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

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(Received 5 May 1969)

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-sec 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-μg target of monoisotopic[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-μg/cm^2 target was bombarded mainly by ^{12}C and ^{13}C ions accelerated by the Berkeley heavy ion linear accelerator (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 μA dc (as completely stripped ions) were used typically and the total number of microampere hours 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-diam orifice into vacuum to be deposited onto the periphery of a 45-cm-diam 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 sec. 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 in-

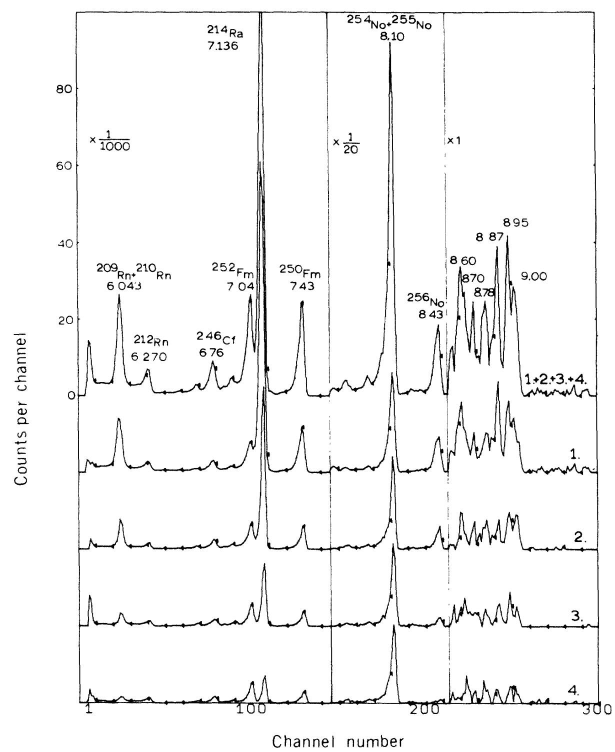

FIG. 1. A series of alpha spectra of the activities produced by bombardment of ^{249}Cf with 71-MeV ^{12}C ions. The top spectrum is the sum of the individual spectra from the four detectors. The 8.60-MeV peak is probably due to ^{258}Lr; the peaks above that energy belong to ^{257}104 with the exception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

tensities 0.15, 0.2, 0.3, and 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/μA h) 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, $4n$) to produce $^{257}104$ and is consistent with the calculations[8] of Sikkeland and Lebeck.

To make certain of the mass and atomic num-

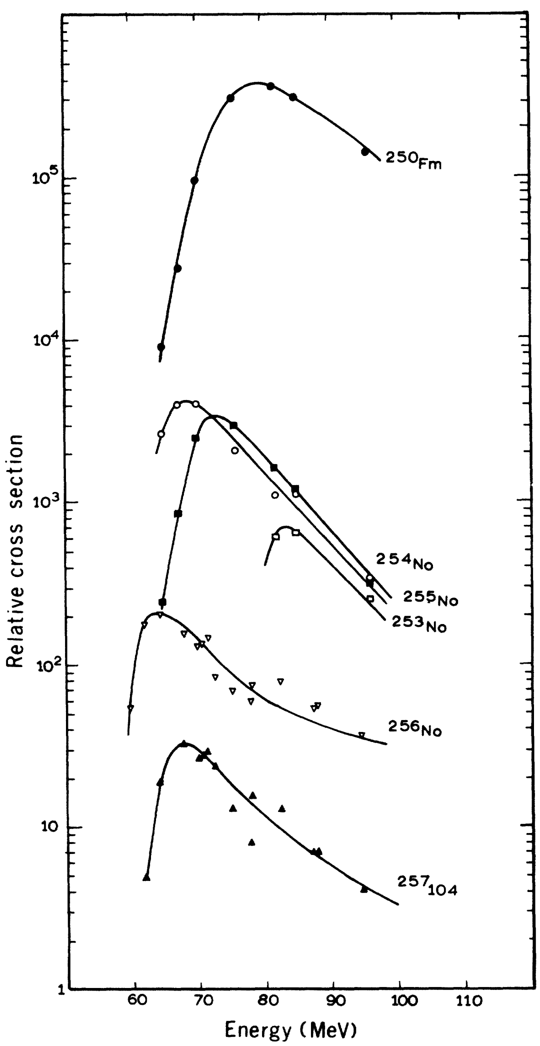

FIG. 2. Excitation curves of several activities produced by bombardment of ^{249}Cf with ^{12}C ions. Note the high yield of the nobelium isotopes produced in (^{12}C, αxn) reactions. There is an uncertainty in the energy scale that could be as much as 5 MeV.

