	78.00	569154 ± 1592	1.72
			839.52	100.00	533861 ± 1023	1.63
			182.33	65.00	204834 ± 1730	1.52
			732.00	22.00	100176 ± 888	1.30
<sup>131</sup> Sb	9.5 min	2.1 h	943.40	46.20	29222 ± 394	2.78
			933.10	25.87	17640 ± 382	2.98
<sup>134</sup> Te	50.1 min	3.3 h	277.95	21.30	411731 ± 1685	4.88
			201.24	8.90	123133 ± 1767	4.92
<sup>134</sup> I	50.1 min	8.6 h	857.29	6.70	432668 ± 1051	7.21
			1613.80	4.31	177159 ± 583	6.50
			1741.49	2.57	107325 ± 487	7.34
<sup>135</sup> I	1.8 h	22.9 h	1260.41	28.70	1844600 ± 1454	5.87
			1131.51	22.59	1416620 ± 1386	5.40
			1038.76	7.95	552762 ± 1017	5.69
			836.80	6.69	476552 ± 1059	5.27
			1678.03	9.56	497395 ± 781	5.65
			1457.56	8.67	499735 ± 818	5.75
			1791.20	7.72	394075 ± 690	5.80
			1124.00	3.62	227641 ± 847	5.40
1706.46	4.10	211984 ± 563	5.67			
<sup>138</sup> Cs	50.1 min	4.7 h	1435.86	76.30	1214440 ± 1177	8.32
			1009.78	29.83	568411 ± 991	7.97
			2218.00	15.18	178761 ± 486	8.32
			871.8	5.11	103228 ± 795	7.77

			2639.59	7.63	$77034 \pm 304$	8.12
$^{141}\text{Ba}$	9.5 min	1.7 h	739.20	4.83	$6597 \pm 467$	7.07
			625.40	3.59	$5681 \pm 529$	7.64
$^{142}\text{La}$	50.1 min	9.3 h	641.29	47.40	$3033430 \pm 2131$	6.28
			894.90	8.34	$460260 \pm 1040$	6.33
			1901.30	7.16	$230758 \pm 578$	5.99
			1011.40	3.93	$202468 \pm 856$	6.34
			1043.70	2.70	$135455 \pm 805$	6.29
$^{143}\text{Ce}$	1.8 h	22.9 h	293.27	42.80	$1816230 \pm 2477$	4.78
			350.62	3.23	$144406 \pm 1899$	4.78
			231.55	2.05	$59502 \pm 2382$	4.08
			880.46	1.03	$34612 \pm 787$	4.82

468 Our photofission products cumulative yields data for  $^{239}\text{Pu}$  are compared in Figure 13 to  
 469 already published data.



470

471 *Figure 13: Fission product yields distribution for the photofission of  $^{239}\text{Pu}$  and comparison with existing data.*

472 In this work, we provide the cumulative photofission yields of  $^{239}\text{Pu}$  fission products  $^{84}\text{Br}$ ,  
 473  $^{91\text{m}}\text{Y}$ ,  $^{128}\text{Sb}$  and  $^{135}\text{I}$  that were not published before (circled points in Figure 13). We can  
 474 note that relative yields were recently published by Parlag *et al.* [33] for  $^{135}\text{I}$  and  $^{91\text{m}}\text{Y}$  for  
 475 a 17.5 MeV Bremsstrahlung endpoint energy, but in this paper, all yields are normalized  
 476 to that of  $^{97}\text{Zr}$  published by Kondrat'ko *et al.* [13] (first dataset published in 1981  
 477 concerning the photofission yields of  $^{239}\text{Pu}$ ), which is significantly lower than other  
 478 published data and particularly our work, see Table 7. More globally, the photofission  
 479 yields provided in our work are a little larger but consistent with the other data. We can  
 480 note a few singular points like the  $^{104}\text{Tc}$  yield from Krishichayan *et al.* [12],  $1.96 \pm 0.14 \%$ ,

481 which is much lower than expected for this mass number. Our work gives  $6.91 \pm 1.12 \%$   
 482 and for comparison, the neutron fission yield is  $5.69 \%$  from ENDF/B-VIII.0 database [23].  
 483 We can also mention that Kondrat'ko *et al.* data [13] are globally low and some points  
 484 also appear as singular, such as the yield of  $^{105}\text{Rh}$ ,  $3.96 \pm 0.20 \%$ , compared to our  
 485 measurement,  $7.08 \pm 1.11 \%$ , and to the yields of  $^{105}\text{Ru}$  with the same atomic number (near  
 486  $6 \%$ ). The yield of  $^{134}\text{Te}$  from Wen *et al.* [11] also looks like an outlier with  $11.6 \pm 0.81 \%$ ,  
 487 compared to our data  $4.88 \pm 0.78 \%$  and to  $^{134}\text{I}$  with this atomic number (near  $6-7 \%$ ).  
 488 Except for the singular data mentioned above, the observed discrepancies in the published  
 489 photofission yields is probably due, for a significant part, to the different experimental  
 490 configurations (geometry of the samples, energy spectrum and intensity of the interrogating  
 491 photon beams, photoneutron production, irradiation-cooling-counting times, detectors,  
 492 etc.) and possibly to data analysis (subtraction of neutron fissions, gamma-ray  
 493 interferences, decay chain calculations to take into account precursors, etc.). In our case,  
 494 analyzing several gamma-ray lines for a same photofission product (when possible, for  
 495 example  $^{87}\text{Kr}$ ,  $^{88}\text{Kr}$ ,  $^{134}\text{I}$ ,  $^{135}\text{I}$ ,  $^{138}\text{Cs}$ ,  $^{142}\text{La}$ ) and observing a good consistency in the different  
 496 yields associated with each line improves the confidence of the weighted average reported  
 497 as cumulative yield. It is also important to mention that our results share a common  
 498 uncertainty of  $14 \%$  related to the photon flux assessment. In addition, our plutonium  
 499 sample is not exclusively made of  $^{239}\text{Pu}$ , the photofission yields of the other actinides  
 500 composing it ( $^{240}\text{Pu}$ ,  $^{242}\text{Pu}$ ,  $^{241}\text{Am}$ ) should be investigated with as pure as possible samples  
 501 in order to evaluate the contribution of each isotope.

502

503 The  $^{238}\text{U}$  cumulative photofission yields measured with the DU sample are given in Table  
 504 9, and the details of the delayed gamma-ray analysis in Table 10.

