EPJ Web of Conferences **146**, 09027 (2017) DOI: 10.1051/epjconf/201714609027
ND2016

C-BORD project: "Effective container inspection at BORDer control points"

Development of an identification method of actinides in containers

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Abstract. In the framework of the C-BORD project of the H2020 programme to provide a safety chain for container inspection at border control points, CEA-DAM is involved to develop an identification method of Special Nuclear Material. Using active interrogation the main purpose is to specifically discriminate fissile actinides from fertile ones through the detection of γ-rays emitted by very short-lived isomers. We present here the interdependance of such a project with nuclear data. We first introduce the SNM identification method. Then, will be detailed the needs in nuclear data and first yields of specific short-lived isomers of ^{252}Cf spontaneous fission. Perspectives and conclusions will end the article.

1. Introduction

In European Union, the statement that has been done is that security at borders is of the most importance for all the country members. In this context the C-BORD project has been initiated to develop efficient non-intrusive inspection (NII) technologies and test a comprehensive cost-effective TRL-7 solution for the generalised inspection of containers, coping with a large range of container targets, including explosives, chemical warfare agents, illicit drugs, stowaways and Special Nuclear Material (SNM). In this framework, CEA-DAM is involved to develop a method for SNM identification using photofission. The method is based on the detection of γ-rays emitted by very short-lived isomers with lifetimes below 3 μs. This active non-destructive method is used as a second-line inspection when passive inspection methods have reached their limits, if there is a suspicion of SNM presence in a container we must provide a method to discriminate fissile actinides from fertile ones. Photofission will be induced with the already working LINAC Varian Linatron M9® used for X-ray imaging by customs. In this method observables are yields of isomers (^{134m,135m}Te and ^{136m}Xe) whose delayed γ-rays are detected during interpulses measurements of pulsed beam of photons. Isomer yields are nuclear data challenging to obtain. Their knowledge goes beyond borders security applications, since they will play a key role in nuclear power plant safety [1], and in the constraint and improvement of existing Monte-Carlo fission particles emission codes [2,3].

2. SNM identification method

Based on the existing 9 MeV LINAC a work-package "Technology sub-system photofission" is developed to be coupled to high energy imaging techniques for better NII results. In a two steps approach (irradiation that will induce photofission reactions followed by a detection phase), two aims have to be achieved within this work-package: the detection of the presence (or absence) of SNM inside a container and its identification i.e., discrimination between fissile and fertile actinides (^{235}U and ^{239}Pu compared to ^{238}U). Imposed by legal constraints on safety regulation the LINAC provides a pulsed beam of end-point Bremsstrahlung photons with energies of 9 MeV. These photons have a distribution peaked at 0.5 MeV and rapidly decreasing. Photons with sufficient energy ($E_\gamma > 6$ MeV) will induce photofission reactions with cross sections of tens of millibarns. The low cross section values are compensated with the high intensity of the beam ($\sim$100 mA). The accumultation of these specific working conditions implies several constraints in physics, detection and the needed nuclear data set.

We describe here the SNM identification method whose principle is based on the interpulses detection of three spectific delayed γ-rays emitted by isomeric fission fragments produced during the photofission process. Chosen for their specific lifetimes, intensities and energies these candidates are presented in Table 1:

- $T_{1/2} > 100$ ns limits the background due to the beam
- $T_{1/2} < 3\,\mu$s allows a better signal-to-background ratio due to a more intense activity compared to a longer-living isomer in a short measurement time
- $E_\gamma > 1$ MeV limits the photon absorption by different material.

We focus on isomeric levels of ^{134,135}Te and ^{136}Xe, ones of the most produced isotopes in the fission process ($\sim 10^{-2}$/fission). Their γ-rays will be easily identifiable on the γ-spectrum acquired during repeated counting times of few μs. Moreover, working with ratios and γ-rays in the same energy range allow us to get rid of geometrical corrections and efficiency corrections. Their short half-lives

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Table 1. Fission fragments characteristics.

Isomer	^{134m}Te	^{135m}Te	^{136m}Xe
$T_{1/2}$ (ns)	164.1(9)	511(20)	2950(90)
Isomer E (keV)	1691	1555	1892
Isomer J^{π}	6^+	$\frac{19}{2}^-$	6^+
ground state spin	0^+	$\frac{7}{2}^-$	0^+
γ of interest (keV)	1279	1180	1313

Table 2. ratio of isotope yields in n-induced fission at 1 MeV [4,5].

n-induced fission at 1 MeV	^{234}U(n,f)	^{237}U(n,f)	^{238}Pu(n,f)
equivalent photofission at 9 MeV	^{235}U(γ,f)	^{238}U(γ,f)	^{239}Pu(γ,f)
$\frac{Y(^{134}Te)}{Y(^{136}Xe)}$	1.64	15.39	1.27
$\frac{Y(^{135}Te)}{Y(^{136}Xe)}$	0.86	15.46	0.64

allow a time and energy identification to meet the requirements of a quick and reliable analysis.

