

Lawrence Berkeley National Laboratory

Recent Work

Title

THE BERKELEY HILAC HEAVIEST ELEMENT RESEARCH PROGRAM

Permalink

<https://escholarship.org/uc/item/55k818vk>

Author

Ghiorso, Albert.

Publication Date

1970-04-01

Submitted to Proceedings of the
Robert A. Welch Foundation Conferences
on Chemical Research. XIII. The
Transuranium Elements, Houston, Texas,
November 17-19, 1969

UCRL-18633
Preprint

c.2

THE BERKELEY HILAC HEAVIEST ELEMENT
RESEARCH PROGRAM

RECEIVED
LAWRENCE
RADIATION LABORATORY

MAY 22 1970

LIBRARY AND
DOCUMENTS SECTION

Albert Ghiorso

April 1970

AEC Contract No. W-7405-eng-48

TWO-WEEK LOAN COPY

*This is a Library Circulating Copy
which may be borrowed for two weeks.
For a personal retention copy, call
Tech. Info. Division, Ext. 5545*

LAWRENCE RADIATION LABORATORY
UNIVERSITY of CALIFORNIA BERKELEY

UCRL-18633

4

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

THE BERKELEY HILAC HEAVIEST ELEMENT

RESEARCH PROGRAM*

Albert Ghiorso

Lawrence Radiation Laboratory
University of California
Berkeley, California

Dr. Ghiorso (University of California Lawrence Radiation Laboratory, Berkeley, Conference Speaker): Thank you, Glenn.

I feel very honored to be giving this talk today as a participant in this celebration of the 100th Centenary of the Mendeleev Periodic System. I was fortunate to be one of those who participated in the discovery¹ of element 101 and I might recall that we named this new element mendelevium at a time when perhaps it was not the popular thing to do. Nonetheless, the name was an excellent choice and has received universal acceptance.

Today I would like to talk about a new element (at least we in Berkeley consider it a new element!) and some of the consequences of our recent work.

Now, this is obviously a very complicated subject and many of you are not familiar with the intimate details of this kind of work, so I have tried to put together this paper in such a way that it will expose you to the maximum amount of information that would be meaningful. I am going to try to do this by using both the transparency projector and slides, and at times they will be on simultaneously. I hope this will make it a little easier for those of you who want to study the detailed data.

Well, let me start out by reminding you of my colleagues who are involved in this work. These are Matti Nurmia and James Harris of the Lawrence Radiation Laboratory and Kari and Pirkko Eskola, a husband and wife

team on leave from the University of Helsinki, Finland. Robert Silva of the Oak Ridge National Laboratory joined us temporarily to help on the chemistry experiments with element 104 which I will describe later.

My talk will cover, first, the element 104 alpha emitters which we have found during the last year and a half;² second, a description and movie of some very recent chemistry experiments which have been completed successfully using a longer-lived isotope of element 104 that we have discovered recently; and third, our work on spontaneous fission emitters which seems to fail to confirm the older claims made by the Dubna group to the discovery of an isotope of element 104.³

A. TARGET MATERIALS USED IN HEAVY ION BOMBARDMENTS

Figure 1 - Heavy-Nuclide Chart

I will start by discussing possible target materials for use in transmutation experiments to make element 104. The chart you see here shows only the heavy elements. The important target elements are made ordinarily by reactor-neutron buildup. Basically, the method starts with the very abundant nuclide, ²³⁹Pu, and builds the heavier elements up by a process of neutron capture and beta decay. The outstanding work in making the intense neutron irradiations and the necessary accompanying chemical separations is being done by the ORNL High Flux Isotope Reactor and TRU facilities in collaboration with the Savannah River Laboratory.

One can progress far up the table--up to ²⁵⁷Fm--but that is as far as one can go with reactors. Along the way, one has some very interesting target nuclides. ²⁴²Pu and ²⁴⁴Pu, which are long-lived, are very good building materials for elements below atomic number 104. Americium, element 95, has two

useful isotopes suitable for targets, ^{241}Am and ^{243}Am , but they are deficient in neutrons when one compares them with the curium isotopes that are available. Curium has a large number of isotopes that can be used, all the way from 5-month ^{242}Cm to long-lived ^{248}Cm . It is this latter isotope, ^{248}Cm , that we have bombarded with ^{18}O ions to make a 70-second alpha emitter, $^{261}_{104}\text{Po}$, for use in our chemistry experiments.

^{250}Cm would be a very desirable target material, but unfortunately it is difficult to make in useful quantities with reactor neutrons. Recently, in a sense, it became available--unfortunately it is down about 2000 feet in Nevada! There is a fair amount of it there, but unhappily for us at the moment, it is very expensive to recover. As George Cowan will describe, it was produced in a nuclear explosion. It is a fairly rich material--about ten per cent in mass relative to the other curium isotopes. It would be very interesting to bombard this material with ^{48}Ca and it is my candidate for being the most useful for making nuclides in the never-never land up there around element 114. But whether we get any of it or not is simply a matter of money, and we will have to see Dr. Seaborg about that! No small amount either. The cost is up in the millions of dollars.

The next good target material one gets to is ^{249}Bk with a half-life of almost a year. Unfortunately, it is not very abundant--it tends to be allowed to beta-decay into the more useful form of this mass as californium. ^{249}Cf is the target material which we used initially to make the isotopes $^{257}_{104}\text{Pu}$, $^{258}_{104}\text{Pu}$ and $^{259}_{104}\text{Pu}$. We would like to use ^{250}Cf , except that it is presently mixed with the 2.6-year ^{252}Cf , which has a 3% decay branch by spontaneous fission. This produces the nuisance that it is also an intense neutron emitter and thus very hard to handle.

When one gets up to element 99, einsteinium, we find that the most abundant isotope, ^{253}Es , has a half-life of only 20 days. Experiments become exceedingly difficult but they can be done when the results warrant it. ^{254}Es is longer-lived, but we do not yet have this nuclide in microgram quantities. ^{255}Es is short-lived, only 40 days, and again we do not really have very much of it yet.

Eighty-day ^{257}Fm is the heaviest nuclide that is obtainable from reactors, but the amount will probably always be miniscule. The best source for this material is a nuclear explosion with the aforementioned recovery difficulties.

B. SOME PROBLEMS ENCOUNTERED IN THE SEARCH FOR NEW ELEMENTS

As a veteran isotope hunter among the heavy elements, I can assure you that, in general, one prefers to look for alpha emitters. This comes about because an alpha-particle decay with its distinctive energy and half-life can be very specific in identifying the particular nuclide that emitted the alpha particle. There are overlapping cases where one has to worry about impurities, but by and large, one alpha particle, I used to say, was worth a hundred spontaneous fissions. Fission decay is not very specific.

Now I must raise this estimate by another factor of a hundred-- thus making one alpha particle worth ten thousand fissions! At least, we have produced thousands of fissions of some activities and we find that even after working very diligently, we still are not really sure of their atomic and mass numbers. On the other hand, a few tens of alpha-particle decays have been sufficient to characterize many nuclides with commendable accuracy.

