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DISCLAIMER

POSITIVE IDENTIFICATION OF TWO ALPHA PARTICLE EMITTING ISOTOPES OF ELEMENT 104*

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ABSTRACT

We have studied isotopes of element 104 formed by bombarding ^{249}Cf with ^{12}C and ^{13}C ions. ^{257}104 and ^{259}104 were positively identified by milking their daughters, ^{253}No and ^{255}No. ^{257}104 is a 4.5-sec alpha particle activity with a complex spectrum; ^{259}104 is likewise an alpha emitter with a half-life of 3 sec. ^{258}104 is tentatively identified as an 11-msec spontaneous fission activity.

In a recent report[1] we have described our unsuccessful attempts to confirm the attribution by Dubna experimenters of a 0.3-second spontaneous fission emitter to an isotope of element 104.[2] In this letter we will describe what we believe are definitive experiments which characterize the isotopes ^{257}104 and ^{259}104. In addition, tentative evidence for ^{258}104 is included.

A 60 microgram target of mono-isotopic[3] ^{249}Cf was electrodeposited from an isopropyl alcohol solution in an area of 0.21 cm^2 on a substrate sandwich consisting of 0.1 mg/cm^2 Pd sputtered onto 2.2 mg/cm^2 Be. This 290 microgram/cm^2 target was bombarded mainly by ^{12}C and ^{13}C ions accelerated by the Berkeley HILAC to make isotopes of element 104 but an appreciable fraction of the bombardments were conducted with ^{11}B, ^{14}N, ^{15}N, and ^{16}O ions. Currents in the range of 4 to 5 microamperes dc (as completely stripped ions) were typically used and the total number of microamperehours was several thousands.

The apparatus used was similar to that described in our papers[4,5] concerning the properties of various isotopes of nobelium. The transmuted atoms recoiling from the target were swept by helium gas from the target region operating at a pressure of about 500 torr through a 0.4 mm diameter orifice into vacuum to be deposited onto the periphery of a 45 cm diameter wheel. The digitally-controlled wheel was periodically rotated to place these atoms into positions adjacent to four Si-Au surface barrier crystal detectors[6] so that after suitable amplification and analysis alpha particle spectra could be obtained. The energy and half-life data were stored in a PDP-9 computer. The bombarding energy was measured with a solid state detector looking at particles scattered from the target at an angle of 20° and was adjusted by means of Be metal foil degraders to reduce the energy of the 10.4 MeV/nucleon particles from the HILAC. The beam current was monitored by a water-cooled Faraday cup. Because of the very high beam densities that were used, it was also necessary to liquid-cool the windows, the degrader foils, and the target.

The ^{12}C bombardments of the ^{249}Cf target produced a new alpha particle activity with a complex energy spectrum (Fig. 1) and a half-life of 4.5 ± 1.0 seconds. The spectra were submitted to a computer analysis[7] with the result that four prominent alpha particle groups with energies 8.70, 8.78, 8.95, and 9.00 MeV and relative intensities 0.15, 0.2, 0.3, 0.35, respectively, were assigned to the activity. For alpha energy calibration, the 6.043 MeV peak of $^{209,210}Rn$ and the 7.43 MeV peak of ^{250}Fm were used and extrapolation was made by use of a pulse generator. The absolute accuracy of the energy values is estimated to be 0.02 MeV. The amount of activity was substantial (~5 counts per microamperehour) and it was possible to measure its excitation function with good accuracy (Fig. 2). The function, with a maximum cross section of approximately 10^{-32} cm^2, corresponds to that expected for the $^{12}C,4n$

reaction to produce $^{257}104$ and is consistent with the calculations[8] of Sikkeland and Lebeck.