The observables allowing actinide discrimination are yield ratios of these isomers: $\frac{Y(^{134m}Te)}{Y(^{136m}Xe)}$ and $\frac{Y(^{135m}Te)}{Y(^{136m}Xe)}$. Nuclear data of interest are isomer yields in actinides. Data on thermal neutron induced fission were obtained by Sund et al. [8] on ^{235}U and ^{239}Pu. These data show differences between isotopic and isomeric yield ratios and between actinides. In this case, the fissioning systems are ^{236}U and ^{240}Pu. In photofission we need to study the ^{235}U and ^{239}Pu fissioning systems in the reactions presented in Table 2. Unfortunately there is only few data available concerning the production of these isomers in neutron-induced fission, ^{234}U(n,f) and ^{238}Pu(n,f), and nothing exist for the isomers in photofission. Few measurements and evaluations show that isotopic yield ratios differ according to the fissioning actinide. Different works [3,6,7] support the hypothesis that the isomeric ratio is weakly dependant on the fissioning system and the projectile energy which means that the large enough difference between the isotope yields in actinide fissions may also be found in the isomer yields. Finally, for our application we need to measure isomer yield ratios in photofission at 9 MeV with a $LaBr_3$(Ce) detector (for its good time and energy resolutions).

The setting-up of the identification method will follow three axes: verify the method feasability in terms of detection and analysis, obtain realiable nuclear data for applications and guarantee a use in operational conditions. The first step has been initiated with the study of the ^{252}Cf spontaneous fission (SF) presented in the following Sect. 3.

3. Thorough study of ^{134m,135m}Te and ^{136m}Xe production in ^{252}Cf (SF)

In order to validate the method and start an isomer yields database, the first experiment concerned the study of ^{252}Cf (SF). Additionnaly, we extracted from these data the isomer yields of interest and compared them to previously published experimental results and Monte-Carlo calculations performed by O. Litaize with the

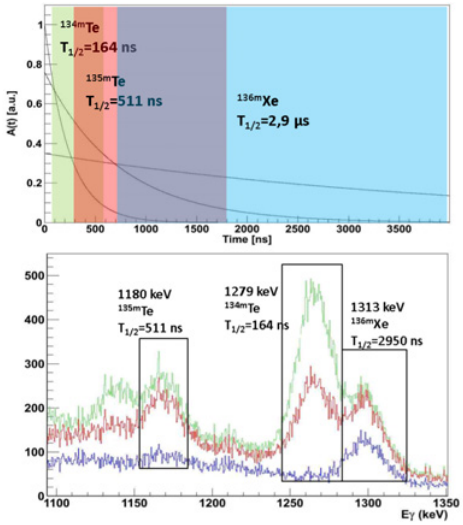

Figure 1. Top: isomer activities normalized to 1 as a function of time and coincidence window. Bottom: $LaBr_3$:Ce γ-energy spectra zoomed on the region of interest 1180 keV (^{135m}Te), 1279 keV (^{134m}Te) and 1313 keV (^{136m}Xe).

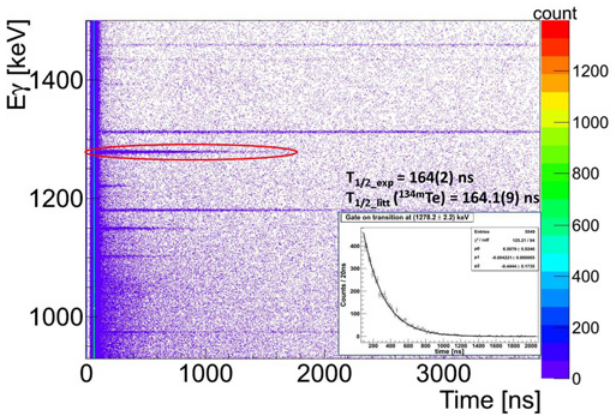

Figure 2. HPGe time-energy matrix. Example of the 1279 keV time decay.

FIFRELIN code (presented in Sect. 5).