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Table 9: Cumulative yields (number of photofission products per 100 fissions) for the photofission of  $^{238}\text{U}$  and comparison with published data

Photon beam type		Bremsstrahlung	Bremsstrahlung	Neutron-capture gamma rays	Bremsstrahlung	Bremsstrahlung	Bremsstrahlung	Bremsstrahlung	Bremsstrahlung	Monoenergetic
Energy (MeV)		17.5	15.8	7.8	16.3	10.0	14.987	8.0	22.0	13.0
Fission Product	$T_{1/2}$	This work	Our previous work [19]	Kahane 1985 [6]	Carrel 2011 [7]	Naik 2011 [8]	Naik 2013 [9]	Naik 2014 [10]	Wen 2016 [11]	Krishichayan 2019 [12]
$^{84}\text{Br}$	31.8 min	$1.01 \pm 0.16$	$1.37 \pm 0.20$	-	-	-	$0.90 \pm 0.06$	$0.30 \pm 0.06$	-	$2.49 \pm 0.19$
$^{87}\text{Kr}$	1.3 h	$1.90 \pm 0.29$	$1.96 \pm 0.31$	$1.82 \pm 0.21$	-	$1.61 \pm 0.20$	$1.86 \pm 0.30$	$1.11 \pm 0.13$	$1.0 \pm 0.06$	$2.29 \pm 0.16$
$^{88}\text{Kr}$	2.8 h	$2.35 \pm 0.38$	$2.24 \pm 0.34$	-	$2.52 \pm 0.23$	$2.77 \pm 0.53$	$2.58 \pm 0.19$	$2.77 \pm 0.38$	$1.0 \pm 0.03$	$2.92 \pm 0.17$
$^{89}\text{Rb}$	15.4 min	$3.44 \pm 0.55$	$3.50 \pm 0.55$	$2.51 \pm 0.40$	$3.30 \pm 0.20$	-	$3.12 \pm 0.16$	$3.42 \pm 0.34$	$1.4 \pm 0.13$	-

<sup>91</sup> Sr	9.7 h	3.93 ± 0.64	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
<sup>92</sup> Sr	2.6 h	4.09 ± 0.66	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
<sup>94</sup> Y	18.7 min	4.87 ± 0.79	4.48 ± 0.77	-	5.06 ± 0.24	-	4.47 ± 0.25	4.25 ± 0.47	-	7.50 ± 0.46
<sup>97</sup> Zr	16.7 h	5.82 ± 0.94	5.95 ± 0.93	5.89 ± 0.66	-	5.43 ± 0.19	5.78 ± 0.17	6.00 ± 0.64	-	6.20 ± 0.19
<sup>99</sup> Mo	2.7 d	6.01 ± 0.97	4.65 ± 0.77	-	-	4.84 ± 0.44	5.11 ± 0.15	4.75 ± 0.50	-	6.57 ± 0.22
<sup>101</sup> Mo	14.6 min	6.56 ± 1.09	5.37 ± 0.86	-	6.78 ± 0.32	-	7.13 ± 0.30	7.56 ± 0.77	-	-
<sup>104</sup> Tc	18.3 min	3.80 ± 0.63	3.60 ± 0.56	4.13 ± 0.50	-	-	3.65 ± 0.28	4.06 ± 0.42	-	4.44 ± 0.64
<sup>105</sup> Ru	4.4 h	2.78 ± 0.46	2.76 ± 0.44	2.95 ± 0.45	-	2.57 ± 0.21	2.55 ± 0.06	2.91 ± 0.38	-	4.06 ± 0.20
<sup>128</sup> Sn	59.1 min	0.58 ± 0.09	0.57 ± 0.09	-	-	-	0.85 ± 0.04	0.21 ± 0.03	-	1.22 ± 0.22
<sup>128</sup> Sb	9 h	0.22 ± 0.04	0.21 ± 0.03	-	0.16 ± 0.01	-	-	-	-	-
<sup>129</sup> Sb	4.4 h	1.24 ± 0.20	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
<sup>130</sup> Sb	39.5 min	0.89 ± 0.14	0.79 ± 0.12	-	1.08 ± 0.05	-	-	-	-	1.61 ± 0.11
<sup>131</sup> Sb	23 min	2.65 ± 0.43	2.51 ± 0.37	-	3.94 ± 0.19	-	4.18 ± 0.18	2.40 ± 0.27	-	5.01 ± 0.33
<sup>131m</sup> Te	1.3 d	0.86 ± 0.14	0.70 ± 0.10	3.12 ± 0.40	-	-	-	-	-	-
<sup>132</sup> Te	3.2 d	4.71 ± 0.77	4.71 ± 0.73	2.43 ± 0.50	-	4.84 ± 0.46	5.48 ± 0.14	6.15 ± 0.65	-	5.44 ± 0.25
<sup>132</sup> I	2.3 h	4.72 ± 0.79	4.87 ± 0.76	3.74 ± 0.46	-	-	-	-	-	-
<sup>133m</sup> Te	55.4 min	3.19 ± 0.51	3.23 ± 0.44	2.35 ± 0.39	4.43 ± 0.21	-	-	-	3.8 ± 0.42	-
<sup>134</sup> Te	41.8 min	5.58 ± 0.92	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
<sup>134</sup> I	52.5 min	7.00 ± 1.25	7.30 ± 1.16	6.29 ± 0.94	-	-	8.06 ± 0.34	8.63 ± 0.87	3.1 ± 0.19	-
<sup>135</sup> I	6.6 h	5.75 ± 0.95	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
<sup>138</sup> Xe	14.1 min	5.59 ± 0.90	3.75 ± 0.59	5.38 ± 0.90	6.60 ± 0.58	-	-	5.91 ± 0.63	1.8 ± 0.41	-
<sup>138</sup> Cs	33.4 min	5.87 ± 1.11	5.91 ± 0.86	6.10 ± 0.71	-	8.00 ± 0.48	6.84 ± 0.25	6.44 ± 0.68	2.6 ± 0.10	-
<sup>141</sup> Ba	18.3 min	5.48 ± 0.86	4.75 ± 0.73	-	-	-	4.51 ± 0.23	5.44 ± 0.56	-	-
<sup>142</sup> La	1.5 h	4.74 ± 0.79	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

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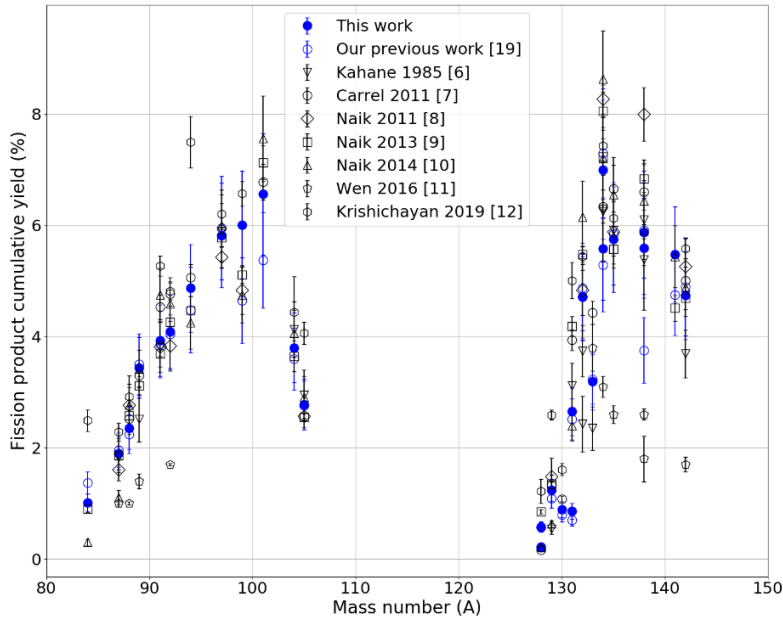
Table 10: Detailed data of the delayed gamma-ray analysis for each <sup>238</sup>U photofission product