ber 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 sec.[4,9] After a period of 200 sec 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 dectors when they were opposite the wheel. During the next 200-sec 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 sec; 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, 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-sec $^{259}104$ 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

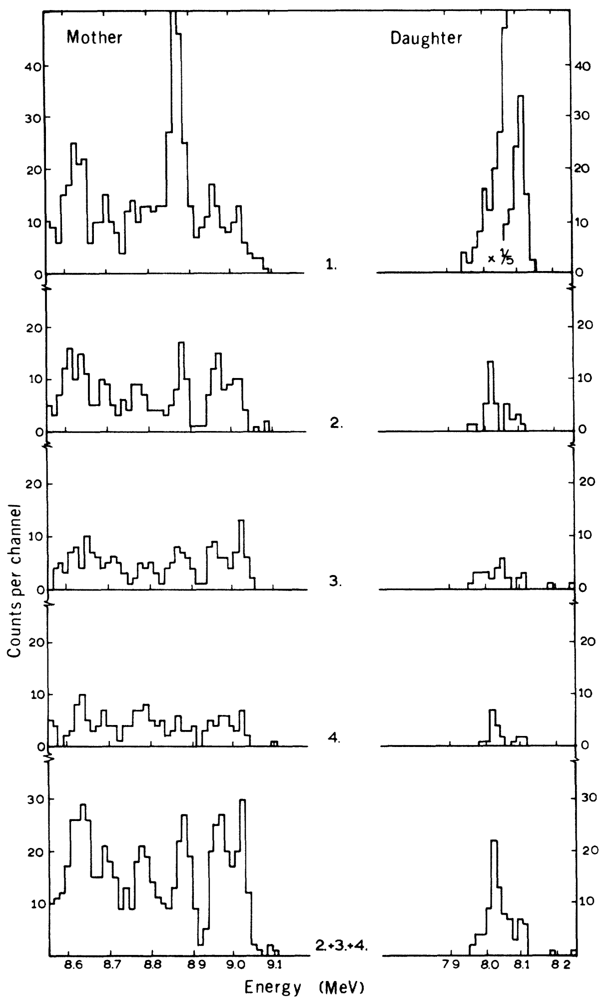

FIG. 3. A set of spectra from the mother-daughter experiment which demonstrates the genetic relationship between $^{257}104$ and ^{253}No. The spectra recorded by the individual crystal pairs are shown on top with the sum of the last three pairs at the bottom.

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-msec period could be easily measured. An activity with a half-life of 11 ± 2 msec 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 msec 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.

We gratefully acknowledge the very considerable assistance afforded by the following people in making these experiments possible: R. M. Latimer in the chemical processing of the target material, C. A. Corum and the mechanical 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. We appreciate the very important help in the initial stages by T. Sikkeland and are very pleased by the interest and encouragement shown to us by G. T. Seaborg. One of us (P.E.) is indebted to the National Research Council for Sciences, Helsinki, Finland, for financial support.

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*This work was done under the auspices of the U. S. Atomic Energy Commission.

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

‡Guest Scientist supported by the National Research Council of Sciences, Helsinki, Finland.

[1]A. Ghiorso, M. Nurmia, and J. Harris, University of California Lawrence Radiation Laboratory Report No. UCRL-18714, 1969 (unpublished).

[2]G. N. Flerov, Yu. Ts. Oganesyan, Yu. V. Lobanov, V. I. Kuznetsov, V. A. Druin, V. P. Perelygin, K. A. Gavrilov, S. P. Tretiakova, and V. M. Plotko, At. Energ. (U.S.S.R.) 17, 310 (1964) [translation: Soviet J. At. Energy 17, 1046 (1964)], and Phys. Letters 13, 73 (1964).

[3]The target material was milked from several hundred micrograms of Bk^{249} at the transuranium processing plant at Oak Ridge National Laboratory by D. Ferguson et al., to whom we would like to express our gratitude.

