Neutron Multiplicities and Yields of Correlated Zr-Ce and Mo-Ba Fragment Pairs in Spontaneous Fission of ^{252}Cf

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High resolution γ-γ and γ-γ-γ coincidences in the spontaneous fission of ^{252}Cf were measured. Relative yields and neutron multiplicities from zero up to 10 were extracted directly for the first time in fission for the Zr-Ce and Mo-Ba correlated fragment pairs. The 0, 8, and the new 10 neutron emission yields for Mo-Ba are significantly larger than those reported previously for total ^{252}Cf spontaneous fission. Our results demonstrate that the use of this high resolution γ-γ-γ coincidence technique can provide important, previously inaccessible data on fission.

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It was shown earlier [1] and confirmed in our recent work [2] that the total intensities of the lowest $2^+ \rightarrow 0^+$ ground state band transitions observed in the deexcitation of even-even fission products reflect, to a high degree of accuracy (<5%), the total independent yields of these isotopes. Based on this observation we have determined the independent yields of individual correlated pairs of fission fragments of ^{252}Cf. Prior to our work, all previous information on the prompt neutron multiplicity distributions has been obtained from measurements of the neutrons emitted in induced or spontaneous fission, with zero neutron emission not observed directly, of course. The extraction of the neutron multiplicity distributions in these experiments involved (a) assumptions about the shape of these distributions, (b) neutron detector efficiencies, and (c) complex unfolding of the experimental data (see Refs. [3–5] for examples). Only the total neutron distributions have been obtained from these complex analyses. More sophisticated measurements (see, for example, Refs. [6,7]) allowed one to deduce the average neutron multiplicities, $\overline{\nu}$, and their variances as a function of the fission fragment mass. Here we report the first direct data on the yields and neutron multiplicities for Zr-Ce (40,58) and Mo-Ba (42,56) correlated pairs. We observe zero up to ten neutron emission, the latter for the first time. Both the zero and eight and new ten neutron emission yields for Mo-Ba are significantly higher than the previously reported total yields for ^{252}Cf spontaneous fission (see Ref. [5]). Our neutron multiplicity data demonstrate that the use of the high resolution γ-γ-γ coincidence technique with large detector arrays opens up new opportunities to understand the fission process by providing significant previously inaccessible information.

In this work we studied the multiple γ rays emitted by different pairs of fragments formed in the spontaneous fission of ^{252}Cf. The experiments were carried out at the Holifield Heavy Ion Research Facility with the ORNL Compton Suppression Spectrometer System. The 20 Compton-suppressed Ge detectors were located at a source-to-detector distance of 14 cm. In this experiment a hermetically sealed ^{252}Cf source with 6×10^4 spontaneous fission events per second was placed in the center of the detector array. All coincidence γ-ray events with multiplicity ≥ 2 were recorded. In a five day run, approximately 2×10^9 γ-γ coincidences were collected event by event.

A two-dimensional matrix of γ-γ coincidences (4096×4096 channels) was created from the initial data by selecting the γ-ray coincidences occurring within 200 ns. The peaks seen in this two-dimensional spectrum arise from the coincidences between γ rays emitted (a) promptly from each of the complementary fission fragments, (b) promptly from only one of the fragments, and (c) from the excited daughter states populated in a fragment β decay. The energy level diagrams of the Zr-Ce and Mo-Ba fragments of interest were taken from Refs. [8,9].

From the initial data, we also formed the so-called "double-gate" spectra. Each double-gate spectrum contained only γ rays in coincidence with the double-gated γ

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rays: e.g., one γ ray emitted by a chosen fission fragment and the other by its complementary partner. For double-gated spectra, lower count rates are obtained compared to the single-gated spectrum, since only the events with multiplicity ≥ 3 contribute in these double-gate spectra. However, the background and the complexity of the spectrum are greatly reduced. Additional γ rays from other isotopes besides the one of interest can occur in a single-gate window, but coincidences with these other γ rays are eliminated in a double-gated spectrum. Thus, one sees only the prompt γ rays belonging to the two correlated partners. Examples of the sensitivity of such double-gated spectra in level structure studies are found in Ref. [10]. Both the single-gated and the double-gated spectra were used for the determination of the transition intensities. This is the first published use of the high resolution γ-γ-γ coincidence technique to study the fission process.