Fission Product	Cooling time	Counting time	Gamma-ray energy (keV)	Relative intensity (%)	Net peak area (counts)	Photofission cumulative yield (%)
<sup>84</sup> Br	20.0 min	3.2 h	881.60	41.60	72262 ± 695	0.88
			1897.60	14.56	24041 ± 318	0.95
			1015.90	6.16	13767 ± 562	1.13
			802.20	5.99	9578 ± 723	0.82
			2484.10	6.66	10377 ± 173	1.03
			3927.50	6.78	8729 ± 95	1.20
			3365.80	2.87	3461 ± 78	0.99
3235.30	2.04	2452 ± 77	0.95			
<sup>87</sup> Kr	19.0 min	7.6 h	402.59	49.60	250860 ± 1359	1.79
			2554.80	9.23	60419 ± 281	1.97
			2558.10	3.92	25636 ± 211	1.97
			2011.88	2.88	19727 ± 313	1.79
<sup>88</sup> Kr	19.0 min	16.7 h	2392.11	34.60	400897 ± 659	2.34
			834.83	12.97	191623 ± 938	2.39
			1529.77	10.93	159206 ± 571	2.42
			2029.84	4.53	58502 ± 374	2.39
			2035.41	3.74	48190 ± 358	2.39
			2231.77	3.39	39608 ± 314	2.26

			1518.39	2.15	31391 ± 456	2.42
<sup>89</sup> Rb	19.0 min	1.7 h	1032.00	63.60	149511 ± 623	3.43
			1248.20	45.60	106176 ± 516	3.44
			657.80	11.00	23747 ± 801	3.47
			2570.10	10.18	17964 ± 177	3.47
<sup>91</sup> Sr	19.0 min	41.6 h	1024.30	33.50	1025360 ± 1223	3.97
			749.80	23.68	666694 ± 1258	3.83
<sup>92</sup> Sr	19.0 min	16.7 h	1383.90	93.00	2317890 ± 1589	4.09
<sup>94</sup> Y	19.0 min	1.9 h	918.74	56.00	260995 ± 1461	4.87
			550.9	4.93	19427 ± 892	4.94
<sup>97</sup> Zr	19.0 min	41.6 h	1149.97	2.62	105437 ± 705	5.83
			1750.24	1.09	39384 ± 399	5.78
<sup>99</sup> Mo	19.0 min	41.6 h	739.50	12.12	215280 ± 1085	6.01
<sup>101</sup> Mo	19.0 min	1.4 h	590.10	19.21	69639 ± 824	6.76
			1012.47	13.02	52392 ± 524	6.49
			1532.49	6.14	23139 ± 337	6.40
<sup>104</sup> Tc	19.0 min	1.9 h	358.00	89.00	178773 ± 1124	3.87
			535.10	14.69	40712 ± 913	3.57
			893.10	10.23	32790 ± 599	3.37
			1596.70	4.18	14101 ± 332	3.80
			1157.40	2.85	11038 ± 479	4.10
			2123.80	2.23	6507 ± 241	3.70
<sup>105</sup> Ru	19.0 min	26.2 h	724.30	47.30	919049 ± 1370	2.81
			676.36	15.66	275906 ± 1169	2.59
			469.37	17.55	260961 ± 1454	2.76
<sup>128</sup> Sn	19.0 min	5.6 h	482.30	59.00	93194 ± 1186	0.58
<sup>128</sup> Sb	1.5 h	40.4 h	754.00	100.00	141414 ± 801	0.22
<sup>129</sup> Sb	19.0 min	26.2 h	812.80	47.60	403876 ± 1076	1.24
			544.70	18.09	141730 ± 1277	1.36
			966.50	8.14	66692 ± 746	1.16
			683.50	5.66	44137 ± 1051	1.19
<sup>130</sup> Sb	1.0 h	3.0 h	839.52	100.00	103362 ± 624	0.85
			793.40	100.00	109564 ± 652	0.92
			330.91	78.00	43614 ± 946	0.92
			732.00	22.00	21619 ± 634	0.84
<sup>131</sup> Sb	19.0 min	1.9 h	943.40	46.20	162988 ± 677	2.66
			933.10	25.87	90233 ± 626	2.63
			1207.40	3.88	13656 ± 458	2.66
			2335.00	1.85	4798 ± 191	2.45
<sup>131m</sup> Te	19.0 min	41.6 h	852.21	21.40	90922 ± 897	0.86
<sup>132</sup> Te	40.2 min	41.3 h	228.33	88.12	301254 ± 8404	4.71
<sup>132</sup> I	19.0 min	41.6 h	667.71	98.7	1128590 ± 1521	4.72
<sup>133m</sup> Te	19.0 min	7.6 h	912.67	55.27	633754 ± 1055	3.17
			647.51	19.40	208071 ± 1112	3.25
			863.96	15.64	181664 ± 862	3.25
			914.77	10.94	123117 ± 772	3.10
<sup>134</sup> Te	19.0 min	3.9 h	767.20	29.60	448907 ± 1026	5.67
			565.99	18.60	230043 ± 1096	5.22
			277.95	21.30	123853 ± 1307	5.49
			464.64	5.03	54548 ± 1119	5.32
<sup>134</sup> I	19.0 min	9.2 h	884.09	65.08	2484463 ± 1738	6.95
			1136.16	9.09	364607 ± 843	7.36
			540.83	7.66	252969 ± 1237	7.49
			1613.80	4.31	159594 ± 543	7.32
<sup>135</sup> I	19.0 min	38.9 h	1260.41	28.70	1282530 ± 1250	5.86
			1131.51	22.59	919069 ± 1151	5.22
			1678.03	9.56	410034 ± 736	6.06
			1457.56	8.67	388947 ± 768	6.07
			1038.76	7.95	352406 ± 900	5.72
			1791.20	7.72	312318 ± 652	5.83

			836.80	6.69	$298895 \pm 1021$	5.95
			1706.46	4.10	$175261 \pm 549$	6.06
			1124.00	3.62	$147227 \pm 746$	5.22
$^{138}\text{Xe}$	19.0 min	1.4 h	1768.26	16.73	$49177 \pm 340$	5.59
$^{138}\text{Cs}$	19.0 min	4.4 h	462.80	30.75	$431772 \pm 1294$	5.83
			871.80	5.11	$100812 \pm 759$	6.13
			408.98	4.66	$58254 \pm 1179$	5.87
$^{141}\text{Ba}$	19.0 min	1.4 h	304.19	25.44	$53238 \pm 1030$	5.28
			343.67	14.44	$37741 \pm 988$	5.60
			739.20	4.83	$23791 \pm 682$	5.78
$^{142}\text{La}$	19.0 min	9.2 h	641.29	47.40	$743908 \pm 1202$	4.93
			2397.80	13.27	$156821 \pm 415$	4.26
			894.90	8.34	$134809 \pm 672$	4.60
			1901.30	7.16	$105968 \pm 397$	4.78
			1011.40	3.93	$66289 \pm 560$	4.76
			2055.20	2.18	$31017 \pm 274$	4.76
			3313.80	0.95	$9499 \pm 104$	4.55

509 Our fission products cumulative yields for  $^{238}\text{U}$  are compared in Figure 14 to other existing  
510 data.



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Figure 14: Fission product yields distribution for the photofission of  $^{238}\text{U}$  and comparison with existing data.