To make certain of the mass and atomic number of this activity, a series of experiments was carried out to identify the ^{253}No daughter, an alpha particle emitter with an energy of 8.01 MeV and a half-life of 105 seconds.[4,9] After a period of 200 seconds during which the $^{257}104$ alpha activity was measured, the four crystal detectors were automatically shuttled to positions opposite four similar detectors. This was done in order to measure the activity at high geometry from the daughter atoms which had been transferred by alpha particle recoil to the detectors when they were opposite the wheel. During the next 200 second period it was then possible to measure the half-life and energy of the daughter atoms free from the interference produced by those atoms produced directly during the bombardment. This cycle was repeated automatically with the beam being turned off during the daughter-measuring intervals. A composite spectrum of the mother and daughter activities is shown in Fig. 3. The activity from the 55-sec 8.10 MeV ^{254}No which was produced directly in good yield during the bombardment served to monitor the amount of nobelium that reached the surfaces of the crystals by a transfer mechanism of unknown origin; such transferred nobelium atoms were observed only in the first detector. The amount of ^{253}No daughter activity, as well as its energy and half-life, correspond quite well with the genetic sequence proposed.

Bombardments with ^{13}C ions of the ^{249}Cf target produced a somewhat similar activity but it was possible to differentiate it from the $^{257}104$ in several ways: (1) The complex alpha particle spectrum consists of two prominent groups with energies 8.77 and 8.86 MeV, and relative intensities 0.6 and 0.4, respectively; (2) the half-life is somewhat shorter and our best value so far is about 3 seconds; and (3) the excitation function and cross section correspond[8] much more closely to those of the $^{13}C,3n$ reaction to make $^{259}104$ than that of the ^{13}C,5n reaction to make $^{257}104$. Further evidence for the assignment to the former nuclide was obtained when the same type of daughter-measuring experiment as in the case of $^{257}104$ was carried out. A known isotope of nobelium, the 185-sec 8.11 MeV ^{255}No [4,10] was identified as the alpha recoil product in these experiments. More recently we have produced the 3-second 104^{259} by the ^{16}O,5n reaction in bombardments of ^{248}Cm.

Spontaneous fission branching decay was not found to be prominent for either one of these nuclides but in a different type of experiment we did uncover a new fission period which is probably due to $^{258}104$. We used an apparatus[11] consisting of a drum rotating next to the target in vacuum to catch the transmutation recoils directly. A peripheral series of mica detectors was placed to record the passage of fragments from spontaneous fission; after suitable etching their tracks could be observed optically.[12] The rotating drum was moved transversely to the direction of the beam to spread out the longer-lived fission activities and thus reduce the background. The speed of the drum was such that a 1 millisecond period could be easily measured. An activity with a half-life of 11 ± 2 milliseconds was observed when the ^{249}Cf target was bombarded by both ^{12}C and ^{13}C ions. Its excitation functions and yields are consistent with its being assigned to $^{258}104$ made by the ^{12}C,3n and ^{13}C,4n reactions and the half-life compares favorably with the 4 ms predicted by our empirical systematics.[5,11] Even though we have observed thousands of spontaneous fission events, because of the prevalence of isomers in the heavy element region which undergo this type of decay, we still are not certain of the correctness of this attribution. On the other hand, according to Nilsson,[13] the appearance of spontaneous fission isomers in this particular region of Z values is unlikely.

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Technicians at the HILAC for the design and fabrication of the apparatus; A. E. Larsh and A. A. Wydler for the electronic hardware used; R. G. Leres and D. F. Lebeck for the advanced computer software solutions; F. S. Grobelch and the HILAC operators for the excellent performance of the accelerator; and P. Shaw and C. Jung, our patient mica scanners.

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FOOTNOTES AND REFERENCES

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FIGURE CAPTIONS

Fig. 1. A series of alpha spectra of the activities produced by bombardment of ^{249}Cf with 71-MeV ^{12}C ions. The top spectrum is the sum of the individual spectra from the four detectors. The 8.60 MeV peak is probably due to ^{258}Lr; the peaks above that energy belong to ^{257}104 with the exception of the one at 8.87 MeV whose origin is uncertain.

Fig. 2. Excitation curves of several activities produced by bombardment of ^{249}Cf with ^{12}C ions. Note the high yield of the nobelium isotopes produced in ^{12}C,αxn reactions. There is an uncertainty in the energy scale that could be as much as 5 MeV.

