DISCLAIMER

Ternary Fission of ^{249}Cf(n,f) and ^{250}Cf(SF)

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Abstract. During the last years, several Cm and Cf isotopes have been studied by our research group in the frame of a systematic investigation of gas emission characteristics in ternary fission. Here we report on new results on the energy distribution and the emission probability of ^{3}H, ^{4}He and ^{6}He particles emitted in the spontaneous ternary fission of ^{250}Cf (E_{exc} = 0 MeV) and in the neutron induced ternary fission of ^{249}Cf (E_{exc} = 6.625 MeV). Both measurements were performed using suited and well-calibrated ΔE-E telescope detectors, at the IRMM (Geel, Belgium) for the spontaneous fission and at the very intense neutron beam PF1b at the Institute Laue-Langevin (Grenoble, France) for the neutron induced fission measurement. In this way, the existing database can be enlarged with new results for Z=98 isotopes, which is important for the systematic investigation. Moreover, the investigation of the 'isotope couple' ^{249}Cf(n,f) – ^{250}Cf(SF), together with corresponding data for other isotopes, will yield valuable information on the influence of the excitation energy on the particle emission probabilities.

Keywords: nuclear reaction ^{249}Cf(n,f), E = 5.4 meV, radioactivity ^{250}Cf(SF), measured ternary fission α, triton and ^{6}He emission probabilities and energy distributions.
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INTRODUCTION

Nuclear fission is essentially a binary process, but roughly 2 to 4 times every thousand fission events, the two heavy fragments are accompanied by a light charged particle. In this so-called ternary fission process, the emission yields for α particles, tritons and ^{6}He particles are the most important.

The ternary fission process is an important source of helium and tritium gas in nuclear reactors and also in used fuel elements due to spontaneous fission. Therefore accurate ternary fission yields for ^{4}He and tritons are requested by nuclear industry. Furthermore ternary fission data are of interest for nuclear physics in order to improve our understanding of the ternary particle emission mechanism and to provide information on the fission process itself.

An interesting characteristic of the process is related to the influence of the excitation energy of the fissioning nucleus on the ternary particle emission probabilities. This effect can be studied by comparing the ternary particle emission probabilities for the same compound nucleus at zero excitation energy (spontaneous fission) and at an excitation energy corresponding to the neutron binding energy (neutron induced fission).

In this paper, we describe new measurements performed on the neutron induced fission of ^{249}Cf (E_{exc} = 6.625 MeV) and on the spontaneous fission of ^{250}Cf (E_{exc} = 0 MeV). More specifically, characteristics of energy distributions and emission probabilities for α particles, tritons and ^{6}He particles will be shown.

The investigation of this 'isotope couple' ^{249}Cf(n,f) – ^{250}Cf(SF), together with corresponding data for other isotopes, measured in previous experiments done by our research group, will yield valuable information on the influence of the excitation energy on the particle emission probabilities.

EXPERIMENTAL SETUP

The spontaneous fission of ^{250}Cf has been studied at the Institute for Reference Materials and Measurements (IRMM) in Geel, Belgium. The ^{249}Cf(n,f) neutron induced measurement was carried out at the PF1b cold neutron guide installed at the high flux reactor of the Institute Laue-Langevin (ILL) in Grenoble, France. The thermal equivalent neutron flux at the sample position was 4×10^{9} neutrons/cm^{2}.s.

Sample Characteristics

The californium samples were prepared at the Lawrence Berkeley National Laboratory (LBNL) in the US. The 5.84 μg ^{249}Cf sample had an activity of 0.88 MBq and an enrichment of almost 100%, while the ^{250}Cf sample had a mass of about 1 μg and an activity of 4 MBq. The isotopic composition of the ^{250}Cf sample is given in Table 1. The contribution due to the spontaneous fission of ^{249,251}Cf was negligible. In both cases, the targets consisted of californium oxide with a diameter of 6 mm deposited on Ti-foils.

TABLE 1. Isotopic composition (in atomic percent) of the ^{250}Cf sample (07/06/2000).