One of the most fundamental problems that has to be faced is that

caused by the very small cross section that is typical for the production of a new heavy nuclide by means of heavy-ion bombardment of a heavy-element target. The reason that the production cross section is small is that it has to compete with fission at each stage of a reaction where neutrons are emitted in a cascade. For example, let us take a heavy ion--say ^{18}O --bombarding a curium nucleus. The resulting neutron-rich compound nucleus of element 104 can have a chance of either evaporating a neutron and going to the next stage or being destroyed by fission. The competition is very rough--about one per cent survive at each step. If very many neutrons have to be evaporated, the losses are excessive and one has to contend with cross sections of 10^{-33} cm^2 , a nanobarn. With a nanobarn cross section experiments can be performed with modern techniques, but it is not the easiest thing in the world. One does not get a lot of activity!

The complexity of the interactions, of course, can become formidable. We find reactions where not only neutrons are evaporated but also alpha particles and protons, or indirect reactions where effectively alpha particles or protons are emitted. Consequently many nuclides can be produced simultaneously both among the heavy elements and among the lighter elements in the region of 126 neutrons between lead and uranium. These "light" alpha emitters become a problem because certain decay chains give rise to alpha activities that resemble the heavy ones in energy and half-life. They can appear in these experiments from reactions with impurities of mercury, thallium, lead, and bismuth at the nanogram level in the target so that special care has to be taken in the purification of the target material.

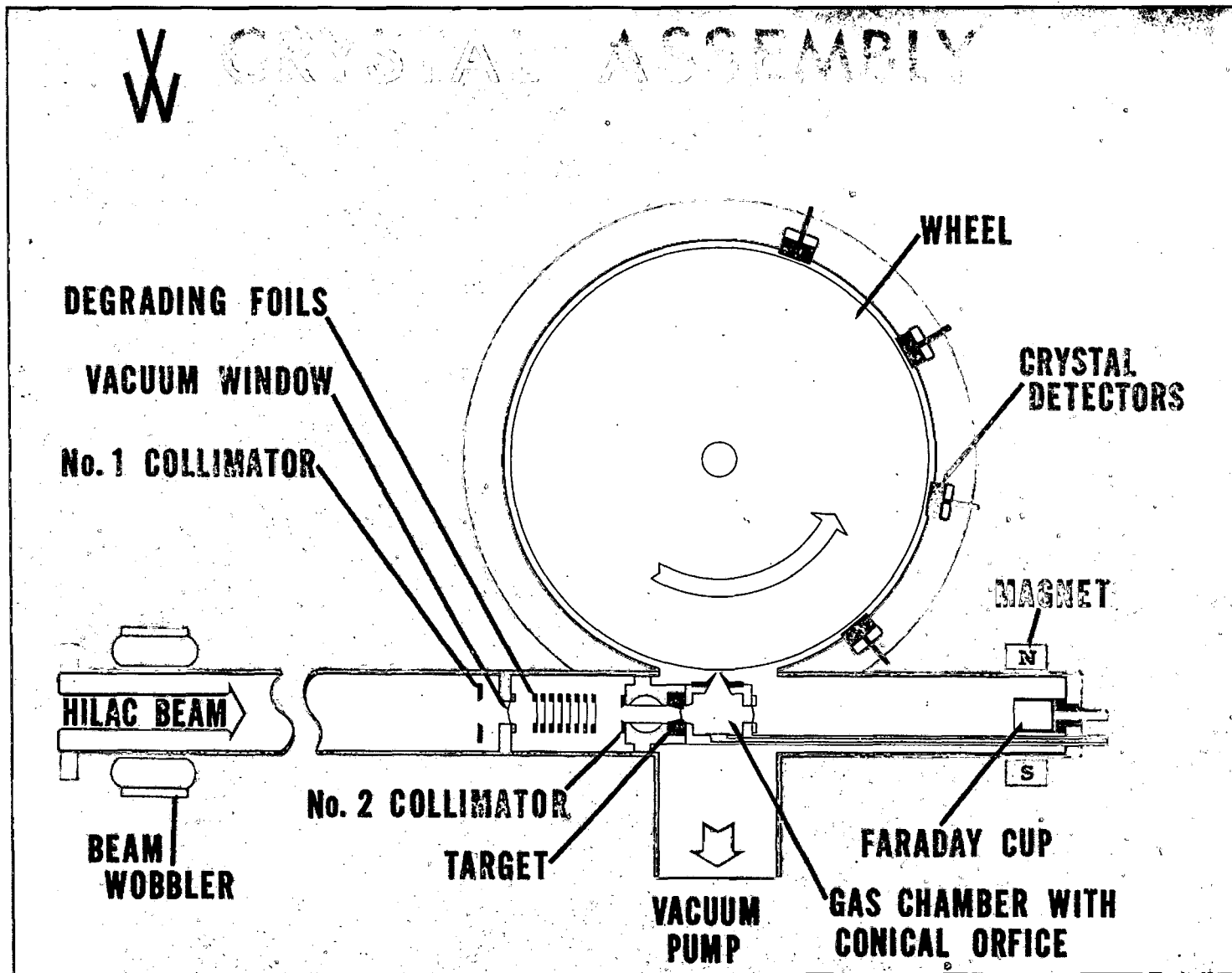
The other approach to looking for a new element is to try to find a spontaneous-fission emitter with a distinctive half-life. The 14-ms

nuclide, ^{242}Am , was discovered in this way by the people at Dubna when they bombarded ^{242}Pu with neon ions hopefully trying to make an isotope of element 104. Of course, it was a tremendous disappointment for them when it turned out to be an isomer of americium. Because of this extremely important find, the usefulness of spontaneous fission was degraded terribly as an indication of atomic number. Since that time, many more fission isomers have been found, and it has become a very interesting field in its own right. The Russians well deserve the credit for uncovering this new type of isomerism, and as Dr. Strutinsky and Dr. Björnholm will show, there are some very important consequences stemming from this research. Later in this paper I will return to the problem of the Russian work on a presumed element-104 spontaneous-fission emitter.

Figure 2 - Vertical wheel system for alpha emitters.

Let me now describe our present method for detecting the alpha-particle emitters that are produced by heavy-ion bombardment of heavy-element targets. The method shown here has become standard for those activities with half-lives longer than a few milliseconds. The particle beam, coming from the left, passes through a series of degrader foils which lower the energy of the beam, and then through the target chamber to be brought to rest in a Faraday cup so as to provide an integrated measure of the number of beam particles. In our case, the Hilac beam energy is 10.4 MeV per nucleon and we typically desire to degrade it to around 5 to 7 MeV per nucleon. Passing through the target, it ejects the recoils of element 104 into a chamber which is filled with helium gas at a pressure of a few hundred torr. The range of these atoms is of the order of a centimeter or two for a reaction produced by oxygen ions.

The recoils stopped in the gas are then carried by the gas stream



CBB 689-5712

Fig. 2. Vertical wheel system for alpha emitters.
This is a schematic assemblage of our four crystal-

through a small orifice into a vacuum maintained at a fraction of a torr by a large mechanical blower pump. The gas moves very rapidly and, depending upon the target-chamber dimensions and the size of the orifice, the time for a recoil atom to move from the point where it comes to rest after ejection from the target to its final resting place on the collector can vary from ~1-100 msec.