Fig. 3. A set of spectra from the mother-daughter experiment which demonstrates the genetic relationship between ^{257}104 and ^{253}No. The spectra recorded by the individual crystal pairs are shown on top with the sum of the last three pairs at the bottom.

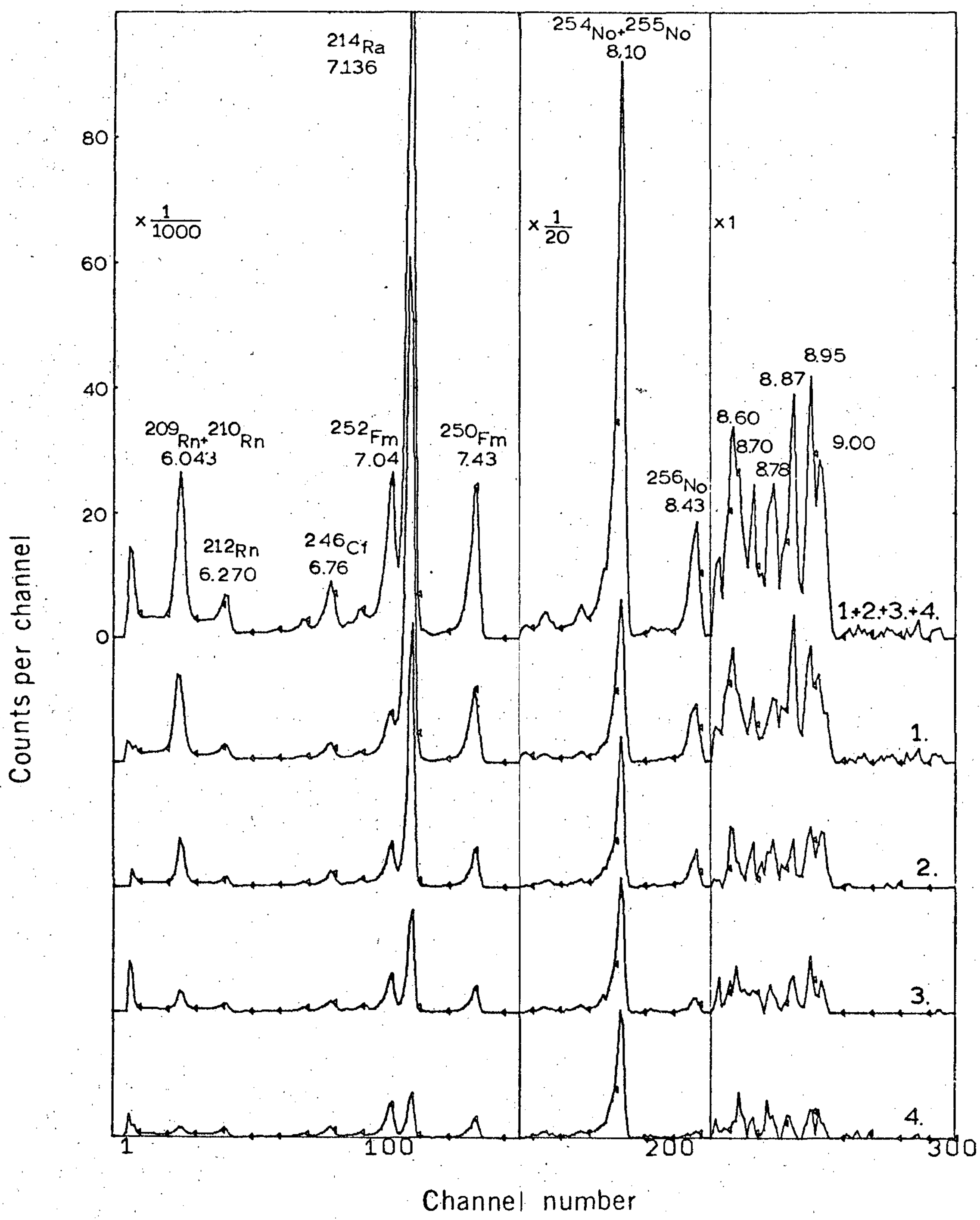
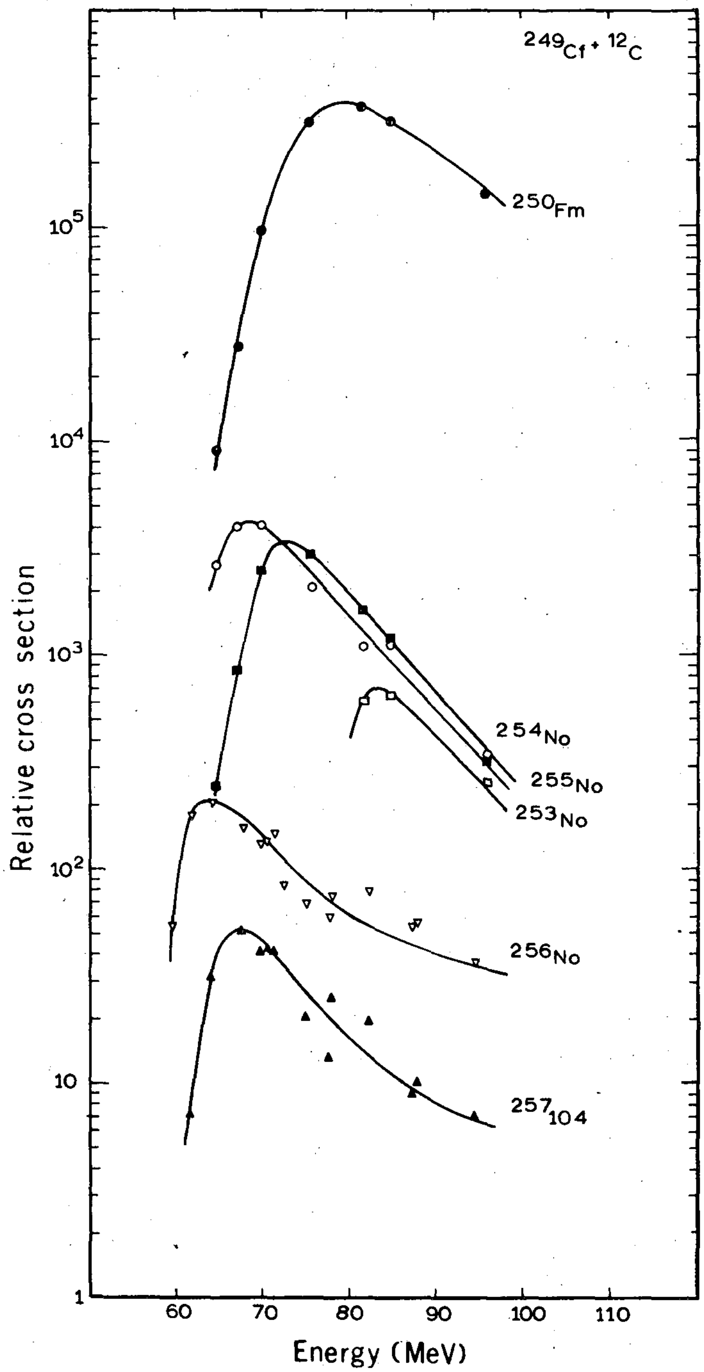