Isotope	**^{249}Cf**	**^{250}Cf**	**^{251}Cf**
Abundance (%)	10.65	77.21	12.14

Detection System

For the ^{249}Cf neutron induced fission measurements the sample was placed in the centre of a vacuum chamber at an angle of 45 degrees with the incoming neutron beam, as shown in Figure 1. For the spontaneous fission of ^{250}Cf the same setup was used, however here we measured without neutron beam, so the sample could be placed right in front of the detectors. In both cases, a polyimide foil was used to cover the sample in order to prevent the contamination of the chamber by recoil nuclei.

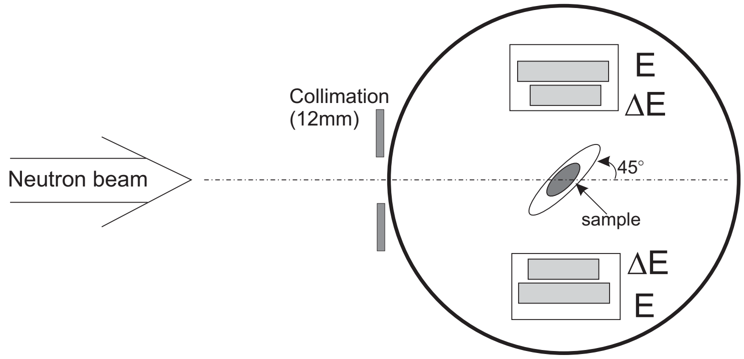

FIGURE 1. Experimental setup for the ^{249}Cf neutron induced fission measurement.

The measurements were performed in two separate steps. In a first step, ternary particles were detected, allowing the determination of both energy distributions and counting rates. Therefore two well-calibrated silicon surface barrier ΔE-E telescope detectors were placed at both sides of the sample.

In addition, ΔE detectors were covered with thin aluminium foils of 25 μm to stop alpha decay particles and fission fragments from penetrating the detector. Depending on the ternary particles we wanted to detect, the sample was turned in order to face the suited ΔE-E telescope detector.

Signals coming from the surface barrier detectors were sent through a pre-amplifier and an amplifier. These signals were digitized in an Analogue to Digital Converter (ADC), coincident ΔE and E signals were stored in a PC.

For both experiments the detector characteristics were chosen in order to have the best setup for detecting α and ^{6}He particles and measuring binary fissions, or for detecting α particles and tritons (Table 2).

TABLE 2. Thickness of surface barrier detectors used.

^{249}Cf	**LRA/B and ^{6}He [μm]**	**LRA and t [μm]**
ΔE	29.8	41
E	500	1500
^{250}Cf		
ΔE	29.8	55.1
E	500	1500

The ΔE-signal is proportional to the energy deposited by the ternary particle traversing the silicon surface barrier detector; the E-signal is proportional to the remaining particle energy.

In a second step, binary fission fragments were detected in order to determine the Binary Fission Yield (B). At this stage, the ΔE detector from the telescope suited to measure LRA/B, was removed, together with the aluminium foil, and replaced by a dummy ring with exactly the same dimensions. In this way, binary fission fragments could be measured with the E-detector under the same detection geometry as ternary particles.

ANALYSIS AND RESULTS

Particle Identification

The procedure used to identify various ternary particles and separate them from the background is the one proposed by Goulding et al. [1]. This method is based on the difference in energy loss of different particles in the same material using the equation: $T/a = (E + \Delta E)^{1.73} - E^{1.73}$, where T is the thickness of the ΔE detector and a is a particle and material specific constant.

The selection of ternary particles was realized by putting a window on the region of interest of the T/a spectrum. This has been done for Long Range Alpha particles (LRA), tritons and ^{6}He particles. In the case of the tritons, an additional correction due to the background was needed. After the selection, ΔE and E spectra were obtained for a given ternary particle and the total energy distribution could be deduced. The thresholds in energy for each ternary particle are due to the thickness of the ΔE detector, the electronic noise and the presence of the Al-foil. The average energy and the Full Width at Half Maximum (FWHM) of the energy distribution were obtained from a Gaussian fit performed on experimental data.

Results for the ^{249}Cf(n,f) Measurement

The spectrum obtained from the binary fission measurement is plotted in Fig. 2 (left). The small alpha pile-up peak due to the radioactive decay of ^{249}Cf has to be separated from the two bumps of the fission fragments. Then the remaining spectrum is extrapolated and the corresponding number of binary fission fragments was deduced after integration of the extrapolated spectrum.

