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ISOTOPES OF ELEMENT 102 WITH MASS 251 TO 258†

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The purpose of this Letter is to outline recent information obtained in our laboratory concerning a wide range of isotopes of the element with atomic number 102. A more detailed article is presently being prepared for submission elsewhere.

We have used ^{12}C and ^{13}C ions accelerated by the Berkeley heavy-ion linear accelerator (HILAC) to bombard essentially monoisotopic targets of ^{244}Cm, ^{246}Cm, and ^{248}Cm for the production of the isotopes $^{251}102$ to $^{258}102$. The apparatus is an elaboration of a simple principle first observed in this laboratory some years ago.[1] Atoms recoiling from the target are stopped in a stream of helium at 600 Torr and carried by this gas through an orifice about 0.2 mm in diameter into an evacuated space. The gas jet impinges a few millimeters away on the periphery of a wheel and a large fraction (~80%) of the heavy atoms attach themselves to its surface. At regular intervals the wheel is digitally rotated about 50° to expose the collected atoms to Au-Si surface-barrier alpha-particle detectors. In this series of experiments four detectors, equally spaced along the circumference of the wheel, were used simultaneously in order to obtain half-life information as well as alpha-particle energies. The targets, made by molecular deposition, were 0.2 to 0.5 mg/cm^2 of curium oxide on 4 to 5 mg/cm^2 beryllium metal. The carbon-ion beam currents used were typically 2×10^{12} particles/sec in an area of 0.2 cm^2. Changes in bombarding energy were made by inserting different thicknesses of Be degrader foils in the beam path so that excitation functions for the alpha-particle activities could be determined.

The electronic circuitry to analyze the pulse outputs from the individual detectors was conventional. After preamplification with charge-sensitive amplifiers located near the detectors, the pulses were shaped by delay lines to one microsecond and further amplified in the counting area. They were then sorted with a two-parameter analyzer into four two-hundred-channel groups. The resolution of the system varied from 25 to 50 keV full width at half-maximum depending on the detector used. Spontaneous fissions were recorded by discriminators set to trigger on pulses greater than 30 MeV in amplitude. It was necessary to gate the system off during each beam pulse to prevent spurious signals from neutron reactions and thus a 20% loss was suffered. The total counting efficiency, defined as the ratio of the counts observed to the alpha disintegrations undergone by the nuclei transmuted from the target, was about 10%. Supplementary measurements of spontaneous fission activity were made in many experiments with the aid of mica detectors.[2]

Fortunately the production cross sections to form the element-102 isotopes by these reactions are in the range from 10^{-31} to 10^{-30} cm^2 so that it has been possible to make mea-

surements of energies and half-lives with relatively good statistical accuracy. Each isotope except for $^{251}102$ was made by more than one reaction as a check on the mass assignment obtained from the peak energy of its excitation function. Half-lives were obtained by the relative amounts of alpha activity in the four detectors after correcting for individual counting efficiencies. These were obtained by measuring the alpha-particle decay of a known activity (typically ^{214}Ra).

A difficulty that was encountered initially was created by the discovery that the 2.6-sec ^{214}Ra decayed to a slight extent (~0.1%) by electron capture to 4-msec, 8.43-MeV ^{214}Fr. The ^{214}Ra was produced in these experiments by reactions of carbon ions with lead isotopes present in the target as impurities. This background activity interfered in some cases with the radiation from the element 102 but it was possible to correct adequately for this effect by referring to the amount of the ^{214}Ra parent activity. (Similarly, corrections for the alpha-particle groups from ^{211m}Po were taken into account where they were a possible source of background.)

Figure 1 shows typical alpha energy spectra for the last 100 channels in which the groups due to alpha decay of the various element-102 isotopes are indicated. The spectra below channel 100, in general, consist mainly of various Pb-induced activities such as ^{211}Ra, ^{212}Ra, ^{213}Ra, ^{214}Ra, ^{211m}Po, and ^{211}Po in addition to ^{249}Fm and ^{250}Fm. Energy calibration was obtained primarily with pulse generators calibrated with the 7.136-MeV ^{214}Ra peak.

Table I summarizes these measurements. As well as half-lives and alpha energies, it includes the formation cross sections at the peaks of the excitation functions. For comparison we have listed the most recent data available from Flerov's groups at Dubna, as published at a recent conference.[3]

It can be seen that within the statistical errors there is reasonable agreement between the two sets of results except in the value for the half-life of $^{256}102$. Several attempts were made to find an alpha-emitting, 8-sec activity that could be attributed to $^{256}102$ but these were unavailing. The source of the discrepancy is unknown and thus the Dubna value for its spontaneous-fission branching ratio must be called into question. It would seem possible that an isomer may be responsible for much or all

