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ISOTOPES OF ELEMENT 102 WITH MASS 251 TO 258

Albert Ghiorso, Torbjorn Sikkeland, and Matti J. Nurmin

February 1967
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 to Physical Review.

We have used $^{12}$C and $^{13}$C ions accelerated by the Berkeley HILAC to bombard essentially monoisotopic targets of $^{244}$Cm, $^{246}$Cm, and $^{248}$Cm for the production of the isotopes $^{251}$102 to $^{258}$102. The apparatus is an elaboration of a simple principle first observed in this laboratory some years ago. Atoms recoiling from the target are stopped in a stream of helium at 600 torr and carried by this gas through an orifice about 0.2 mm in diameter into an evacuated space. The gas jet impinges a few millimeters away on the periphery of a wheel and a large fraction (~80%) of the heavy atoms attach themselves to its surface. At regular intervals the wheel is digitally rotated about 50° to expose the collected atoms to Au-Si surface barrier alpha particle detectors. In this series of experiments four detectors, equally spaced along the circumference of the wheel, were used simultaneously in order to obtain half-life information as well as alpha particle energies. The targets, made by molecular deposition, were 0.2 to 0.5 mg/cm$^2$ of curium oxide on 4 to 5 mg/cm$^2$ beryllium metal. The carbon ion beam currents used were typically $2 \times 10^{12}$ 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 pre-amplification 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 percent 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 percent. Supplementary measurements of spontaneous fission activity were made in many experiments with the aid of mica detectors.

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 measurements 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-second $^{214}$Ra decayed to a slight extent ($\sim 0.1\%$) by electron
capture to 4 ms 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 adequately correct 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 G. N. Flerov's groups at Dubna, U.S.S.R. 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-second activity that could be attributed to $^{256}$102 but 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 of the spontaneous fission activity reported by Kuznetsov, et al since in their experiments a half-life of 8 seconds was clearly observed. We have seen what could be spontaneous fission branching by the 3-sec $^{256}$Fm at a level approximately a factor of two 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 $^{252}$Fm were first observed in 1959 in our laboratory. 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 $^{252}$Fm. In 1959 the "3-second 8.3 MeV" alphas and spontaneous fissions were thought to be due to $^{254}$Fm because the 3-second value coincided with a half-life determined the year before by a milking method in which $^{250}$Fm was observed. 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 1958 milking experiments $^{244}$Cf would have been produced as the granddaughter of 2-sec $^{252}$Fm ($^{244}$Cm($^1\text{H},4\text{n}$) reaction); 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 $^{254}$Fm, 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. In 1961 as a part of the experiments in which element 103, lawrencium, was discovered.

*The curium targets used in 1958-1959 had an isotopic composition of 95% $^{244}$Cm and ~5% $^{246}$Cm.
alpha particles of 8.2 MeV and 15-second 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 $^{258}\text{E}_{\text{102}}$ 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 $^{248}\text{Cm} (^{13}\text{C},3\text{n})$ reaction of the order of $10^{-31} \text{ cm}^2$.

We should have readily observed its presence either directly or via its daughter $^{254}\text{Tm}$ 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 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 $^{246}\text{Cm}$, 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.
This work was done under the auspices of the U. S. Atomic Energy Commission.

†On leave of absence from the Department of Physics, University of Helsinki, Finland.


4. V. I. Kuznetsov, Yu. V. Lobanov, V. P. Perelygin, Dubna preprint 2525, Dec. 1965. This reference indicates $8.2 \pm 1.0$ sec for the spontaneous fission activity.


<table>
<thead>
<tr>
<th>Isotope</th>
<th>Reaction used</th>
<th>Peak cross section (10^{-20}cm^2)</th>
<th>Half-life (sec)</th>
<th>CE (MeV)</th>
<th>SF/α ratio</th>
<th>Reaction used</th>
<th>Half-life (sec)</th>
<th>CE (MeV)</th>
<th>SF/α ratio</th>
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<td>0.8±0.3</td>
<td>8.60 (80%)</td>
<td>8.68 (20%)</td>
<td>239_{18}O_{5n}</td>
<td>4.5±1.5</td>
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<td></td>
<td>244_{13}C_{5n}</td>
<td>0.096</td>
<td>~2.5^b</td>
<td>8.41</td>
<td>1/2^d</td>
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<td>1/2^d</td>
<td>239_{18}O_{5n}</td>
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<td></td>
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<td>1/200</td>
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Data in columns 2-6 are from present work; those in the last four columns are taken from Ref. 3.

No error given due to rather poor statistics.

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

The mass assignment of the SF emitter is not conclusive.
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 $\mu$Ah +6 ions
(b) $^{244}$Cm + 70.9 MeV $^{12}$C, 4.0 " "
(c) $^{244}$Cm + 62-74 MeV $^{13}$C, 6.8 " 
(d) $^{246}$Cm + 70 MeV $^{12}$C, 2.0 " 
(e) $^{246}$Cm + 70.8 MeV $^{13}$C, 1.0 " 
(f) $^{248}$Cm + 71-73 MeV $^{13}$C, 3.0 " 
(g) $^{248}$Cm + 63-68 MeV $^{13}$C, 9.4 " 

Fig. 1
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