After going through the orifice the atoms strike the wheel. In vacuum, under experimental conditions which can sometimes be very mysterious, either all will stick or perhaps as little as thirty per cent of them. We have experienced a large variation with different techniques. At the moment we believe that we are approaching 100% efficiency but are not certain why. It seems to depend to some extent on what impurities are in the gas. One explanation that we can offer is that a very heavy conglomerate of the element-104 recoil and an oil droplet, for instance, has a mass that is so very heavy that the particle can be carried through the orifice and stick to the wheel with a high efficiency. We have observed that if the gas is very pure the efficiency is much less. It is a very ticklish kind of thing and can depend upon outgassing of surfaces, etc.

After a given period of deposition, the wheel is cyclically rotated very swiftly to a position where the alpha-energy spectrum emitted by the recoils can be measured with a solid-state detector.* The number of events with a given energy and their decay are measured during the interval of time that elapses until the next group of atoms is placed in front of the crystal detector. The angular rotation of the wheel is chosen such that the same point of deposition on the wheel is not repeated until all 239 others have been used.

* The detectors were manufactured to our specifications by ORTEC. These semiconductor devices were made of silicon with a gold surface barrier and had an 8 mm diameter and a depletion layer thickness of 100 microns. The lip of the supporting ceramic ring was kept very small so that the crystals could be mounted with their faces close (~0.7 mm) to the wheel periphery. The detector performance was excellent--being only marred by fast-neutron degradation. When the resolution had become degraded to a noticeable extent, the crystals were easily replaced.

By using a number of detectors around the periphery of the wheel, each group of atoms can be measured that number of times and so provide further decay data. The slide is a schematic version of our old four crystal-position system. The equipment has now been modified to provide five crystal positions. Later I will show a slide of that system which demonstrates how we use twenty crystals to record the milking of the alpha-recoil daughters coming from decay of isotopes of element 104.

Figure 3 - Nuclide chart in the 126-neutron region.

The impurity problem that I referred to earlier can be very taxing at times. Fortunately, once it is known that this is a problem it can be taken into account if its magnitude is not too intense. As an example, one of the activities that is found is ^{211m}Po , the 25-second isomer which has an alpha energy (8.87 MeV) in the range that we are searching, but fortunately has a 25-second half-life. It also has a very prominent group at 7.27 MeV which can be used to identify how much of an 8.87 MeV activity it is responsible for.

One also makes ^{212m}Po , a 46-second isomer with the very high alpha-particle energy of 11.65 MeV and, again, it can be used as a measure of the amount of lead or bismuth in the target. Other ways of determining the extent of light impurities can be utilized--such as, measuring the various radium and francium isotopes produced by ^{12}C or ^{11}B ion bombardment.

We have recently uncovered another problem that has plagued us in these experiments. It is a phenomenon which we do not yet understand that we call "self-transfer." We find that, in vacuum, a fraction of the atoms on the wheel, of their own volition, will leave the wheel and go over to a facing crystal. We have observed this phenomenon in the case of ^{250}Fm and ^{254}No . Only a few per cent of the atoms do this but we do not understand why it happens at all. Qualitatively, the effect does not seem to depend upon the

surface. It happens from plastic, platinum, or aluminum surfaces. It does not depend upon the carrier gas; it happens to the same extent with hydrogen as with helium. It does not depend on the amount or surface density of the activity. We have changed the pressure in the general region near the crystals without seeming to affect it. It is simply that some of these atoms spontaneously seem to leave the collecting surface. If the wheel is not moved, the effect is not observed, so obviously it is not caused by direct collection of the activity on the crystals from the gas jet. On the other hand, the effect is observed simply by quickly removing a crystal which is facing the wheel and counting it away from the wheel. The interesting thing about this phenomenon is that its apparent "half-life" in the case of fermium is about two seconds; but in the case of ^{254}No , where we have also observed it, the half-life is only a fraction of a second. We have not observed the self-transfer of the prominent activities, ^{214}Ra and ^{213}Fr . The problem was troublesome to us because in certain cases it could simulate the legitimate transfer of daughter atoms recoiling as a result of the alpha decay of element 104 mother atoms. The odd half-life effect also troubled us because at first we attributed it to a large loss of activity from the wheel. We worried that we were measuring half-lives properly. We were reassured when we found that over a wide range of half-life we obtained the same periods for well-known nuclides as those which had been measured by other techniques.*

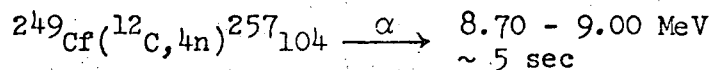
This then is the method with some of its problems. Let us pass on to the results obtained.

* Subsequent to the Conference we have performed new experiments which seem to indicate that the transfer effect is caused by nuclear recoil from isomeric transitions to the ground state in ^{250}Fm and ^{254}No .

C. THE ELEMENT 104 ALPHA EMITTERS

Figure 4 - Alpha-particle energy versus neutron number.

This is a standard kind of alpha-particle energy versus neutron-number plot for the heaviest even-Z elements and demonstrates the typical behavior around the 152-neutron subshell. When this slide was made some years ago, these were the predicted positions for element 104, and I will show a slide later on of where they fit in now.