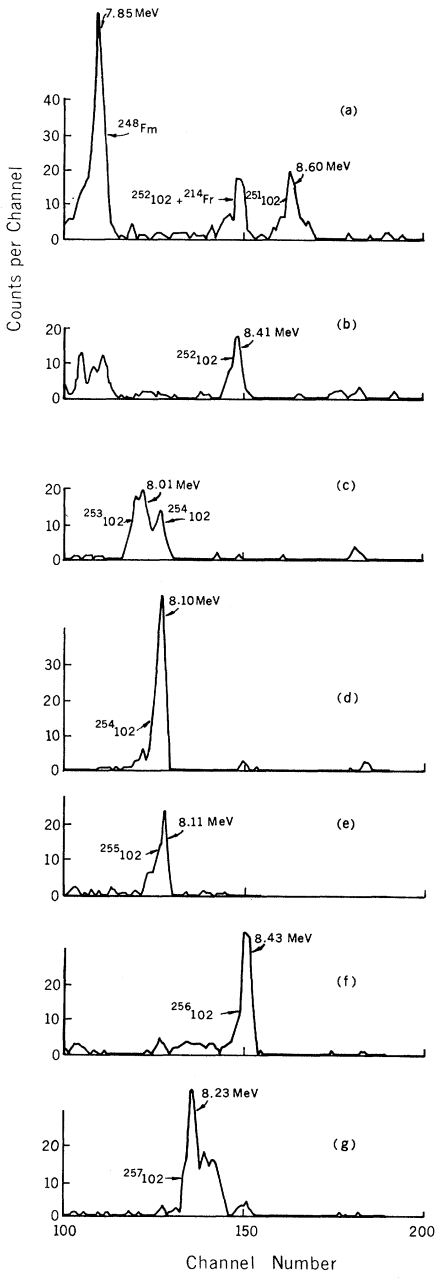

FIG. 1. Alpha spectra obtained in the bombardments of various Cm isotopes with C ions: (a) ^{244}Cm + 78-90 MeV ^{12}C, 28.5 μA h +6 ions; (b) ^{244}Cm + 70.9 MeV ^{12}C, 4.0 μA h +6 ions; (c) ^{244}Cm + 62-74 MeV ^{13}C, 6.8 μA h +6 ions; (d) ^{246}Cm + 70 MeV ^{12}C, 2.0 μA h +6 ions; (e) ^{246}Cm + 70.8 MeV ^{13}C, 1.0 μA h +6 ions; (f) ^{248}Cm + 71-73 MeV ^{13}C, 3.0 μA h +6 ions; and (g) ^{248}Cm + 63-68 MeV ^{13}C, 9.4 μA h +6 ions.

of the spontaneous fission activity reported by Kuznetsov, Lobanov, and Perelygin,[4] since in their experiments a half-life of 8 sec was clearly observed. We have seen what could

Table I. Production schemes and decay properties of various isotopes of element 102.[a]

Isotope	Reaction used	Peak cross section[c] (10^{-30} cm^2)	Half-life (sec)	αE (MeV) (±.02)	SF/α ratio	Reaction used	Half-life (sec)	αE (MeV) (±.03)	SF/α ratio
$^{251}102$	^{244}Cm(^{12}C,5n)	0.09	0.8±0.3	8.60(80%) 8.68(20%)		Not reported			
$^{252}102$	^{244}Cm(^{12}C,4n)	0.13	2.3±0.3	8.41	1/2[d]	^{239}Pu(^{18}O,5n)	4.5±1.5	8.41	
	^{244}Cm(^{13}C,5n)	0.096	~2.5[b]	8.41	1/2[d]				
$^{253}102$	^{244}Cm(^{13}C,4n)	0.29	105±20	8.01		^{242}Pu(^{16}O,5n)	95±10	8.01	
	^{246}Cm(^{12}C,5n)	0.25	~100[b]	8.01		^{239}Pu(^{18}O,4n)			
$^{254}102$	^{246}Cm(^{12}C,4n)	0.89	55±5	8.10		^{243}Am(^{15}N,4n)	--	8.11	≤1/1800
	^{246}Cm(^{13}C,5n)	0.54	~50[b]	8.10		^{238}U(^{22}Ne,6n)	50±10		
	^{244}Cm(^{13}C,3n)	0.096	~50[b]	8.10		^{242}Pu(^{16}O,4n)	75±15		
$^{255}102$	^{246}Cm(^{13}C,4n)	0.47	185±20	8.11		^{238}U(^{22}Ne,5n)	~120	8.08	
	^{248}Cm(^{12}C,5n)	0.38	~180[b]	8.11		^{242}Pu(^{18}O,5n)	180±10	8.09	
$^{256}102$	^{248}Cm(^{12}C,4n)	0.74	2.9±0.5	8.43	~1/400[d]	^{238}U(^{22}Ne,4n)	6±2	8.41	1/200
	^{248}Cm(^{13}C,5n)	0.75	3.2±0.2	8.43		^{242}Pu(^{18}O,4n)	9±3	8.42	
	^{246}Cm(^{13}C,3n)	0.09		8.43					
$^{257}102$	^{248}Cm(^{13}C,4n)	1.1	23±2	8.23(50%) 8.27(50%)		Not reported			
	^{248}Cm(^{12}C,3n)	0.08	~20[b]	8.25					

[a]Data in columns 2-6 are from present work; those in the last four columns are taken from Ref. 3.