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The values provided for the cumulative photofission yields of  $^{238}\text{U}$  through this experimental campaign confirm the results obtained in our previous work [19]. No significant difference of the photofission yields is observed between the 15.8 MeV and 17.5 MeV electron endpoint energy, except that of  $^{138}\text{Xe}$  for which counting statistics was greatly improved in our new measurement, allowing to refine this yield (and in the same way for  $^{101}\text{Mo}$ , to a lesser extent). Wen *et al.* [11] data seem to show a systematic bias and are mostly below the expected values for the different mass numbers of the reported

520 photofission products. The yield of  $^{94}\text{Y}$  from Krishichayan *et al.* [12] also looks like an  
 521 outlier with  $7.50 \pm 0.46 \%$ , compared to our data  $4.87 \pm 0.79 \%$ , which is coherent with  
 522 all the other datasets displayed.

523

524 The  $^{235}\text{U}$  cumulative photofission yields measured with the HEU sample are given in Table 11  
 525 and the details of gamma analysis in  
 526 Table 12.

527  
 528

Table 11: Cumulative yields (number of photofission products per 100 fissions) for the photofission of  $^{235}\text{U}$  and comparison with published data

<i>Photon beam type</i>		Bremsstrahlung	Bremsstrahlung	Bremsstrahlung	Monoenergetic
<i>Energy (MeV)</i>		17.5	15.8	16.3	13.0
<b>Fission product</b>	<b>T<sub>1/2</sub></b>	<b>This work</b>	<b>Our previous work [19]</b>	<b>Carrel 2011 [7]</b>	<b>Krishichayan 2019 [12]</b>
$^{84}\text{Br}$	31.8 min	$1.71 \pm 0.30$	-	-	$1.77 \pm 0.13$
$^{87}\text{Kr}$	1.3 h	$3.07 \pm 0.53$	$4.64 \pm 0.74$	-	$3.45 \pm 0.28$
$^{88}\text{Kr}$	2.8 h	$3.59 \pm 0.59$	$5.34 \pm 0.84$	$3.63 \pm 0.32$	$3.87 \pm 0.22$
$^{89}\text{Rb}$	15.4 min	$4.76 \pm 0.82$	$6.89 \pm 1.08$	$4.69 \pm 0.28$	-
$^{91}\text{Sr}$	9.7 h	$4.62 \pm 0.82$	$7.71 \pm 1.18$	$5.37 \pm 0.26$	$6.08 \pm 0.26$
$^{92}\text{Sr}$	2.6 h	$5.17 \pm 0.90$	$7.92 \pm 1.24$	$5.59 \pm 0.26$	$6.52 \pm 0.21$
$^{94}\text{Y}$	18.7 min	$6.03 \pm 1.04$	-	$5.81 \pm 0.27$	$6.84 \pm 0.37$
$^{99}\text{Mo}$	16.7 h	$4.28 \pm 0.77$	-	-	$5.32 \pm 0.20$
$^{101}\text{Mo}$	14.6 min	$5.43 \pm 0.96$	-	$4.19 \pm 0.20$	-
$^{104}\text{Tc}$	18.3 min	$1.99 \pm 0.36$	$1.52 \pm 0.24$	-	$2.37 \pm 0.14$
$^{105}\text{Ru}$	4.4 h	$1.28 \pm 0.23$	$1.86 \pm 0.29$	-	$1.90 \pm 0.11$
$^{128}\text{Sn}$	59.1 min	$0.96 \pm 0.16$	$1.38 \pm 0.22$	-	$1.22 \pm 0.16$
$^{129}\text{Sb}$	4.4 h	$1.67 \pm 0.28$	$2.46 \pm 0.36$	$1.60 \pm 0.08$	$2.47 \pm 0.12$
$^{130}\text{Sb}$	39.5 min	$1.12 \pm 0.19$	$1.49 \pm 0.25$	$1.12 \pm 0.05$	$0.82 \pm 0.06$
$^{131}\text{Sb}$	23 min	$2.03 \pm 0.37$	$3.42 \pm 0.53$	$2.75 \pm 0.13$	$1.59 \pm 0.13$
$^{131\text{m}}\text{Te}$	1.3 d	$1.15 \pm 0.17$	$1.98 \pm 0.33$	-	-
$^{132}\text{Te}$	3.2 d	$4.57 \pm 0.80$	-	-	$4.98 \pm 0.26$
$^{132}\text{I}$	2.3 h	$4.89 \pm 0.86$	-	-	-
$^{133\text{m}}\text{Te}$	55.4 min	$3.18 \pm 0.55$	-	$4.21 \pm 0.20$	-
$^{134}\text{Te}$	41.8 min	$4.12 \pm 0.74$	$5.33 \pm 0.84$	$3.16 \pm 0.15$	$5.37 \pm 0.39$
$^{134}\text{I}$	52.5 min	$5.00 \pm 1.87$	$7.52 \pm 2.22$	-	-
$^{135}\text{I}$	6.6 h	$4.72 \pm 0.85$	$7.34 \pm 1.14$	$5.06 \pm 0.32$	$4.72 \pm 0.21$
$^{138}\text{Xe}$	14.1 min	$4.35 \pm 0.78$	$7.36 \pm 1.18$	$4.62 \pm 0.41$	-

<sup>138</sup> Cs	33.4 min	4.99 ± 1.80	7.79 ± 2.61	-	7.74 ± 0.33
<sup>141</sup> Ba	18.3 min	5.25 ± 0.88	6.92 ± 1.06	-	4.43 ± 0.27
<sup>142</sup> La	1.5 h	5.29 ± 0.92	6.39 ± 2.18	5.15 ± 0.24	5.98 ± 0.19

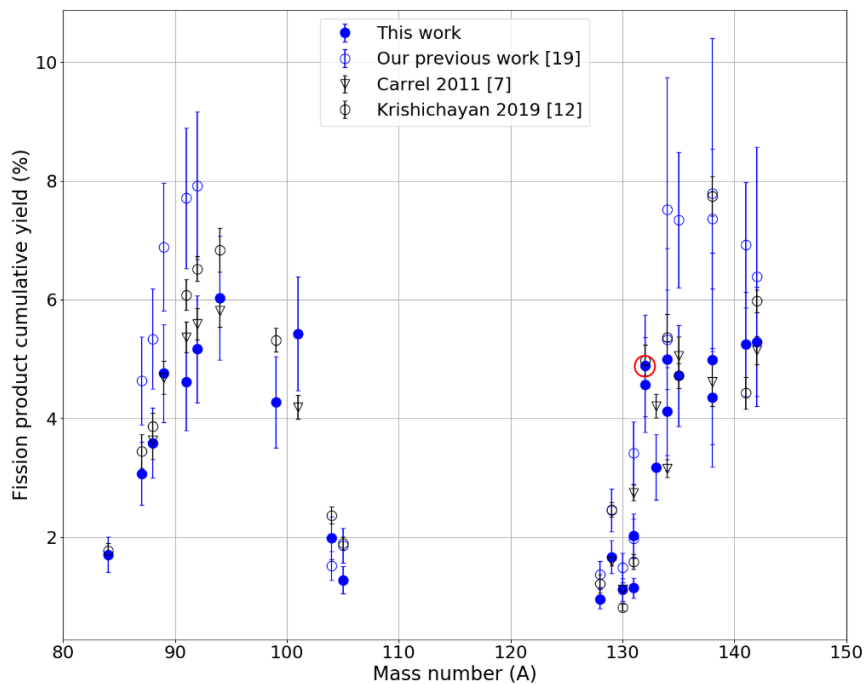
529  
530Table 12: Detailed data of the delayed gamma-ray analysis for each <sup>235</sup>U photofission product