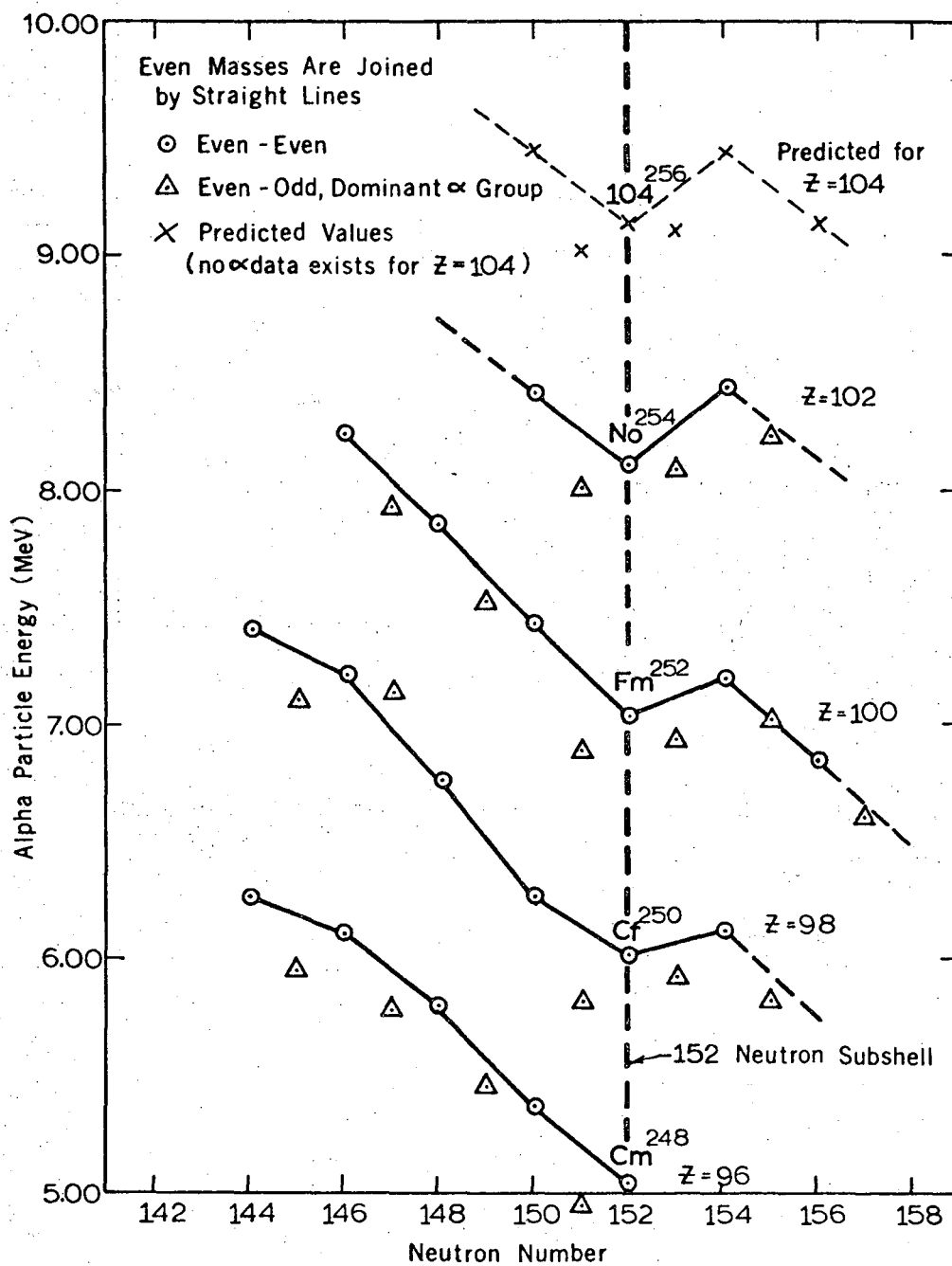


This is the reaction used in the experiments first performed in July of 1968 when we observed an alpha-emitting isotope of element 104. The latest target used has 60 micrograms of ^{249}Cf in a circle 3/16 inch in diameter. Two other targets were used before this one which gave similar results. We find a very complex alpha-particle structure in the decay of $^{257}_{104}$ with groups at 8.70, 8.78, 8.95, and 9.00 MeV.

Figure 5 - Alpha spectra from $^{257}_{104}$.

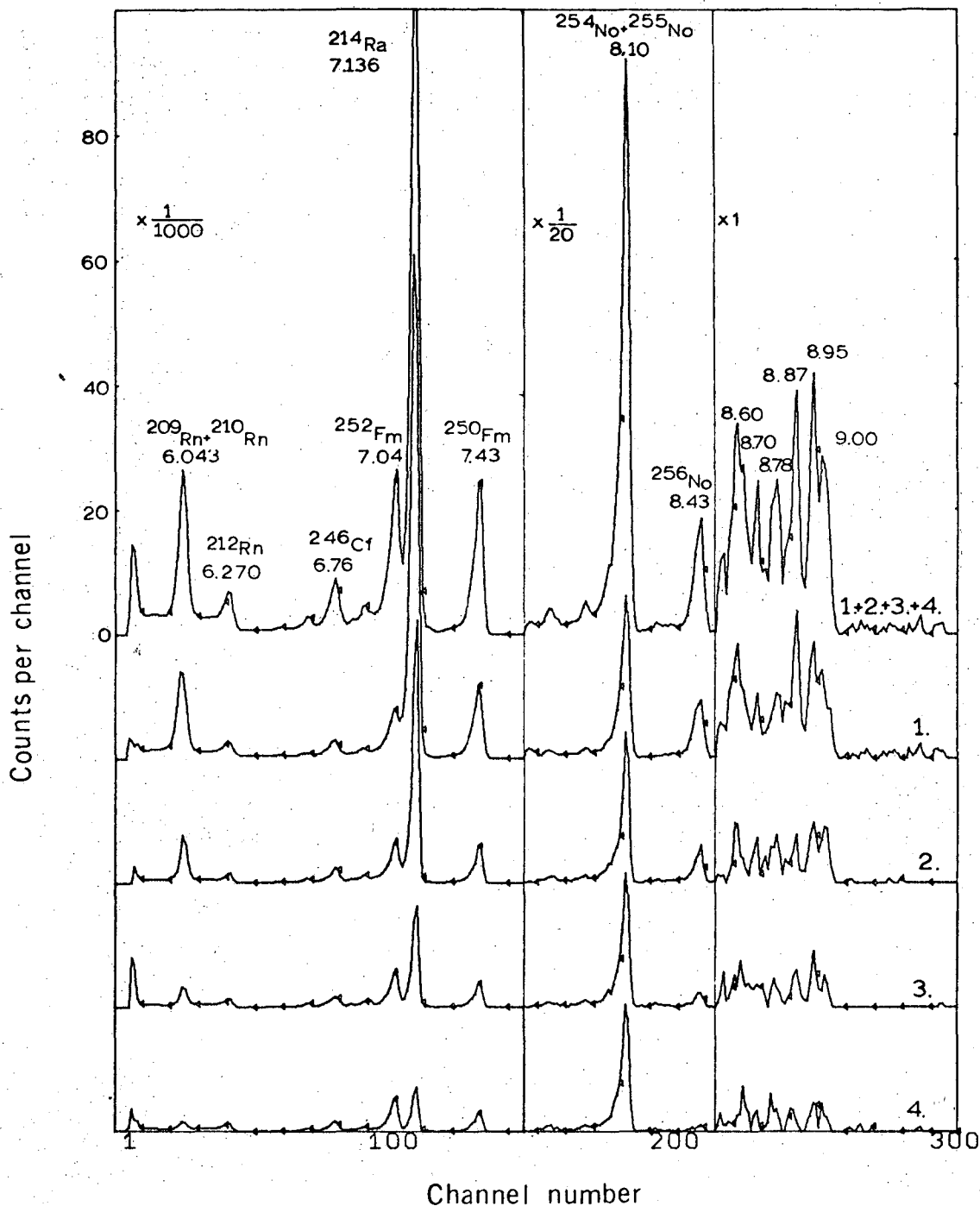
The cross section, 10 nanobarns (10^{-32} cm^2), is relatively large by our present standards. We observe about 30 alpha counts per hour at a beam level which dares not be increased by a factor of one and one-half without risking the destruction of the target. You can see that we are pushing our sensitivity to the absolute maximum. But a production rate of 30 counts per hour of distinctive alpha-energy groups produces a substantial amount of activity in a day.

The energy resolution fortunately is quite good. Typically we obtain about 30 kilovolts full-width at half-maximum in these experiments. The half-life for $^{257}_{104}$ is quite distinctive and the peaks stand out against the



XBL6732028

Fig. 4. Alpha-particle energy versus neutron number for the heavy even-Z elements.