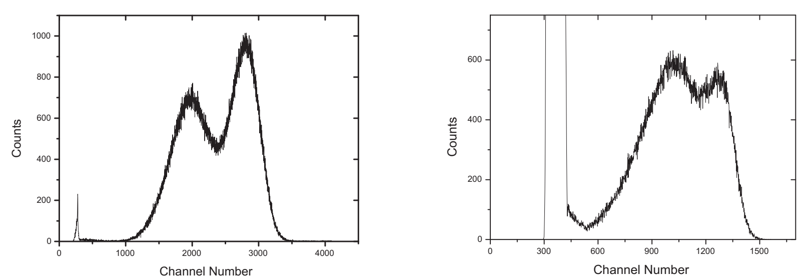

FIGURE 2. Measured binary fission spectrum for ^{249}Cf(n,f) (left) and for ^{250}Cf(SF) (right).

Fig. 3 (left) shows the spectra for the LRA, triton and ^{6}He measurements. The characteristics of these energy distributions are given in Table 3. Emission probabilities relative to ^{4}He are shown in Table 3 as well, together with the absolute emission probabilities. All the uncertainties given correspond to the sum of statistical and systematical uncertainties.

For LRA particles, a Gaussian fit was performed on experimental data with an energy above 12.5 MeV. In the case of ^{6}He particles the fit started at 9.9 MeV. For tritons, the fit was performed starting at 6 MeV. The triton yield was obtained after subtracting the contribution of the background.

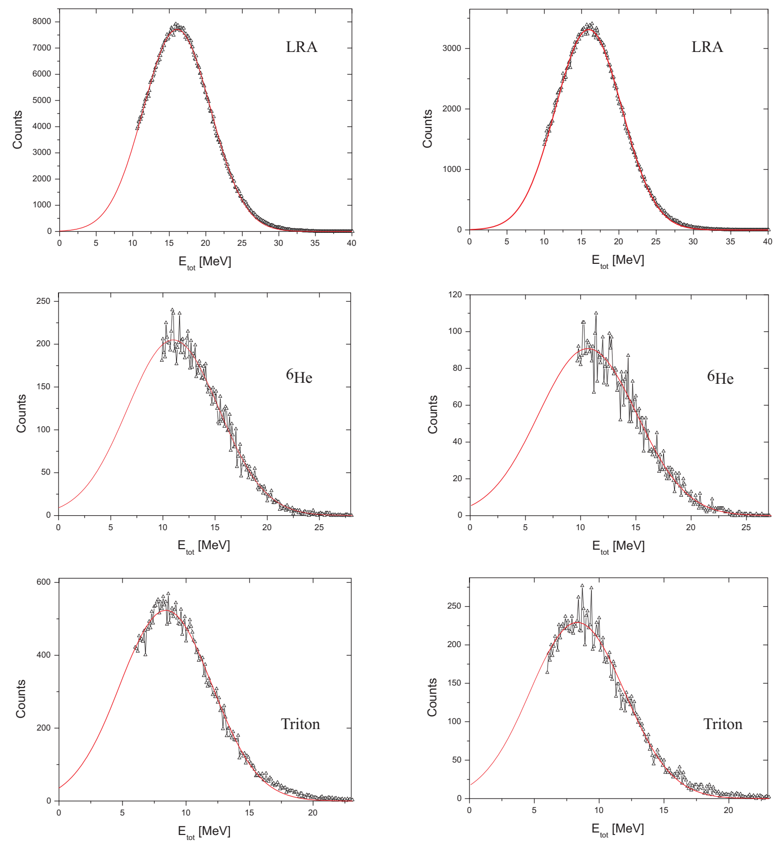

FIGURE 3. Energy distributions for LRA, ^{6}He and tritons for ^{249}Cf(n,f) (left) and ^{250}Cf(SF) (right).

TABLE 3. Values for average energy (E) and FWHM, as well as values for relative and absolute emission probabilities (per fission) for the various ternary particles measured.