After the corrections of the measured peak areas for the known detection efficiencies and internal conversion probabilities, we obtained the relative transition intensities. We neglected the small variations in the detection efficiency caused by the dependence of the prompt fission γ-ray multiplicity on the chosen fragment pair.

For each of the fragments, ^{146}Ce and ^{148}Ce, the yields were obtained in their combinations with different Zr isotopes. For even isotopes of Zr these yields were determined from the measured intensities of their $2^+ \rightarrow 0^+$ transitions in coincidence with the $2^+ \rightarrow 0^+$ transitions in the Ce isotopes. In the case of the odd Zr isotopes, the intensities of all the transitions to their ground states were summed up. From our measured relative yields, we obtained the independent yields of the fragment pairs by normalization to known integral independent yields of ^{146}Ce and ^{148}Ce [11] Table I gives these independent yields of the fragment pairs and the number of neutrons emitted for each correlated Zr-Ce pair (ν). These go from zero to eight neutrons emitted. In the last column of Table I are given the integral yields of ^{146}Ce and ^{148}Ce ($\sum Y$) normalized to that from Ref. [11] and mean numbers of prompt neutrons ($\bar{\nu}$) emitted with these Ce fragments. When plotted as functions of mass number, the yields of the Zr isotopes accompanying ^{146}Ce and ^{148}Ce lie on smooth curves. This supports the correctness of our determination of the yields of the odd Zr isotopes. It is interesting to note that the average neutron multiplicity for ^{148}Ce, $\bar{\nu} = 3.2$, is considerably less than for ^{146}Ce, $\bar{\nu} = 4.6$.

A more complete set of relative yields of different pairs of fission fragments was measured for the charge division of ^{252}Cf, $Z_L/Z_H = 42/56$ (Mo/Ba). The γ-ray coincidences investigated in the extraction of these yields involved the $2^+ \rightarrow 0^+$ transitions in the even-even Mo and Ba isotopes, the 117-keV ground state transition in ^{143}Ba, and known ground state transitions in odd Mo isotopes. We normalized the obtained relative yields to the sum of the independent yields of ^{140}Ba, ^{142}Ba, ^{143}Ba, ^{144}Ba, and ^{146}Ba known from Ref. [11]. The independent yields of the Mo-Ba fragment pairs obtained in this way are presented in Table II (for each pair the frequency of occurrence is given with relation to 100 fission events). As seen in Table II, we have obtained the yields of the fragment pairs corresponding to the emission of prompt neutrons ranging in number from zero to ten. This is the first direct observation of zero neutron emission and the first evidence for ten neutron emission in spontaneous and thermal neutron induced fission. The double-gated spectra provided the only way to unravel the ^{104}Mo-^{138}Ba ($\nu = 10$) and ^{108}Mo-^{138}Ba ($\nu = 6$) and the ^{104}Mo-^{144}Ba ($\nu = 4$) and ^{108}Mo-^{144}Ba ($\nu = 0$) channels because the $2^+ \rightarrow 0^+$ transitions in ^{104}Mo and ^{108}Mo are 192.8 and 192.9 keV, respectively. The mean numbers of neutrons ($\bar{\nu}$) emitted with different Ba fragments are given in the last rows of Table II.

The detailed experimental distributions of correlated fragment pair yields and distributions of numbers of emitted neutrons as given in Tables I and II are obtained for the first time. The literature information about the mass and charge distributions of fission fragments corresponds to the more integral fission characteristics. In order to compare our results with previously reported semiempirical data of total independent yields of individual fragments, we summed our data. We obtained first the independent yields of barium fragments by summing up the numbers in the columns of Table II. These yields ($\sum Y$) are given in the lower part of Table II. Then we estimated the independent yields for the missing fragment pairs involving odd barium isotopes (^{139}Ba, ^{141}Ba, ^{145}Ba, and ^{147}Ba) by linear interpolations of the data for adjoining pairs of the corresponding even barium isotopes. This enabled us to draw the complete isotopic distribution patterns for barium and molybdenum fragments shown in Fig. 1. These independent yield distributions are compared with the estimations made in Ref. [11] (A distribution). The agreement is quite good with the

TABLE I. Yields of the correlated Zr-Ce fragment pairs in spontaneous fission of ^{252}Cf (the numbers are per 100 fission events).