XBL 694 4816

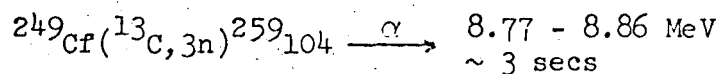
Fig. 5. A series of alpha spectra of the activities produced by bombardment of ^{249}Cf target with ^{12}C ions. The wheel was cycled at a rate of 3 seconds per crystal position. The spectrum from each of the four crystals along with their sum is shown. The total number of microampere hours was 81, measured as fully stripped ions. The 8.60-MeV peak is due to ^{258}Lr .

small 8.87-MeV $^{211\text{m}}\text{Po}$ background produced from the lead impurity. The stand-in for this experiment was a ^{248}Cm target which had a similar amount of lead as determined by ^{11}B experiments to make Fr isotopes. When the ^{248}Cm target is substituted in the $^{249}\text{Cf} + ^{12}\text{C}$ experiment, we find not the element-104 alpha emitter but instead the 3-second ^{256}No activity with an entirely different alpha spectrum.

Figure 6 - Energy levels from ^{253}Fm , ^{255}No , and $^{257}_{104}$ data.

The spectrum that we have obtained for $^{257}_{104}$ shows these particular levels. Shown for comparison are nuclides which are similar. We have not tried to interpret this yet, but it looks quite interesting.

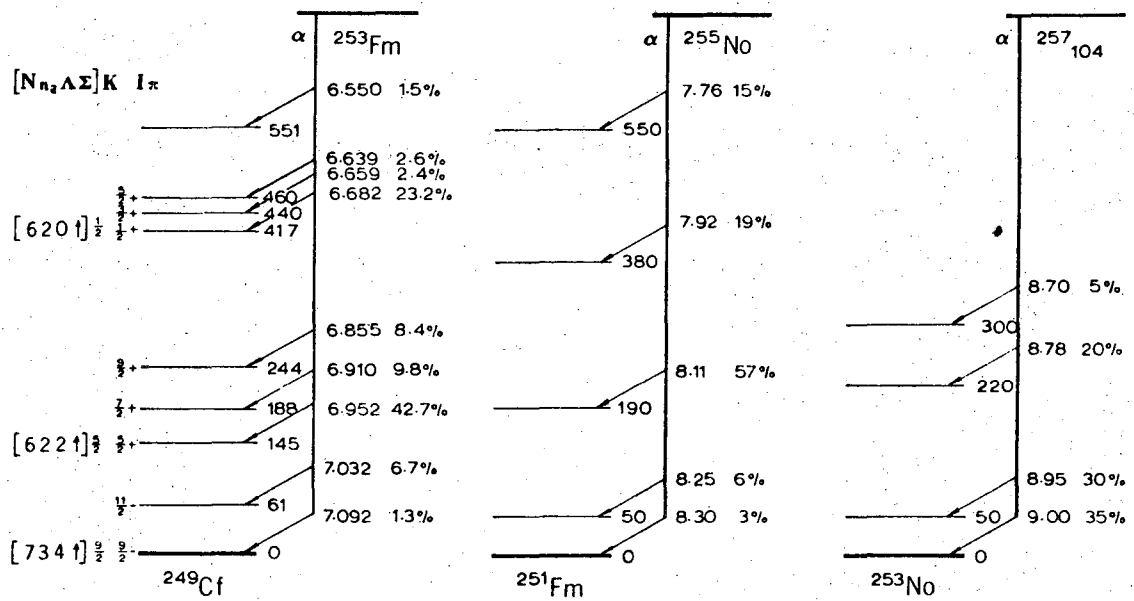
Figure 7 - Alpha spectra from $^{259}_{104}$.



In this next experiment we expected to make $^{259}_{104}$, and we do find principally these two alpha-particle groups which we believe belong to that isotope. We have also made oxygen bombardments of the ^{248}Cm target which tend to show the same spectrum and thus confirm it. These data are not yet as good as that for $^{257}_{104}$, but they are consistent and the spectrum looks definitely different from that for $^{257}_{104}$. We believe its proper assignment to be to the isotope $^{259}_{104}$.

The 8.6-MeV peak that is present in these runs is very interesting. In other experiments involving ^{248}Cm plus ^{15}N and ^{14}N ions, we have made this same 4-second activity and we believe it to be due to ^{258}Lr .

In the 1961 element 103 discovery experiments ⁴ we found an 8.6 MeV alpha emitter and reported a half-life of about 8 seconds. At that time we bombarded a mixture of californium isotopes with masses 249-252 with ^{11}B and



XBL 698 4885

Fig. 6. Energy levels from ^{253}Fm , ^{255}No , and $^{257}_{104}$ data.