^{249}Cf	**E [MeV]**	**FWHM [MeV]**	**rel. em. prob. [%]**	**abs. em. prob.**
LRA	16.07 ± 0.11	10.84 ± 0.14	100	(2.74 ± 0.08)x10^{-3}
^{6}He	10.99 ± 0.32	10.35 ± 0.60	2.54 ± 0.23	(6.99 ± 0.66)x10^{-5}
Tritons	8.42 ± 0.27	8.55 ± 0.43	7.68 ± 0.61	(2.10 ± 0.18)x10^{-4}
^{250}Cf				
LRA	15.95 ± 0.13	10.49 ± 0.16	100	(2.93 ± 0.10)x10^{-3}
^{6}He	10.64 ± 0.30	10.49 ± 0.54	2.74 ± 0.33	(8.03 ± 1.00)x10^{-5}
Tritons	8.31 ± 0.30	8.58 ± 0.49	6.96 ± 0.89	(2.08 ± 0.27)x10^{-4}

Results for the ^{250}Cf(SF) Measurement

The spectrum obtained from the binary fission measurement is shown in Fig. 2 (right). The determination of the binary fission yield is done in the same way as for the ^{249}Cf measurement.

The spectra for LRA, ^{6}He particles and tritons are plotted in Fig. 3 (right). For the LRA particles, a Gaussian fit was performed starting at 12.5 MeV. For ^{6}He particles a

Gaussian fit to all data points (starting at 9.7 MeV) was done. For the tritons, the fit was performed from 6 MeV and the triton yield was determined taking into account the background contribution. The values for the energy distribution of ternary particles are given in Table 3, as well as values for the emission probabilities.

DISCUSSION

Some two years ago, another measurement to determine the neutron induced fission of ^{249}Cf was performed, however with much less statistics and a higher detection limit in the case of tritons and ^{6}He particles [2]. Nevertheless, comparing our new results with the ones obtained before, a good agreement within the uncertainties exists. Therefore a weighted average of all the results of the two measurements can be calculated and will be used in order to perform the systematic study. Combining now our results for ^{249}Cf(n,f) and ^{250}Cf(SF) with previous results for a whole series of isotopes [3], interesting observations can be made.

Energy Distributions

By making a weighted average of all the average energies for the different isotopes for a certain ternary particle, we can conclude that this average energy remains constant within the uncertainties for a certain ternary particle. For LRA particles a value of (16.0 ± 0.1) MeV is obtained, for tritons (8.4 ± 0.1) MeV and for ^{6}He particles an average energy of (10.8 ± 0.2) MeV is calculated.

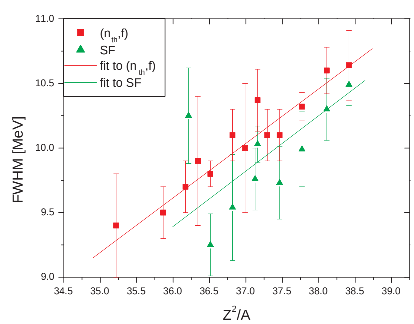

FIGURE 4. Graphic overview of the values for the FWHM for the ternary α's as a function of the fissility parameter of the fissioning nucleus.

Another striking observation, illustrated in Fig. 4, is that for the same compound nucleus, the FWHM for the ternary α energy distribution is systematically 0.3 MeV smaller for spontaneous fission than for neutron induced fission. It is the first time that this phenomenon could be demonstrated, thanks to our systematic study involving 9 spontaneously fissioning nuclides and 13 neutron induced fission reactions. Furthermore, the FWHM of a certain ternary particle linearly increases with increasing fissility parameter Z^2/A which can be seen too in Figure 4.

Emission Probabilities

To examine the influence of the excitation energy on the emission probabilities, Fig. 5 is shown. In the left figure the LRA emission probability is plotted as a function of Z^2/A of the compound nuclei.

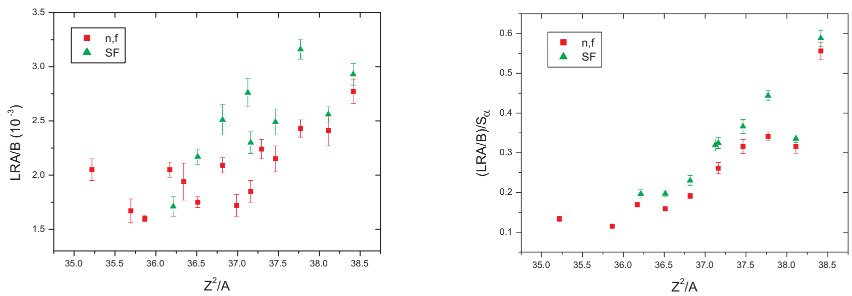

FIGURE 5. LRA/B (left) and $(LRA/B)/S_\alpha$ (right) as a function of Z^2/A of the compound nucleus.