[b]No error given due to rather poor statistics.

[c]The relative values are good to within 25%, the absolute values to within a factor of two.

[d]The mass assignment of the SF emitter is not conclusive.

be spontaneous fission branching by the 3-sec $^{256}102$ at a level approximately a factor of 2 below that found in the Dubna experiments. Because of the low branching ratio and a longer-lived background produced in the experiment we have not yet procured enough data concerning its half-life and excitation function to define its assignment exclusively to that nuclide.

As a matter of historical interest it is worth pointing out that the activities, both alpha particles and spontaneous fissions, that are now best ascribed to $^{252}102$ were first observed in our laboratory.[5] We find the same ratio of these activities now as we did then and feel that the assignment of the spontaneous fission activity observed is most reasonably made to the isotope $^{252}102$. In the earlier work the "3-sec 8.3-MeV" alphas and spontaneous fissions were thought to be due to $^{254}102$ because the 3-sec value coincided with a half-life determined the year before by a milking method in which ^{250}Fm was observed.[6] We now believe that because of resolution and drift problems it was possible to confuse 20-min, 7.22-MeV ^{244}Cf with 30-min, 7.43-MeV ^{250}Fm. In the milking experiments ^{244}Cf would have been produced as the granddaughter of 2-sec $^{252}102$ [reaction ^{244}Cm(^{12}C, $4n$)][7]; the yield of this isotope we now find is consistent with this hypothesis. In the chemical verification experiments which proved ^{250}Fm to be the alpha-recoiling daughter of $^{254}102$, there was no resolution problem since ^{244}Cf was separated by cation-exchange columns. In these experiments there was however no half-life measurement since all of the catchers were used for the chemical separations. As a part of the experiments in which element 103, lawrencium, was discovered,[8] alpha particles of 8.2 MeV and 15-sec half-life were correctly ascribed to element 102 but now it is clear that they belong to mass 257 rather than 255 as suggested at that time.

We have searched diligently for alpha radiation from the isotope $^{258}102$ without success. From the data for all of the nuclides in this region we would predict an alpha-decay half-life of about a minute and a production cross section via the reaction $^{248}Cm(^{13}C, 3n)$ of the order of 10^{-31} cm^2. We should have readily observed its presence either directly or via its daughter ^{254}Fm in alpha-recoil milking experiments and consequently we feel that its most likely mode of decay is by spontaneous fission. Preliminary experiments set a half-life limit of much less than a second for spontaneous fission.

This work was aided greatly by the efforts of many people but we would particularly like to acknowledge the assistance of the following: T. Bowman for preparation of the targets, R. Latimer for target material purification, P. Fields and J. Lerner of the Argonne National Laboratory for the isotopically separated ^{246}Cm, A. Larsh for electronics support, C. Corum for mechanical design, and F. Grobelch and the HILAC crew for the superb performance of the accelerator. We also thank Jaromir Maly for many helpful discussions.

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TERNARY FISSION OF HEAVY NUCLEI*

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A heightened interest has lately centered on that fission process by which three large fragment masses are formed. We wish to present preliminary results from the study of ternary fission of a variety of heavy nuclei. Although the energy and mass distributions (and implications therefrom) are in themselves of considerable interest, the main purpose of this communication is to point out how these data are inconsistent with an explanation based on scattering phenomena, a view which is currently popular in some circles.

The experimental arrangement has been previously described.[1] Briefly, it consists of three solid-state detectors positioned 120° apart in a plane about a fission source. The physical properties of the sources were essentially identical (excepting the Cf^{252} source[1]) and consisted of a tetrafluoride deposit on 20-μg/cm^2 VYNS film support; areal densities varied around 0.100 μg/cm^2. Calibration against the binary-fission fragment spectrum was made for each foil before and after each experiment. The output pulse from each detector was digitally analyzed and recorded event by event on punched paper tape whenever a parallel triple-coincidence circuit was satisfied. Energy calibration was achieved by comparing the binary-fission fragment spectrum with that from time-of-flight data[2] in order to locate the energy position of the average light and heavy masses. A straight-line calibration was then applied; alternatively, a mass-dependent approach as proposed by Schmitt et al.[3] was used.

The following fissioning systems were investigated:

$$Cf^{252} \rightarrow (\text{spontaneous})TF, \quad (1)$$

$$Pu^{241} + n_{th} \rightarrow TF, \quad (2)$$

$$Pu^{239} + n_{th} \rightarrow TF, \quad (3)$$

$$U^{235} + n_{th} \rightarrow TF, \quad (4)$$