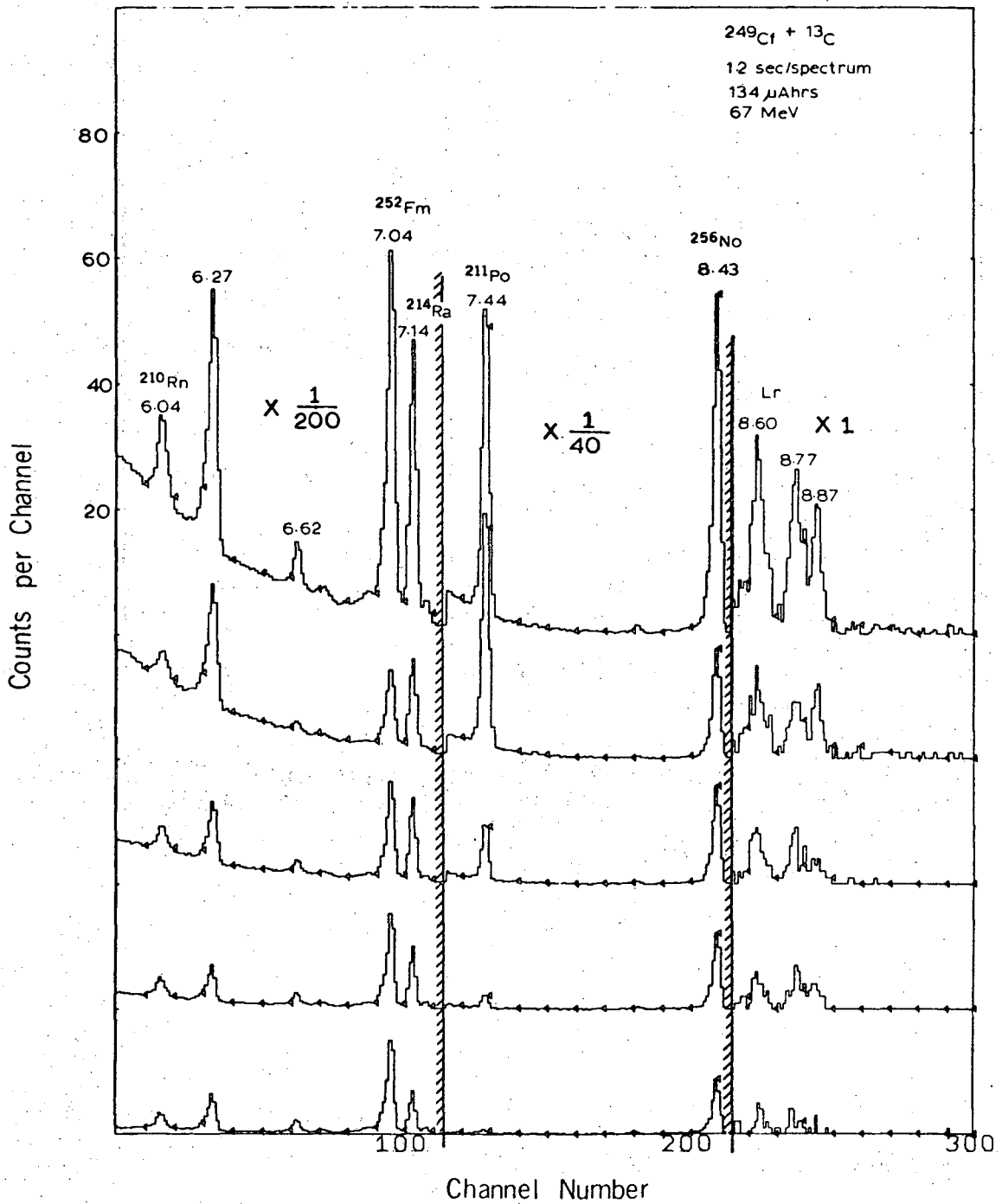


Fig. 7. Alpha spectra from $^{259}_{104}$ produced by the $^{249}_{98}\text{Cf}(^{13}_6\text{C}, 3n)$ reaction. The wheel-cycle rate was 1.2 seconds per crystal.

^{10}B ions and we have not repeated those experiments since. This was the first attempt to do it in a different way and the results indicate that this is the nuclide that we were dealing with at that time. We have not completed the new lawrencium experiments and feel that there could be ^{259}Lr mixed in here with a similar or longer half-life and this would complicate the interpretation of the 1961 results. We have not had time to go back to it yet.*

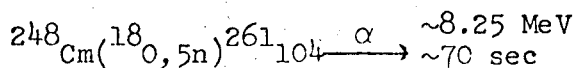
Figure 8 - Alpha spectra for $^{259}_{104}$ produced via $^{248}_{\text{Cm}}(^{16}_0, 5n)$ reaction.

The next slide shows you the same spectrum but without the complication of the lawrencium isotope. This is $^{248}_{\text{Cm}}$ plus $^{16}_0$ and we find that the $^{259}_{104}$ spectrum is much cleaner.

Figure 9 - Excitation function for $^{259}_{104}$ produced via $^{248}_{\text{Cm}}(^{16}_0, 5n)$ reaction.

The next slide is the excitation function for the $^{259}_{104}$. Notice that we also make isotopes of nobelium with very good yield. One can make large amounts of nobelium in this way and it provides a good method for studying nobelium isotopes.

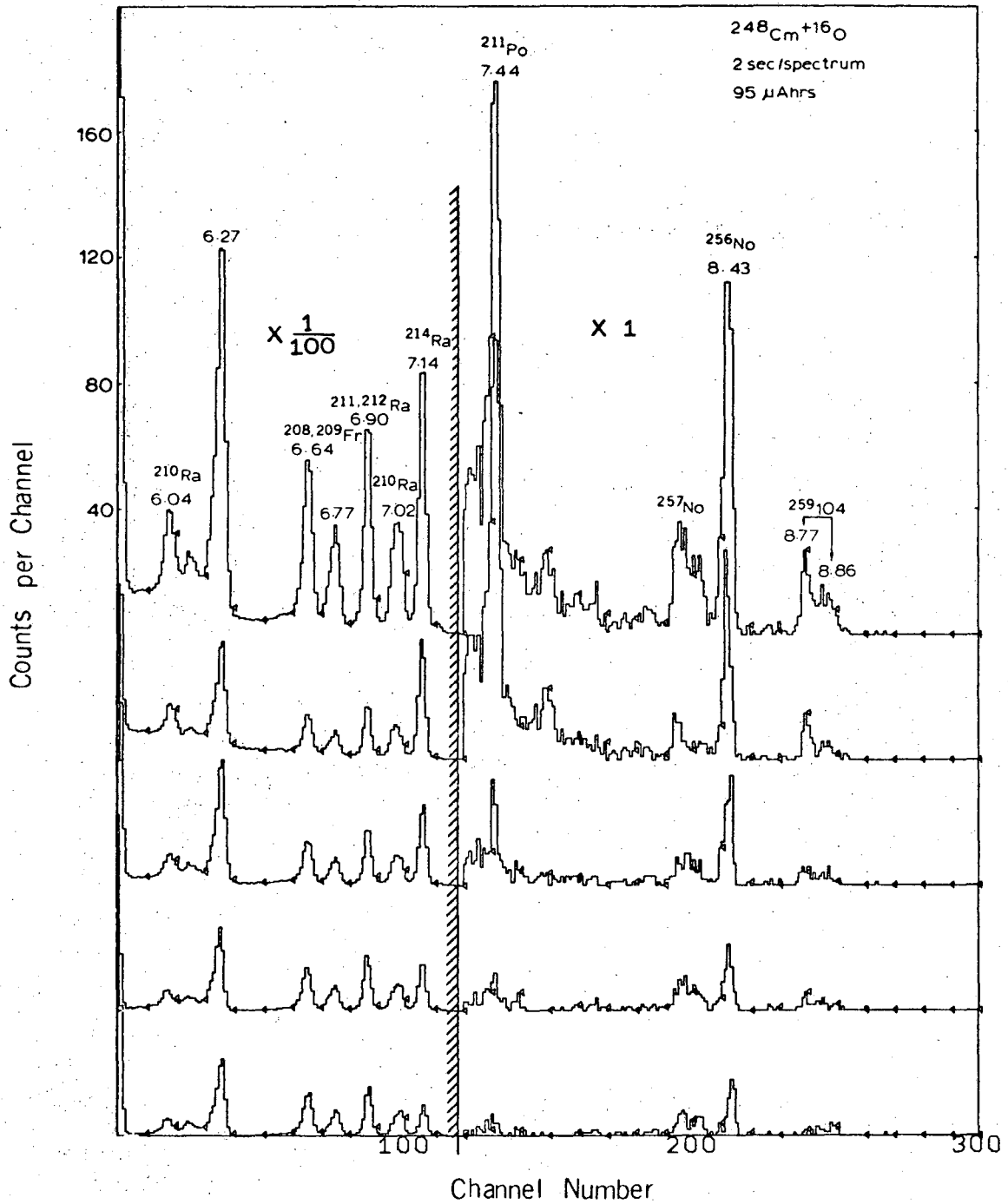
The two methods of producing $^{259}_{104}$ are consistent. They are not as elegant as we would like, but we have not had time to complete them properly. The final proof of the assignments was furnished by alpha-recoil milking experiments which show that 257 and 259 are indeed the mass numbers of these two element 104 activities. I have a slide of that later.



We next turned our attention to trying to find the mass number 261. We believed that $^{261}_{104}$ should have a half-life that could be as long as a minute or so. We felt that we should be able to round out this triad, and, if successful, we might be able to do chemistry with it.