Fission Product	Cooling time	Counting time	Gamma-ray energy (keV)	Relative intensity (%)	Net peak area (counts)	Photofission cumulative yield (%)
<sup>84</sup> Br	19.9 min	2.8 h	881.60	41.60	27836 ± 387	1.79
			2484.10	6.66	2322 ± 84	1.48
			3927.50	6.78	1765 ± 42	1.56
<sup>87</sup> Kr	5.8 min	7.5 h	402.59	49.60	178305 ± 914	3.09
			2554.80	9.23	16155 ± 149	2.96
			2558.10	3.92	7386 ± 116	3.20
			2011.88	2.88	5792 ± 182	2.95
<sup>88</sup> Kr	42 s	17.0 h	196.30	25.98	102192 ± 1261	3.46
			834.83	12.97	60791 ± 570	3.48
			1529.77	10.93	34211 ± 325	3.71
			2231.77	3.39	10623 ± 184	3.55
<sup>89</sup> Rb	16.9 min	1.8 h	1248.20	45.60	28161 ± 258	4.80
			2570.10	10.18	4169 ± 85	4.58
<sup>91</sup> Sr	5.8 min	23.8 h	1024.30	33.50	194351 ± 588	4.68
			749.80	23.68	146727 ± 674	4.47
			925.8	3.85	23247 ± 462	4.68
<sup>92</sup> Sr	42 s	17.0 h	1383.90	93.00	532609 ± 778	5.17
<sup>94</sup> Y	7.8 min	1.8 h	918.74	56.00	93343 ± 448	6.03
			1138.9	5.99	9275 ± 271	6.03
<sup>99</sup> Mo	9.8 min	23.8 h	739.50	12.12	21083 ± 571	4.24
			777.92	4.28	7828 ± 530	4.56
<sup>101</sup> Mo	42 s	1.4 h	1012.47	13.02	19380 ± 432	5.43
<sup>104</sup> Tc	5.8 min	2.1 h	358.00	89.00	67074 ± 763	2.00
			1596.70	4.18	1782 ± 188	1.71
			3149.20	1.16	334 ± 35	1.74
<sup>105</sup> Ru	46.2 min	23.2 h	724.30	47.30	85493 ± 557	1.28
			676.36	15.66	29663 ± 534	1.31
			316.44	11.12	23348 ± 897	1.32
<sup>128</sup> Sn	42 s	5.9 h	482.30	59.00	56919 ± 796	0.93
			680.50	15.93	16847 ± 590	1.12
<sup>129</sup> Sb	1.5 h	22.4 h	812.80	47.60	114447 ± 637	1.67
			544.70	18.09	49999 ± 766	1.71
<sup>130</sup> Sb	1.0 h	2.9 h	793.40	100.00	26825 ± 361	1.10
			839.52	100.00	26971 ± 346	1.13
<sup>131</sup> Sb	5.8 min	2.1 h	943.40	46.20	36721 ± 387	2.08
			933.10	25.87	18752 ± 366	1.87
<sup>131m</sup> Te	42 s	23.9 h	852.21	21.40	14450 ± 527	1.00
			1125.44	11.90	9653 ± 380	1.33
<sup>132</sup> Te	40.1 min	23.3 h	228.33	88.12	129579 ± 1055	4.57
<sup>132</sup> I	42 s	23.9 h	667.71	98.70	189532 ± 779	4.89
			522.65	15.99	32271 ± 782	4.87
			630.19	13.32	26635 ± 693	4.93
<sup>133m</sup> Te	42 s	8.0 h	912.67	55.27	151897 ± 584	3.13
			647.51	19.40	61265 ± 665	3.23
			863.96	15.64	43916 ± 520	3.14
			978.30	4.86	13696 ± 411	3.30
<sup>134</sup> Te	42 s	4.1 h	767.20	29.60	95959 ± 587	4.12
<sup>134</sup> I	42 s	9.4 h	884.09	65.08	413068 ± 789	5.00
			1072.55	14.93	88376 ± 471	5.00



			595.36	11.10	$78034 \pm 708$	4.87
$^{135}\text{I}$	42 s	23.9 h	1260.41	28.70	$176093 \pm 518$	4.80
			1131.51	22.59	$134550 \pm 516$	4.44
			1038.76	7.95	$48496 \pm 460$	4.43
			1678.03	9.56	$52684 \pm 322$	4.92
			1457.56	8.67	$52650 \pm 352$	5.09
$^{138}\text{Xe}$	42 s	1.4 h	1768.26	16.73	$15536 \pm 246$	4.34
			258.41	31.50	$43390 \pm 1181$	4.39
$^{138}\text{Cs}$	42 s	4.7 h	1435.86	76.30	$271157 \pm 573$	4.99
			1009.78	29.38	$122123 \pm 506$	5.03
			2218.00	15.18	$42828 \pm 255$	4.87
$^{141}\text{Ba}$	2.7 min	1.7 h	190.328	46.00	$62679 \pm 942$	4.98
			304.19	25.44	$54247 \pm 828$	5.37
			276.95	23.41	$48450 \pm 836$	5.43
			343.67	14.44	$30891 \pm 758$	5.28
$^{142}\text{La}$	1.1 h	8.7 h	641.29	47.4	$189460 \pm 632$	5.30
			894.90	8.34	$30424 \pm 362$	5.37
			1901.30	7.16	$18446 \pm 188$	5.17
			2187.20	3.70	$9598 \pm 146$	5.65
			2971.00	3.13	$5885 \pm 80$	4.97
			2055.20	2.18	$5271 \pm 136$	5.04

531 Our fission products cumulative yields for  $^{235}\text{U}$  are compared in Figure 15 to existing  
 532 published data.



533

534

Figure 15: Fission product yields distribution for the photofission of  $^{235}\text{U}$  and comparison with existing data.

535

536

537

Contrary to our previous work [19], we were not able to measure the cumulative yields of some short half-life photofission products (like  $^{93}\text{Sr}$  or  $^{142}\text{Ba}$ , with respective radioactive periods of 7.4 min and 10.6 min) because of the dead time issues mentioned in section 2.

538 However, we measured new yields ( $^{84}\text{Br}$ ,  $^{91\text{m}}\text{Y}$ ,  $^{99}\text{Mo}$ ,  $^{101}\text{Mo}$ ,  $^{132}\text{Te}$ ,  $^{132}\text{I}$ ,  $^{133\text{m}}\text{Te}$ ) that can  
539 be compared with existing data. We also provide the  $^{235}\text{U}$  cumulative yield for  $^{132}\text{I}$  (circled  
540 in Figure 15), which was not reported so far in prior publications. For the others, the  
541 cumulative yields obtained in this work are consistent with existing data. In particular, we  
542 observe a better agreement with Carrel *et al.* [7] than in our previous work. The prior  
543 discrepancy was probably due to the poor knowledge of the geometry of the low-mass  
544 HEU sample used in the previous experimental campaign [19]. The HEU sample used in  
545 this work has a simpler geometry, i.e., a single fissile core instead of several cores separated  
546 by Zircaloy spacers.