[4]A. Ghiorso, T. Sikkeland, and M. J. Nurmia, Phys. Rev. Letters 18, 401 (1967).

[5]A. Ghiorso and T. Sikkeland, Phys. Today 20, No.9,

25 (1967).
[6]Manufactured by Oak Ridge Technical Enterprises Corporation, Oak Ridge, Tenn.
[7]J. T. Routti and S. G. Prussin, University of California Lawrence Radiation Laboratory Report No. UCRL-17672, 1968 (unpublished).
[8]T. Sikkeland and D. F. Lebeck, unpublished work.
[9]V. L. Mikheev, V. I. Ilyushchenko, M. B. Miller, S. M. Polikanov, G. N. Flerov, and Yu. P. Kharitonov, At. Energ. (U.S.S.R.) 22, 90 (1967) [translation: Soviet J. At. Energy 22, 93 (1967)].
[10]G. N. Flerov, S. M. Polikanov, V. L. Mikheev, V. I. Ilyushchenko, V. F. Kushniruk, M. B. Miller, A. M. Sukhov, and V. A. Shchegolev, Yadern Fiz. 5, 1186 (1967) [translation: Soviet J. Nucl. Phys. 5, 848 (1967)].
[11]M. Nurmia, K. Eskola, P. Eskola, and A. Ghiorso, University of California Lawrence Radiation Laboratory Report No. UCRL-18712, 1969 (unpublished).
[12]P. B. Price and R. M. Walker, Phys. Rev. Letters 3, 113 (1962).
[13]S. G. Nilsson, University of California Lawrence Radiation Laboratory Report No. UCRL-18355, Revised 1968 (unpublished).

EVIDENCE FOR DISCOVERY OF GRAVITATIONAL RADIATION*

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Coincidences have been observed on gravitational-radiation detectors over a base line of about 1000 km at Argonne National Laboratory and at the University of Maryland. The probability that all of these coincidences were accidental is incredibly small. Experiments imply that electromagnetic and seismic effects can be ruled out with a high level of confidence. These data are consistent with the conclusion that the detectors are being excited by gravitational radiation.

Some years ago an antenna for gravitational radiation was proposed.[1] This consists of an elastic body which may become deformed by the dynamic derivatives of the gravitational potentials, and its normal modes excited. Such an antenna measures, precisely, the Fourier transform of certain components of the Riemann curvature tensor, averaged over its volume. The theory has been developed rigorously, starting with Einstein's field equations to deduce[2] equations of motion. Neither the linear approximation nor the energy-flux relations are needed to describe these experiments, but their use enables discussion in terms of more familiar quantities. All aspects of the antenna response and signal-to-noise ratio can be written in terms of the curvature tensor. The theory was verified experimentally by developing a high-frequency source[3] and producing and detecting dynamic gravitational fields in the laboratory.

Several programs of research are being carried out. One employs laboratory masses in the frequency range 1-2 kHz.[4] Another is concerned with expected gravitational radiation from the pulsars.[5] Some designs for such antennas suggest a pulsar detection range approaching 1000 pc. A third class of antennas employs the quadrupole modes of the earth,[1] the moon, and planets[6] for the range 1 cycle/h to 1 cycle/min. This array is a new set of windows for studying the universe.

Search for gravitational radiation in the vicinity of 1660 Hz.—A frequency in the vicinity of 1660 Hz was selected because the dimensions are convenient for a modest effort and because this frequency is swept through during emission in a supernova collapse. It was expected that once the technology was refined, detectors could be designed for search for radiation from sources with radio or optical emission, such as the pulsars. A knowledge of the expected frequency and Q of a source enormously increases the probability of successful search.

However, occasional signals were seen at 1660 Hz and small numbers of coincidences were observed on detectors[7,8] separated by a few kilometers. To explore these phenomena further, larger detectors were developed. One of these is now operating at Argonne National Laboratory. My definition of a coincidence is that the rectified outputs of two or more detectors cross a given threshold in the positive direction within a specified time interval. For the present experiments the time interval was 0.44 sec. The magnitudes of the outputs at a coincident crossing enable computation of the probability that the coincidence was accidental. Observation of a number of coincidences with low probability of occurring