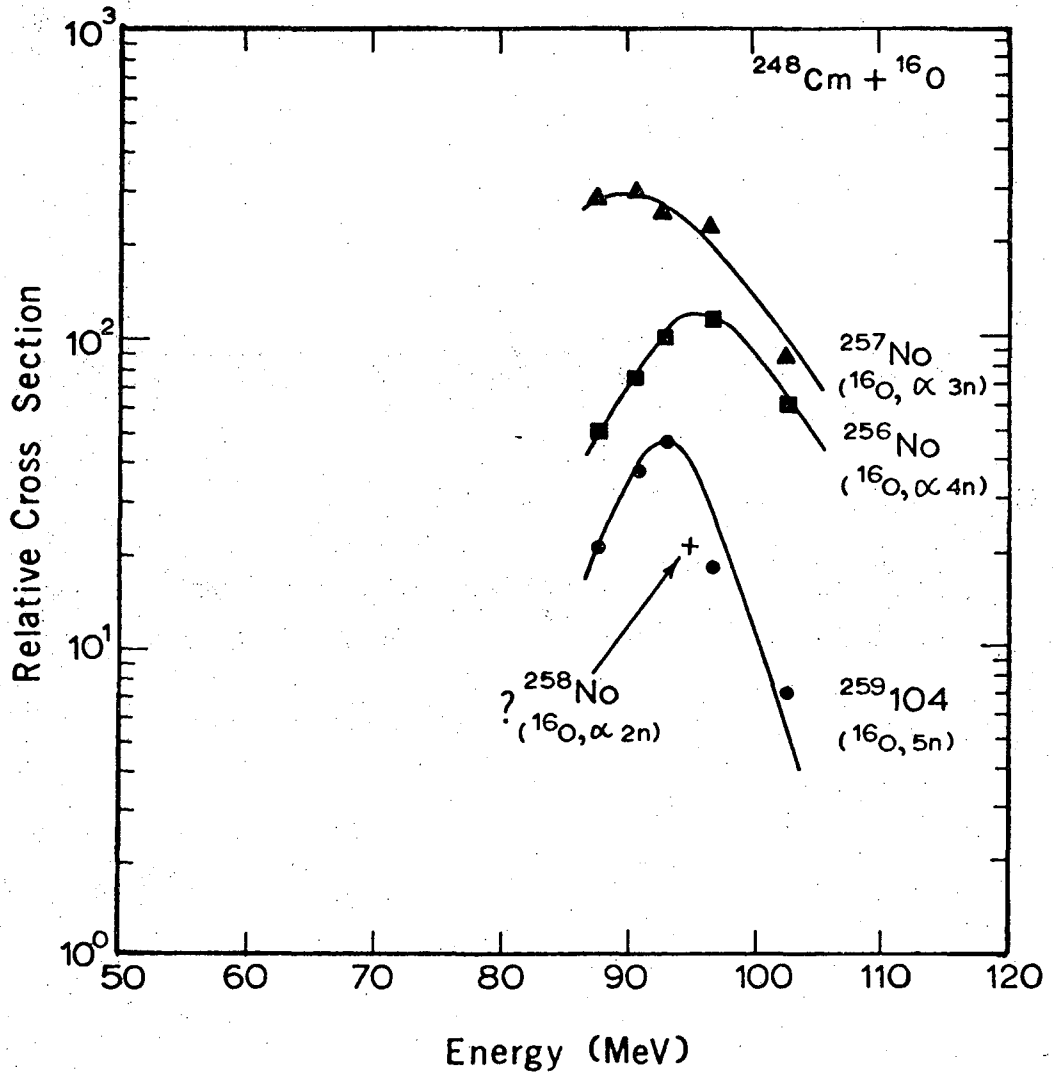
We bombarded $^{248}_{\text{Cm}}$ with $^{18}_0$ and at first had a very difficult

* Since the conference we have found an activity which is best assigned to ^{259}Lr , produced by low-energy $^{15}_\text{N}$ bombardment of $^{248}_{\text{Cm}}$. We find alpha particles of 8.43 MeV with a half-life of 6.5±2 seconds.



XBL 698 4878

Fig. 8. Alpha spectra from $^{259}_{104}$ produced by the $^{248}\text{Cm}(^{16}\text{O}, 5n)$ reaction. The wheel-cycle rate was 2 seconds per crystal.



XBL 6984884

Fig. 9. Excitation functions for the $^{248}\text{Cm}(^{16}\text{O}, 5n)^{259}_{104}$, $^{248}\text{Cm}(^{16}\text{O}, \alpha 3n)^{257}\text{No}$, and $^{248}\text{Cm}(^{16}\text{O}, \alpha 4n)^{256}\text{No}$ reactions.

time in finding what we were looking for. The reason was that it was lost underneath its daughter's spectrum. The $^{261}_{104}$ alpha-particle energy is right on top of the 8.3-MeV ^{257}No . When we got around to examining this region in detail we saw that the half-life was not quite right to be only ^{257}No and we then knew that the $^{261}_{104}$ must be hiding there, too.

Figure 10 - Alpha spectra at 48 seconds per crystal.