## 547 **6. Differentiation of actinide isotopes**

548 As photofission cross sections are of the same order of magnitude for all actinides, the  
549 measured delayed gamma-ray signal can provide an estimation of the total nuclear material  
550 mass. However, it does not indicate whether it is uranium or plutonium (with very different  
551 specific activities, in Bq/g, which is essential for waste management especially for the long-  
552 term alpha activity [34]), fissile or fertile isotopes (for criticality safety purpose in waste  
553 package transport, interim storage, or final repository). This section deals with the  
554 possibility to differentiate actinides using delayed gamma-ray ratios of their photofission  
555 products.

556 When photofission occurs, the formation of two asymmetric fission fragments is the  
557 most likely to happen, resulting in a mass distribution curve of the fission products  
558 comporting two bumps: one for a heavy nucleus centered around mass number 140, and  
559 one for a light nucleus centered around 95. Examples of this theoretical mass distribution  
560 for photofission products are given in the work of Bernard *et al.* [22] and reported in Figure  
561 16 for several actinides.

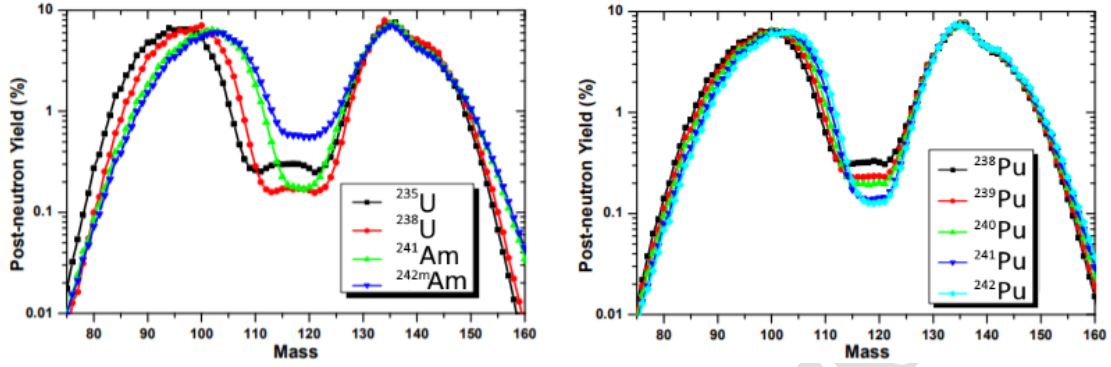


Figure 16: Independent photofission mass yields obtained with the GEF code [35], as reported in ref. [22].

A differentiation information can be obtained between uranium or plutonium isotopes thanks to their different photofission product yields, through the measurement of the relative intensities of specific delayed gamma lines emitted by photofission products having different yields for the different actinides [1][17]. The enrichment, defined as the proportion of the actinide(s) of interest in the mixture, can thus be computed from the ratio of two net areas and from the ratios of the photofission yields of their two emitting photofission products. The objective is to select specific photofission products pairs showing the largest differences, according to the actinide, in their gamma ratios. Some important parameters such as the energy of the gamma rays emitted by the photofission products (related to possible interferences with gamma rays of activation products, matrix attenuation effects, and the level of the Compton continuum under the peaks) and their intensity that determines the achievable counting statistics will also need to be taken into account to identify the most efficient photofission product couples.

For a mixture of  $^{235}\text{U}$  and  $^{238}\text{U}$ , the net peak area of a delayed gamma-ray line of energy  $E_i$  emitted by photofission product  $j$  is given by (5):

$$A_j(E_i) = \frac{I(E_i)\varepsilon(E_i)N_A}{\lambda_j} \times (1 - e^{-\lambda_j t_{irr}}) \times e^{-\lambda_j t_{cool}} \times (1 - e^{-\lambda_j t_{count}}) \times \dots \left[ Y_{j5} \frac{\alpha m_u}{\mathcal{M}_5} \int_{E_{threshold}}^{E_{max}} \varphi(E) \sigma_5(E) dE + Y_{j8} \frac{(1 - \alpha) m_u}{\mathcal{M}_8} \int_{E_{threshold}}^{E_{max}} \varphi(E) \sigma_8(E) dE \right] \quad (5)$$

Where:

- $A_j(E_i)$  is the net peak area of the gamma-ray line of energy  $E_i$  emitted by photofission product  $j$ ;
- $I(E_i)$  is the gamma-ray emission intensity;

- 583 -  $\varepsilon(E_i)$  is the detection efficiency at energy  $E_i$ ;  
 584 -  $N_A$  is the Avogadro constant (in  $\text{mol}^{-1}$ );  
 585 -  $\lambda_j$  is the radioactive decay constant of photofission product  $j$  (in  $\text{s}^{-1}$ );  
 586 -  $t_{irr}$ ,  $t_{cool}$  and  $t_{count}$  are respectively the irradiation, cooling and counting times  
 587 (in s);  
 588 -  $Y_{j5}$  and  $Y_{j8}$  are the cumulative yields of photofission product  $j$  for  $^{235}\text{U}$  and  $^{238}\text{U}$ ,  
 589 respectively;  
 590 -  $m_u$  is the total uranium mass (in g);  
 591 -  $\mathcal{M}_5$  and  $\mathcal{M}_8$  are respectively the molar masses of  $^{235}\text{U}$  and  $^{238}\text{U}$  (in  $\text{g}\cdot\text{mol}^{-1}$ );  
 592 -  $\alpha$  is the enrichment, i.e. the proportion of  $^{235}\text{U}$  in the uranium mixture;  
 593 -  $E_{threshold}$  is the threshold energy of the photofission reaction;  
 594 -  $E_{max}$  is the endpoint energy of the Bremsstrahlung photon beam;  
 595 -  $\varphi(E)$  is the Bremsstrahlung photon flux at energy  $E$  (in  $\text{cm}^{-2}\cdot\text{s}^{-1}$ );  
 596 -  $\sigma_5(E)$  and  $\sigma_8(E)$  are the photofission cross sections (in  $\text{cm}^2$ ) at energy  $E$  for  $^{235}\text{U}$   
 597 and  $^{238}\text{U}$ , respectively.