214Ra
7.136
254No+255No
8.10
80
60
40
20
0
x 1/1000
x 1/20
x1
8.95
8.87
8.60
8.70
8.78
9.00
209Rn+210Rn
6.043
252Fm
7.04
250Fm
7.43
256No
8.43
212Rn
6.270
246Cf
6.76
1+2+3+4.
1.
2.
3.
4.
Counts per channel
1
100
200
300
Channel number
XBL 694 4816

Fig. 1



XBL 694 4815

Fig. 2

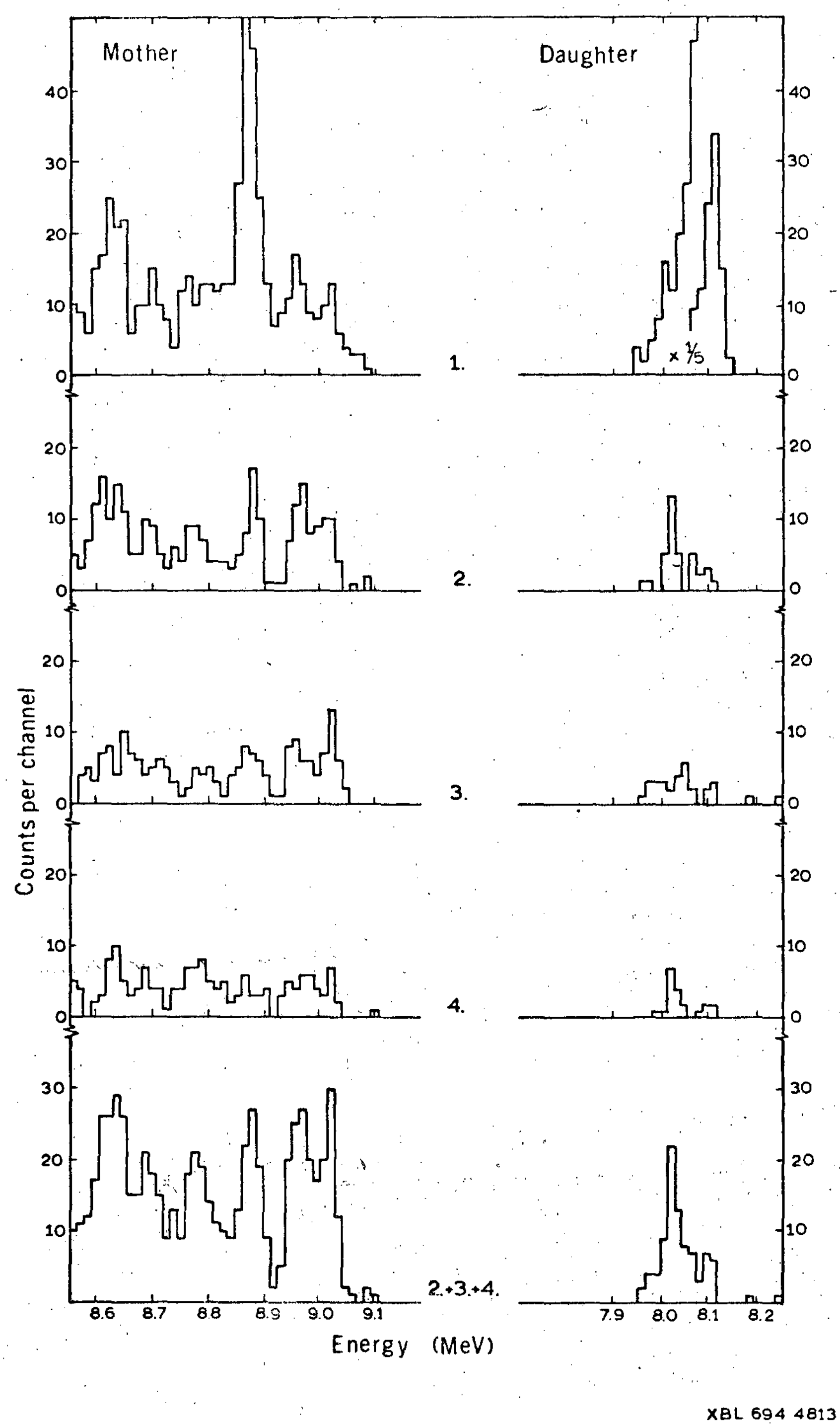

Mother
Daughter
× 1/5
1.
2.
3.
4.
2.+3.+4.
Counts per channel
Energy (MeV)
8.6
8.7
8.8
8.9
9.0
9.1
7.9
8.0
8.1
8.2
XBL 694 4813

Fig. 3

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