	^{98}Zr	^{99}Zr	^{100}Zr	^{101}Zr	^{102}Zr	^{103}Zr	^{104}Zr	
^{146}Ce	0.04 ± 0.1	0.04 ± 0.03	0.09 ± 0.02	0.40 ± 0.10	0.35 ± 0.03	0.13 ± 0.04	0.04 ± 0.02	$\sum Y = 1.0$
ν	8	7	6	5	4	3	2	$\bar{\nu} = 4.6$
^{148}Ce	0.04 ± 0.1	0.14 ± 0.04	0.57 ± 0.05	1.06 ± 0.17	0.32 ± 0.03	0.10 ± 0.04	0.04 ± 0.03	$\sum Y = 2.2$
ν	6	5	4	3	2	1	0	$\bar{\nu} = 3.2$

TABLE II. Yields of the correlated Mo-Ba fragment pairs in spontaneous fission of ^{252}Cf (the numbers are per 100 fission events).

	^{138}Ba	^{140}Ba	^{142}Ba	^{143}Ba	^{144}Ba	^{146}Ba	^{148}Ba
^{102}Mo			0.05 ± 0.04	0.03 ± 0.03	0.07 ± 0.05	0.13 ± 0.05	0.05 ± 0.03
^{103}Mo			0.02 ± 0.02	0.15 ± 0.09	0.59 ± 0.07	0.47 ± 0.09	0.05 ± 0.05
^{104}Mo	0.11 ± 0.03	0.16 ± 0.04	0.30 ± 0.06	0.44 ± 0.08	1.12 ± 0.04	0.36 ± 0.04	0.04 ± 0.03
^{105}Mo	0.02 ± 0.02	0.09 ± 0.04	0.72 ± 0.07	1.04 ± 0.16	1.47 ± 0.09	0.17 ± 0.07	
^{106}Mo	0.01 ± 0.01	0.12 ± 0.02	0.93 ± 0.04	0.97 ± 0.11	0.69 ± 0.04	0.03 ± 0.02	
^{107}Mo	0.02 ± 0.02	0.18 ± 0.08	0.44 ± 0.08	0.22 ± 0.08	0.11 ± 0.06		
^{108}Mo	0.04 ± 0.02	0.10 ± 0.05	0.16 ± 0.08	0.09 ± 0.07	0.04 ± 0.02		
$\sum Y$	0.20 ± 0.05	0.65 ± 0.12	2.62 ± 0.16	2.93 ± 0.25	4.07 ± 0.15	1.16 ± 0.13	0.14 ± 0.06
$\bar{\nu}$	>6	6.0 ± 1	4.3 ± 0.4	3.7 ± 0.5	3.40 ± 0.2	2.4 ± 0.2	1.0 ± 0.7

exception of two points for the lightest Ba isotopes. In Fig. 1 we also show the curves which are Gaussian fits to the data of Table II. These curves show a better agreement with the data of Ref. [11]. We believe that the obtained higher yields for the lightest Ba isotopes are an indication of a possible real deviation of the fission fragment isotopic distribution from the data of Ref. [11] which include approximations such as a Gaussian shape and systematics.

On the basis of the yields of the Mo-Ba fragments, we obtained the neutron multiplicity distribution for ^{252}Cf spontaneous fission that results in this particular charge division. In Fig. 2 this distribution is compared with the total neutron multiplicity distribution for ^{252}Cf known from the literature [5]. We note that the Mo/Ba charge division corresponds to about a 15% branch of the ^{252}Cf spontaneous fission. Our data indicate an enhancement of the lower (0 and 1) and higher (7–10) neutron multiplicities. Are these differences the manifestation that some less abundant fission channels are enhanced for the specific charge division? It will be interesting to study other Z_L/Z_H neutron multiplicity distributions to see if