598  
 599 (5) can be rearranged and written as (6):

$$A_j(E_i) = k_j(E_i) \times m_u \times [Y_{j8}(\alpha - 1) - \Sigma \alpha Y_{j5}] \quad (6)$$

600 With:

- 601 -  $k_j(E_i)$  a constant defined for the sake of simplification, which depends on the  
 602 photofission product radioactive constant, the gamma-ray intensity and detection  
 603 efficiency, and the measurement time parameters (irradiation, cooling, counting):

$$604 \quad k_j(E_i) = \frac{I(E_i)\varepsilon(E_i)N_A}{\lambda_j} \times (1 - e^{-\lambda_j t_{irr}}) \times e^{-\lambda_j t_{cool}} \times (1 - e^{-\lambda_j t_{count}}) \text{ (in } \text{s}^{-1}\cdot\text{mol}^{-1}\text{);}$$

- 605 -  $\Sigma$  another simplification constant depending on the actinide mixture, defined as:

$$606 \quad \Sigma = \frac{\frac{1}{\mathcal{M}_5} \int_{E_{threshold}}^{E_{max}} \varphi(E) \sigma_5(E) dE}{\frac{1}{\mathcal{M}_8} \int_{E_{threshold}}^{E_{max}} \varphi(E) \sigma_8(E) dE} \approx \frac{\int_{E_{threshold}}^{E_{max}} \varphi(E) \sigma_5(E) dE}{\int_{E_{threshold}}^{E_{max}} \varphi(E) \sigma_8(E) dE}$$

607

608 Then, the ratio of the net areas of two delayed gamma rays emitted by two different  
 609 photofission products is given by (7):

$$R = \frac{k_1}{k_2} \times \frac{Y_{18}(\alpha - 1) - \Sigma\alpha Y_{15}}{Y_{28}(\alpha - 1) - \Sigma\alpha Y_{25}} = \frac{k_1}{k_2} \times R' \quad (7)$$

610 Where:

- 611 -  $k_1$  and  $k_2$  correspond to constants  $k_j(E_i)$  described in (6). It is important to
- 612 mention that we are not limited to ratios of close-in-energy gamma rays. This
- 613 implies that detection efficiency at both energies has to be estimated and requires
- 614 a prior localization of actinides, for example with the method described in [18];
- 615 -  $\alpha$  and  $\Sigma$  have the same meaning as in (5) and (6);
- 616 -  $Y_{18}$  and  $Y_{15}$  are the cumulative yields for the first photofission product for  $^{238}\text{U}$  and
- 617  $^{235}\text{U}$ , respectively;
- 618 -  $Y_{28}$  and  $Y_{25}$  are the cumulative yields for the second photofission product for  $^{238}\text{U}$
- 619 and  $^{235}\text{U}$ , respectively;
- 620 -  $R'$  is the net area ratio R corrected by the ratio of constants  $k_1$  and  $k_2$  taking into
- 621 account gamma-ray intensities and detection efficiencies of the two photofission
- 622 product gamma rays, and irradiation parameters of the two photofission products
- 623 (exponential terms in (5)).

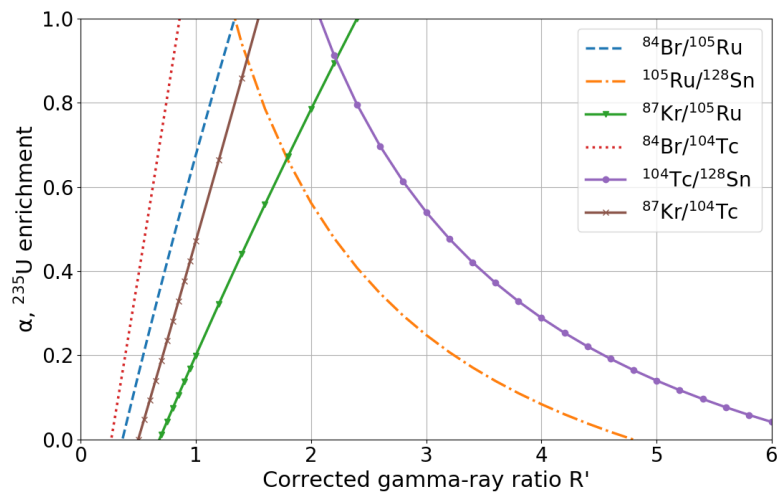
624

625 The efficiency of this differentiation method can be assessed by defining a criterion  $\delta$   
 626 based on the variability of the calculated enrichment compared to the variability of the  
 627 corrected gamma-ray ratio (8).

$$\delta = \frac{d\alpha}{dR'} \frac{R'}{\alpha} = \frac{\Sigma(Y_{15}Y_{28} - Y_{25}Y_{18})}{\left[ \left( \frac{Y_{18}(\alpha - 1) - \Sigma\alpha Y_{15}}{Y_{28}(\alpha - 1) - \Sigma\alpha Y_{25}} \right) (Y_{28} - \Sigma Y_{25}) - Y_{18} + \Sigma Y_{15} \right]^2} \cdot \frac{Y_{18} \frac{\alpha - 1}{\alpha} - \Sigma Y_{15}}{Y_{28}(\alpha - 1) - \Sigma\alpha Y_{25}} \quad (8)$$

628 By minimizing this criterion over the whole enrichment range, the most appropriate  
 629 photofission product couples can be identified based on their cumulative photofission  
 630 product yields reported in previous section for  $^{235}\text{U}$  and  $^{238}\text{U}$ . The six most efficient  
 631 photofission product couples for  $^{235}\text{U}$  vs.  $^{238}\text{U}$  discrimination are thus  $^{84}\text{Br}/^{105}\text{Ru}$ ,  
 632  $^{105}\text{Ru}/^{128}\text{Sn}$ ,  $^{87}\text{Kr}/^{105}\text{Ru}$ ,  $^{84}\text{Br}/^{104}\text{Tc}$ ,  $^{104}\text{Tc}/^{128}\text{Sn}$  and  $^{87}\text{Kr}/^{104}\text{Tc}$ . The curves representing the  
 633  $^{235}\text{U}$  enrichment as a function of the  $R'$  corrected ratio (deduced from the R measured ratio)  
 634 are provided in Figure 17, showing that it seems possible to estimate  $\alpha$  (or at least the

635 enrichment range) with the different couples if  $R'$  is measured with a reasonably low  
 636 uncertainty, which will mainly depend on counting statistics uncertainties of the delayed  
 637 gamma-ray net areas. For instance, a 20 % uncertainty on  $R'$  will convert to a relative  
 638 uncertainty of 65 % on the  $^{235}\text{U}$  enrichment,  $\alpha$ , for the couple  $^{87}\text{Kr}/^{105}\text{Ru}$  and a limited  
 639 enrichment ( $\alpha=0.2$ ). Besides, a 20 % uncertainty on  $R'$  for the couple  $^{84}\text{Br}/^{105}\text{Ru}$   
 640 corresponds to a relative uncertainty of 30 % on  $\alpha$  for a higher enrichment ( $\alpha=0.6$ ). A  
 641 combination of the results, such as a weighted average of the enrichments obtained with  
 642 the different couples, will probably be valuable. Further investigation will be conducted to  
 643 demonstrate the differentiation efficiency of these photofission product couples on real  
 644 actinide mixtures placed inside a matrix.



645

646 *Figure 17: Evolution of  $^{235}\text{U}$  enrichment as a function of  $R'$  corrected ratio for the 6 most efficient photofission product*  
 647 *couples in a  $^{235}\text{U}/^{238}\text{U}$  mixture.*

648 Beyond a simple uranium mixture only, the photofission product yield values show that a  
 649 differentiation of the fertile isotope ( $^{238}\text{U}$ ) and the fissile ones ( $^{235}\text{U}$  and  $^{239}\text{Pu}$  considered  
 650 as a whole), could be achieved since the photofission cross-sections of  $^{235}\text{U}$  and  $^{239}\text{Pu}$  are  
 651 similar and that of  $^{238}\text{U}$  is way different, as shown in Figure 18. Indeed, there is only a 21%  
 652 difference between the photofission rate in  $^{239}\text{Pu}$  and  $^{235}\text{U}$  for the 17.5 MeV endpoint  
 653 energy Bremsstrahlung spectrum presented in this paper.