A fission chamber with a 500 μm thick ^{252}Cf deposit on a carbon backing was used as a trigger for the data acquisition system. The fission chamber is composed of 2 mm thin Al walls, filled with 0.5 bar of Ar gas. γ-rays were detected in coincidence with a fission chamber event (α-decay or fission) with two detectors placed at 90°: a $LaBr_3$:Ce detector (chosen for its good compromise of energy and time resolution) to validate this kind of detector in such experiment and a HPGe for fine spectroscopy. Events (time and energy) are recorded for 4 μs after the fission chamber trigger. After time and energy identification (Figs. 1 and 2) of the three γ-rays of interest, the peak areas were calculated and corrected from detection efficiency, counting time, cooling time, branching ratio and gamma attenuation in aluminium. The number of fissions determined from the analysis of the fission chamber events permits to extract the yields of the three isomers of interest ^{134m,135m}Te and ^{136m}Xe. The isomer yields and ratios are compared to those of the references [8], [9] and [10] and are presented in Table 3

Table 3. Isomer yields experimental results.

γ-ray (keV)	Prod. yield $\times 10^{-2}$ this work	Sund [8]	John [9]	Clarck [10]
^{134m}Te				
297	1.10(4)	1.50(20)	1.03(7)	1.016(51)
1279	1.14(4)	1.14(16)	1.26(11)	1.01(16)
^{135m}Te				
325	0.31(6)	0.42(19)	0.31(3)	0.268(19)
1180	0.31(6)	0.29(13)	0.30(4)	0.286(31)
^{136m}Xe				
197	0.46(16)	0.92(24)	0.60(5)	0.300(15)
381	0.57(2)	0.86(29)	0.73(6)	0.449(?)
1313	0.61(4)	0.86(44)	0.57(14)	0.490(44)

Table 4. Isomer yield ratios experimental results.

Ratio	this work	Sund [8]	John [9]	Clarck [10]
$\frac{Y(^{134m}Te)}{Y(^{136m}Xe)}$	1.92(14)	1.33(70)	2.21(58)	2.06(38)
$\frac{Y(^{135m}Te)}{Y(^{136m}Xe)}$	0.53(10)	0.34(23)	0.53(15)	0.58(8)

and Table 4. One can notice the good agreement between the different measurements.

One can notice that the evaluated isotope yield of these nuclei in ^{252}Cf (SF) [4] ($Y(^{134}Te) = 0.0237(38)$, $Y(^{135}Te) = 0.0180(57)$, $Y(^{136}Xe) = 0.0082(37)$) differ from the experimental isomer yield, respectively, by a factor 1.2, 5.8 and 1.3.

The structure effects in the yields are more important than expected. As expected the structure effect linked to the isomer is found in the yield. Also, this experiment confirmed, that a $LaBr_3$:Ce detector presents the adequate qualities for this kind of measurement and that the isomer yield ratios have to be measured.

4. Nuclear data: Isomer yields

Neutron-induced fission have been quite investigated due to its importance in nuclear energy and safety associated. Neutron-induced fissions present also the advantage of larger cross sections compared to the photofission ones. Although, many nuclear data are available for (n,f) reactions allowing very well furnished evaluated databases, efforts have to be continued regarding experimental values of isomer yields.

As mentionned in the introduction, few data concerning photofission of SNM (at 9 MeV) are available but efforts are and must be undertaken in order to overcome this lack. Photofission data do exist but in a scattered manner. In the last decades, most important efforts have been done to measure photofission cross sections of several actinides on a large energy range [11]. Concerning the nuclear data of interest i.e., fission fragment yields in actinide photofission the most accomplished work has been realised by De Frenne and co-workers on charge and isotopic distributions and elemental yields for the photofission of ^{235}U and ^{238}U [12], [13]. To this date, there exist some data concerning isomeric ratios obtained in fission from a Bremsstrahlung beam with an end-point energy ranging from 8.5 MeV to 70 MeV for various actinides (^{233,235,238}U, ^{232}Th, ^{237}Np or ^{239}Pu). Due to experimental difficulties to have a clear signal in such a γ-background, studied nuclei only concerned the long living ones ($T_{\frac{1}{2}}$ > minute).

From the theoritical point of view, Monte Carlo simulation for particle emissions in fission codes have been developed, such as CGMF [14], FREYA [15], FIFRELIN [16]. These new developments bring complementary informations concerning the fission process and nuclear data of prompt neutron and gamma, but do not reproduce all of the experimental results and are still being ameliorated.

Hence, concerning the nuclear data of interest for the presented application and the difficulties to obtain them, two approaches are possible: direct measurements of the yields (ratios) of the isomers of interest in (γ,f) reactions or obtaining data through simulations with reliable models evaluated on experimental yields results from alternative experiments such as spontaneous fission, neutron-induced fission or surrogate reactions.