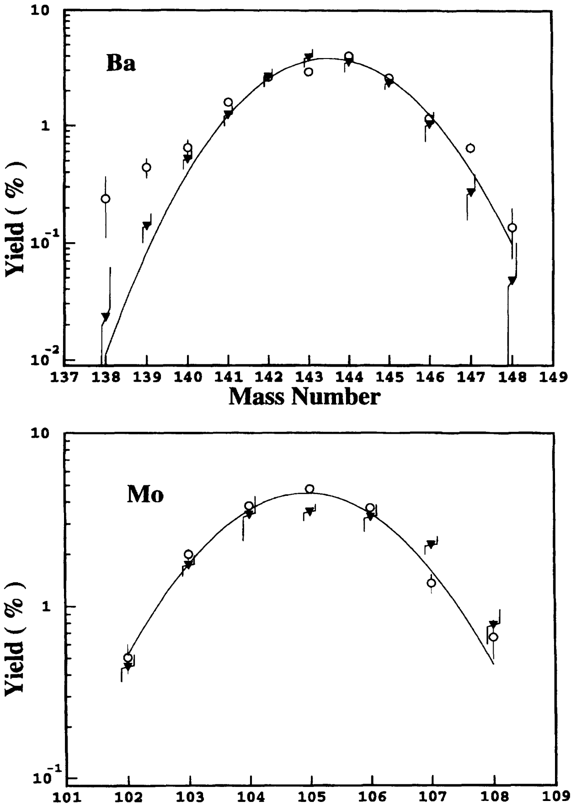

FIG. 1. Isotopic distributions of Ba and Mo fission fragments. Circles and triangles represent the results of this work and Ref. [11], respectively. Our mean values ($\bar{A}$) and variances (σ_A) are as follows. For Ba, $\bar{A} = 143.3$, $\sigma_A = 1.95$; for Mo, $\bar{A} = 104.9$, $\sigma_A = 1.37$. The solid curves in the figure represent a Gaussian fit to the data of Table II.

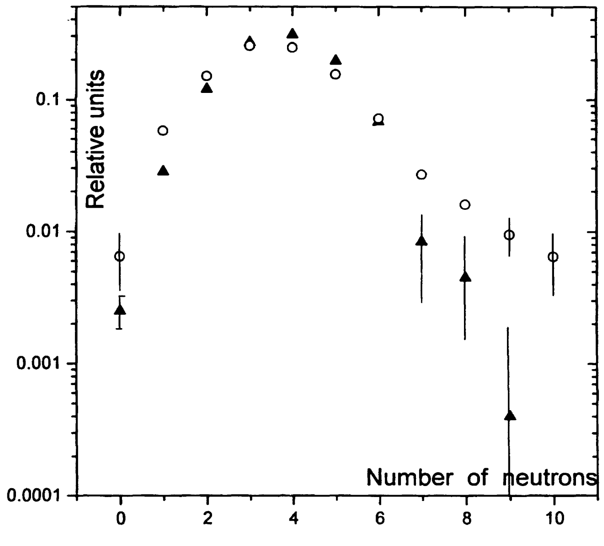

FIG. 2. Neutron multiplicity distributions. The distribution obtained in this work for the mode resulting in the formation of Mo-Ba fragments is shown by circles ($\langle \nu \rangle = 3.79$, $\sigma_\nu = 1.65$). Triangles show the distribution obtained in Ref. [5] for the whole sample of fragments formed in the ^{252}Cf spontaneous fission ($\langle \nu \rangle = 3.77$, $\sigma_\nu = 1.27$). Both distributions are normalized to a total of 1 as in Ref. [5]. If our data are normalized to the peak multiplicities ($\nu = 3, 4, 5$) of Ref. [5], the enhancement of our high and low multiplicities would be greater.

there is a duplication of our currently observed enhancement at the low and high neutron multiplicities in the Mo-Ba pairs compared to the total distribution. Again, we would like to emphasize that the whole procedure of obtaining directly the neutron multiplicity distribution as exploited in this work differs entirely from the procedures used earlier. Thus it may be that the reported total neutron multiplicities may be underestimated at high and low multiplicity.

Finally, the results presented here demonstrate the power of a new approach to the study of the fission process that can provide a wide range of previously inaccessible data on the fission process. More data similar to those presented in Tables I and II can be obtained for ^{252}Cf spontaneous fission. Another important class of information that can be extracted from such data is the relative populations of different spin states in the fission process. Also, it will be interesting to use such data for reconstructing the charge, mass, and excitation energy distributions of the primary fission fragments formed just after scission. Alternatively, different theoretical predications for such distributions can then be compared to experimental data.

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