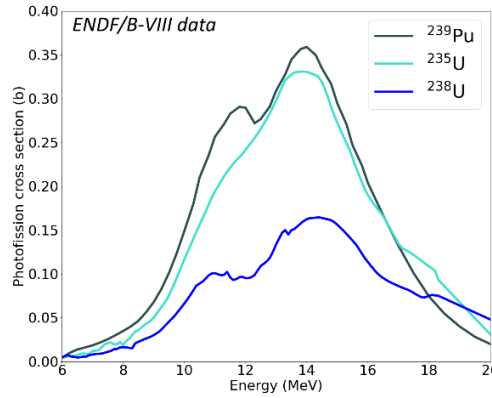


Figure 18: Photofission cross sections of  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{239}\text{Pu}$  [23].

654

655

656 The most efficient photofission product couple for that purpose is  $^{128}\text{Sn}/^{134}\text{Te}$ . The two

657 isotopes share a similar ratio of cumulative yields for  $^{235}\text{U}$  ( $\frac{Y_c(^{128}\text{Sn})}{Y_c(^{134}\text{Te})} = 0.23 \pm 0.06$ ) and

658  $^{239}\text{Pu}$  ( $\frac{Y_c(^{128}\text{Sn})}{Y_c(^{134}\text{Te})} = 0.23 \pm 0.05$ ), and a different ratio for  $^{238}\text{U}$  ( $\frac{Y_c(^{128}\text{Sn})}{Y_c(^{134}\text{Te})} = 0.10 \pm 0.02$ ).

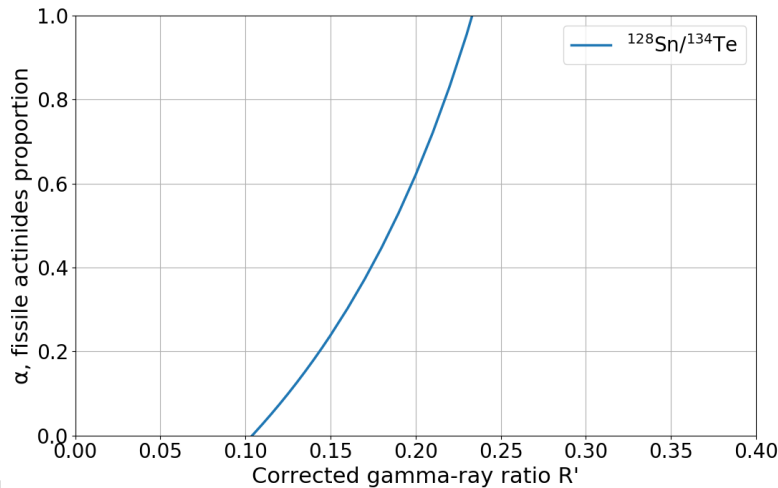
659 Therefore,  $^{235}\text{U}$  and  $^{239}\text{Pu}$  can be regarded as a global fissile mass that can be differentiated

660 from  $^{238}\text{U}$  mass. The curve representing the fissile actinides proportion as a function of the

661  $R'$  corrected ratio (deduced from the  $R$  measured ratio) is provided in Figure 19. Note that

662 the contrast for the differentiation between  $^{235}\text{U}+^{239}\text{Pu}$  and  $^{238}\text{U}$  is lower compared to the

663 differentiation between  $^{235}\text{U}$  and  $^{238}\text{U}$  in Figure 17.



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Figure 19: Evolution of the fissile actinides proportion in a  $^{235}\text{U}/^{239}\text{Pu}/^{238}\text{U}$  mixture as a function of  $R'$  corrected ratio for the couple  $^{128}\text{Sn}/^{134}\text{Te}$

667 It is worth noting that such a global fissile vs. fertile actinides differentiation based on

668 photofission had not been proposed yet, as the yields of these fission products reported in

669 the literature are scarce for  $^{235}\text{U}$  or  $^{239}\text{Pu}$ , for instance only one measured yield was reported  
670 so far concerning  $^{128}\text{Sn}$  and  $^{134}\text{Te}$  photofission products of  $^{239}\text{Pu}$ .

## 671 7. Conclusion

672 New measurements of cumulative photofission yields with a 17.5 MeV endpoint  
673 Bremsstrahlung photon beam produced by a SATURNE electron LINAC have been  
674 performed in CINPHONIE facility, at CEA Cadarache IRESNE Institute, France. To this  
675 aim, a characterization of the Bremsstrahlung photon beam has been first carried out by  
676 photon activation analysis with different samples of Au, Ni, U, Zn and Zr. In a former  
677 study, we highlighted that most of neutron fissions arise from fast neutrons produced in the  
678 actinide sample itself, but not from the photoneutron production in the LINAC components  
679 (target, collimator). As a result, the neutron fission rate in the different samples has been  
680 numerically estimated with MCNP to subtract it from the total fission rate. Finally, the  
681 cumulative production yields of 26 photofission products have been measured for  $^{239}\text{Pu}$   
682 and  $^{235}\text{U}$  and 28 have been evaluated for  $^{238}\text{U}$ . Four of them are not reported in the literature  
683 for  $^{239}\text{Pu}$ , and one for  $^{235}\text{U}$ . Among these available photofission product yields, some show  
684 large discrepancies between actinides and thus appear as good candidates for their  
685 differentiation based on gamma-ray ratios. To that extent, the six most efficient couples  
686 enabling the differentiation between  $^{235}\text{U}$  and  $^{238}\text{U}$  have been determined. Furthermore, a  
687 photofission products couple ( $^{128}\text{Sn}/^{134}\text{Te}$ ) has been identified for the differentiation  
688 between fissile ( $^{235}\text{U}+^{239}\text{Pu}$ ) and fertile ( $^{238}\text{U}$ ) isotopes in a mixture of uranium and  
689 plutonium.

690 Further work will be dedicated to the selection of the most efficient photofission product  
691 couples in presence of a waste matrix, causing gamma attenuation effects that depend on  
692 actinide localization. To this aim, a new experimental campaign will be carried out to test  
693 the differentiation of actinide isotopes inside a concrete matrix, as reported in [18] for  
694 actinide localization. One of the main objectives of this long-term R&D program is to  
695 assess nuclear materials in heterogeneous technological waste blocked in 870 L cemented  
696 drums. Therefore, the quantification of their mass and the differentiation of actinides will  
697 be finally tested with an 870 L mock-up drum with uranium and plutonium samples inside.



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Highlights

- Photofission is a promising technique to assay nuclear waste packages
- Photon activation analysis is used to characterize a 17.5 MeV Bremsstrahlung beam
- Photofission products cumulative yields have been determined for  $^{239}\text{Pu}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$
- The contribution of neutron fission has been estimated with the MCNP code
- Photofission product yield difference makes actinides differentiation practicable

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CRedit author statement:

**M. Delarue:** Methodology, Investigation, Software, Writing – Original Draft, Visualization, Formal analysis

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**Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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