5. Experimental and theoritical efforts

A detailed study was conducted on the specific case of ^{134m}Te production in ^{252}Cf (SF). From cold fission measurements [17] and the results presented Sect. 3, it seems that in the spontaneous fission process of ^{252}Cf, an important part of ^{134}Te is produced after neutron emission, below the 6^+ (at 1.691 MeV, $T_{\frac{1}{2}} = 164$ ns) isomeric level (on the 4^+, 2^+ or 0^+ state). We compared experimental results with the fission modelling calculation code FIFRELIN [16]. The calculations of the spontaneous fission of ^{252}Cf were performed by O. Litaize (CEA, Cadarache, France). The computed values of ground state yields of ^{134}Te, ^{135}Te and ^{136}Xe are slightly above the evaluated ones but within the error bars. On the other hand the isomer yield values were positively above the experimental ones. The fission fragments decays are based on the RIPL-3 level schemes which do not offer ^{134}Te* many alternatives to decay to the ground state by-passing through the isomeric level. The study of the spin distributions of ^{134}Te* after neutron evaporation shows that spins of excited nuclei are too high which explains the overfeeding of two isomeric states (12^+ at 5.804 MeV, $T_{\frac{1}{2}} = 18$ ns and 6^+ at 1.691 MeV, $T_{\frac{1}{2}} = 164$ ns).

Although in Hauser-Feshbach simulations of the prompt neutron and gamma emission from fission fragments such as FIFRELIN or CGMF the average multiplicities of prompt particles are in good agreement with experimental data, it is not always the case when it comes to isomer yields of fission fragments as we can see with the overfeeding of some isomeric levels in ^{134,135}Te and ^{136}Xe in FIFRELIN or in [2]. Indeed, initial spin distribution in the fragments has still to be constrained since it controls the competition between neutron and photon emission. This is why, nuclear data on prompt γ-rays (such as short-lived yields and isomeric ratios) would help in constraining the initial angular momentum distribution of fission fragments.

Getting such experimental data is quite challenging, nevertheless experiments have been realized and planned in order to fulfil this nuclear data need. Concerning (n,f) reactions, an experiment on the 4 MV accelerator will be conducted at CEA DAM in collaboration with Los Alamos National Laboratory. The purpose of this experiment is

the study of isomer in the ^{235}U(n,f) reaction to determine isomer yields. A ^{3}He transfer reaction on ^{238}U performed at the Tandem accelerator (ALTO facility, Orsay, France) will be used to study isomer yield ratios of interest as a function of fissioning nucleus and its excitation energy.

For (γ,f) reactions, experimental conditions being more difficult, experimental programs are being set up. One of the first experiments will be performed at the linear electron accelerator ELSA [18] (CEA/DAM, Bruyères-le-Châtel, France) which produces a Bremsstrahlung radiation (up to 18 MeV) through the interaction of high energy electrons on a Ta target. A research program on photofission yields of SNM has also been initiated in the United States of America [19].

In addition to experiments for nuclear data collection, efforts must also be done concerning technical developments for real conditions measurements. R&D on the detection device has been done in the past months. In order to have an operational detector $\sim$100 ns after the γ-flash we use the "gating function". That is to say we control the electrical potential on a dynode in the photomultiplier tube (coupled to a $LaBr_3$:Ce crystal) blocking the light collection on the anode. The voltage switch of the gating module is fast ($\sim$100–120 ns), consequences are oscillations on the detector signal during few hundred nanoseconds. Consequently the signal cannot be treated with classical analogue electronics, thus, we use a numerical acquisition system. A waveform analysis program has been created to treat detector signal acquired during few μs to few tens of μs: this program takes into account the oscillations due to the gate opening (or closing depending of the working mode of the blocking function) and pile-up events to reconstruct time and energy spectra. The whole acquisition system has been tested and validated with high intensity γ sources during an experiment at CEA LIST (Saclay, France).

Experiments in working conditions close to those of Rotterdam customs (where final trials will take place) are planned at ELSA facility (its versatility allows us to have a beam as intense as X-ray scanners used at border control points) and at SAPHIR facility (CEA LIST, Saclay, France) where samples of SNM will be irradiated with a Varian Linatron M9.

Nowadays experimental and theoretical efforts are being made in order to compensate the lack of nuclear data on short-lived fission fragments. Experimental research program on photofission are still rare and difficult, it is important to study alternative solutions for (n,f) and surrogate reactions experiments.

6. Conclusion

The main assesment is that short-lived isotope yields measurements in photofission experiments is challenging, this is why there is no nuclear data but many efforts must be done in order to fulfil this need.

An operational detection device is now available for measurements from $\sim$100 ns after the beam pulse, associated with a fission trigger mechanism and adequate working conditions (shielding, nuclear material quantities, etc.) we should be able to measure isomer yield ratios in photofission during the upcoming months.

Isomeric ratios or isomer yields in fission fragments are good means to test and improve angular momentum distribution in fission models.

The authors would like to thank O. Litaize (CEA, Cadarache, France) for providing the FIFRELIN calculations. The project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 653323. This text reflects only the author's views and the Commission is not liable for any use that may be made of the information contained therein.

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