First of all the general trend is demonstrated that the ternary particle emission probability increases with increasing fissility, although still strong fluctuations are seen. Another observation is that we see a decrease of the LRA emission probability with increasing excitation energy.

Fig. 6 shows the triton emission probability as a function of Z^2/A. Here we see a smooth increasing of t/B with increasing fissility and no decrease of the triton emission probability with increasing excitation energy is observed.

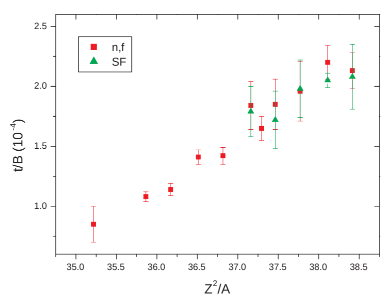

FIGURE 6. t/B as a function of Z^2/A of the compound nucleus.

This difference can be explained by introducing the so called alpha cluster preformation probability factor S_α. The LRA emission process is strongly influenced by this factor. When the fissioning nucleus is formed after capture of a neutron, S_α is likely to decrease due to the excitation energy. With this information, in fact $(LRA/B)/S_\alpha$ corresponds to the escape probability of an α-particle from the scissioning nucleus. Therefore, a new plot (Fig. 5, right) is made for the LRA particles, showing $(LRA/B)/S_\alpha$ as a function of Z^2/A. In this figure it can be seen that the fluctuations, visible in the left part of Figure 5, are mostly disappeared, and the data vary now in an almost smooth way as a function of Z^2/A as triton particles do.

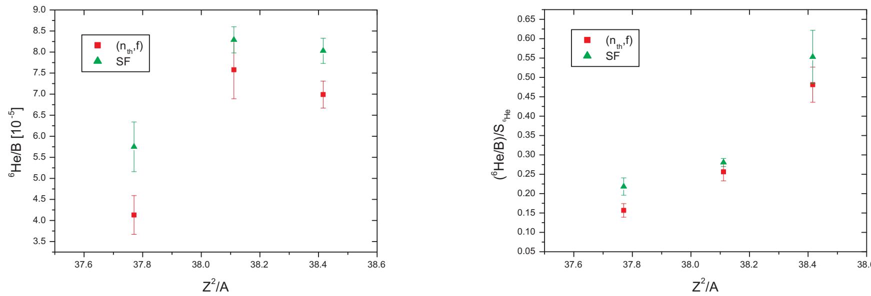

FIGURE 7. ^{6}He/B (left) and (^{6}He/B)/$S_{^{6}\mathrm{He}}$ (right) as a function of Z^2/A.

Let us now have a look at the ^{6}He particles. Fig. 7 (left) shows the absolute emission probability for the ^{6}He particles plotted as a function of Z^2/A. Again an indication of an increase of ^{6}He/B with increasing fissility is demonstrated. In addition, in all cases a higher value for spontaneous fission than for neutron induced fission can be observed. In analogy with the ternary α emission, a ^{6}He cluster preformation probability can be introduced, taking into account the following relation proposed by Blendowske [4]: $S = S_\alpha^{(A-1)/3}$, with A the mass of the emitted cluster. A new plot is made to illustrate the effect of that factor (Fig. 7, right). It can be seen that the data vary now in a more smooth way as a function of Z^2/A. Both observations permit to conclude that ^{6}He particles behave more like α-particles than like tritons.

CONCLUSION

In the present paper new results for the main characteristics (energy distributions and emission probabilities) of LRA, tritons and ^{6}He particles emitted in the neutron induced fission of ^{249}Cf and the spontaneous fission of ^{250}Cf are presented. Our systematic investigation of fissioning systems in the ground state (=spontaneous fission) and at an excited state (=neutron induced fission) permitted to put into evidence the strong impact of particle preformation on the ternary particle emission probability.

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