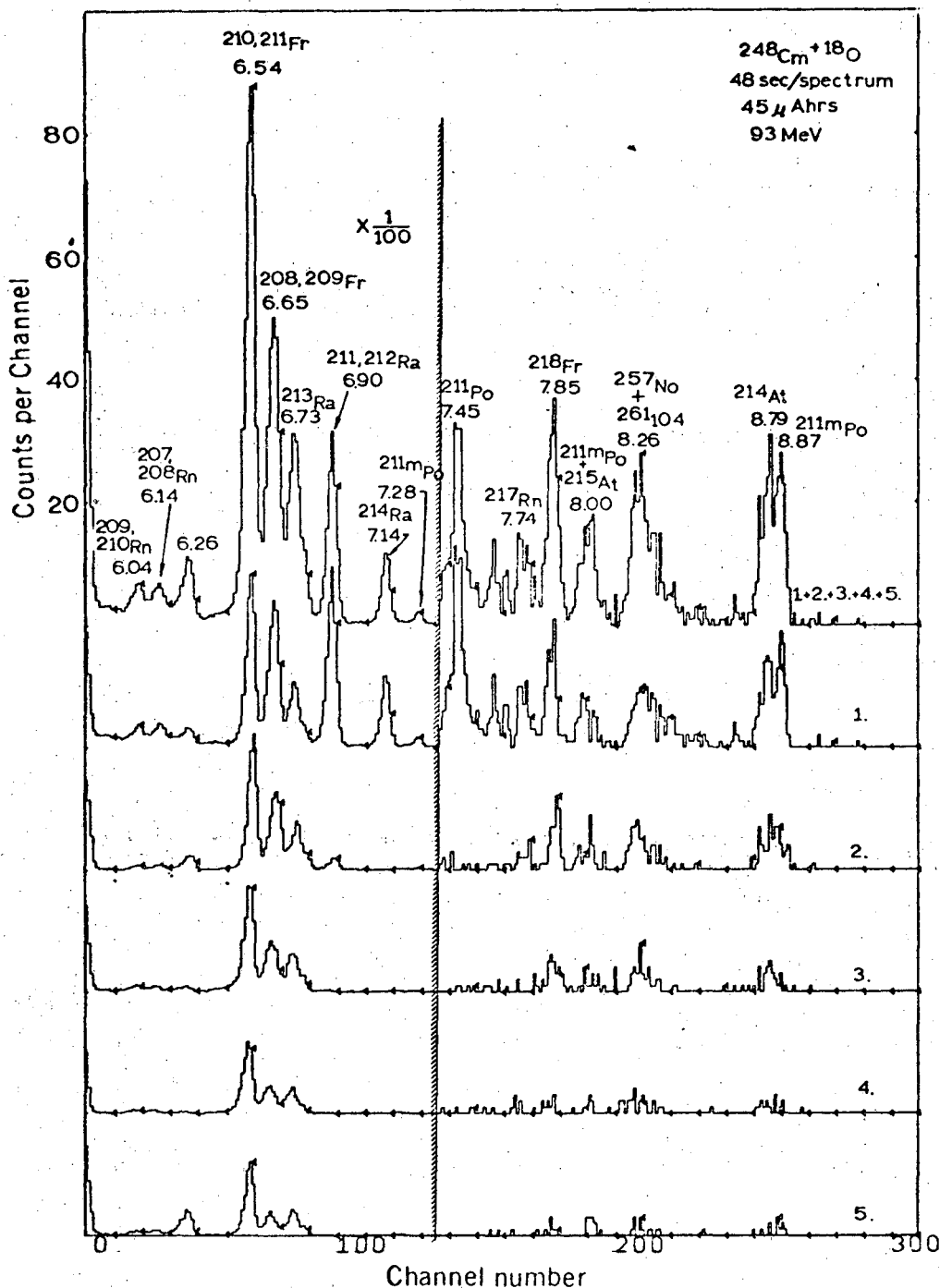
The next transparency shows the spectra taken at 48 seconds per crystal. At this wheel cycle rate we can discriminate somewhat against the ^{257}No that is produced directly. By analyzing these spectra with the computer, we were able to show that it was consistent with a 70-second $^{261}_{104}$ growing into the 25-second ^{257}No . We have not yet made more than a crude excitation function with this activity because of its low production rate. We make a maximum of five counts per microamperehour of the mixture of 104 and its daughter at the calculated peak for the excitation function.

Figure 11 - Alpha spectrum of lead plus ^{18}O ions for comparison.

To show that this is not produced by a lead impurity, here is a lead plus ^{18}O spectrum under similar conditions. This is just the total spectrum. We find nothing that corresponds to the $^{261}_{104}$ - ^{257}No chain; instead we see alpha particles corresponding to the $^{218}\text{Fr} \longrightarrow ^{214}\text{At}$ sequence decaying with a 1-minute half-life. This is odd since their precursor, ^{222}Ac , is known to have a half-life of only 5 seconds and suggests that there must be an isomer of this nuclide with the longer half-life to keep the chain alive.

Figure 12 - Alpha spectra of ^{256}No and ^{257}No produced in $^{248}\text{Cm} + ^{13}\text{C}$ reactions.

In order to be sure that we were really dealing with mass number 261 we felt it necessary to go back and prove once again / that its daughter, ^{257}No , was an 8.3-MeV 26-second alpha emitter. We bombarded the ^{248}Cm target with ^{13}C ions and here are the results of the runs made at 20 seconds per crystal using the



XBL 6911 4925

Fig. 10. A series of alpha spectra of the activities produced by bombardment of ^{248}Cm target with ^{18}O ions. Minute lead and mercury impurities in the target give rise to most of the alpha peaks in the spectrum, the former contributing mostly to the peaks at higher energy and the latter to those at lower energy. The wheel-cycle rate was 48 seconds per crystal position. This experiment was performed using the five crystal-position apparatus and the spectra are thus of individual crystals and their sum. The $^{218}\text{Fr} \rightarrow ^{214}\text{At}$ chain is presumably kept alive by a 1-minute alpha-emitting isomer of ^{222}Ac .

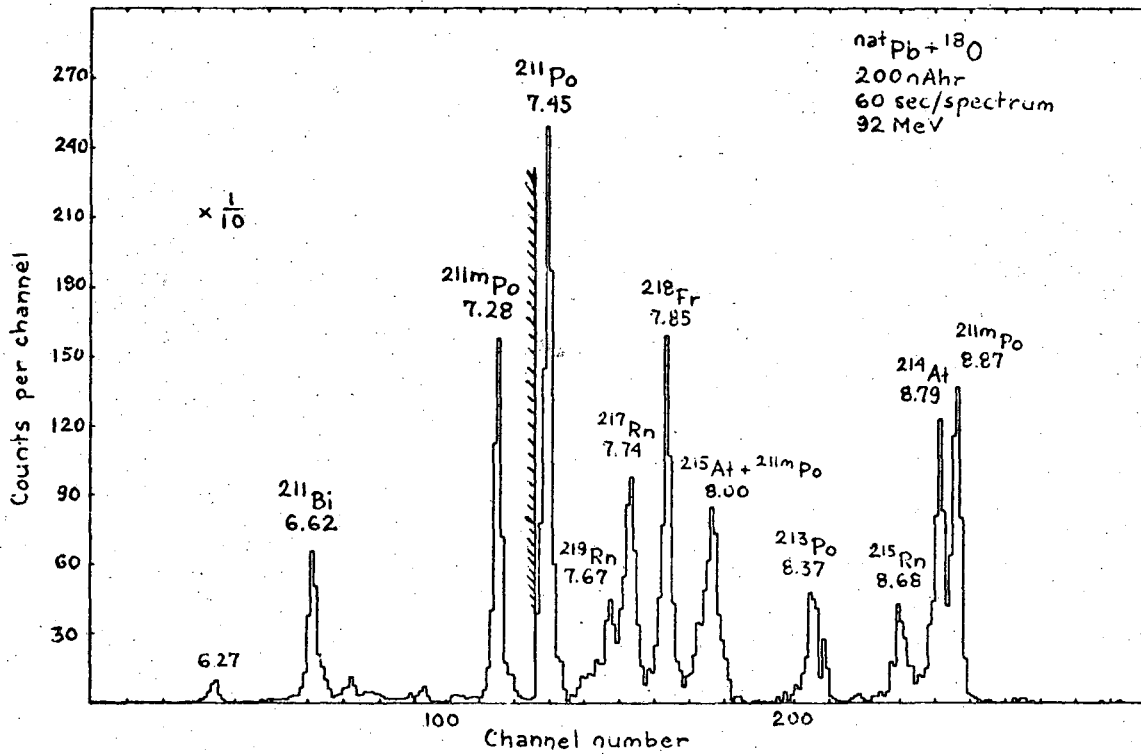


Fig. 11. Alpha spectrum of lead plus ¹⁸O. This spectrum is for comparison with the sum spectrum of Fig. 10, since the wheel-cycle rate was 60 seconds per crystal and the four crystal-position system was used.

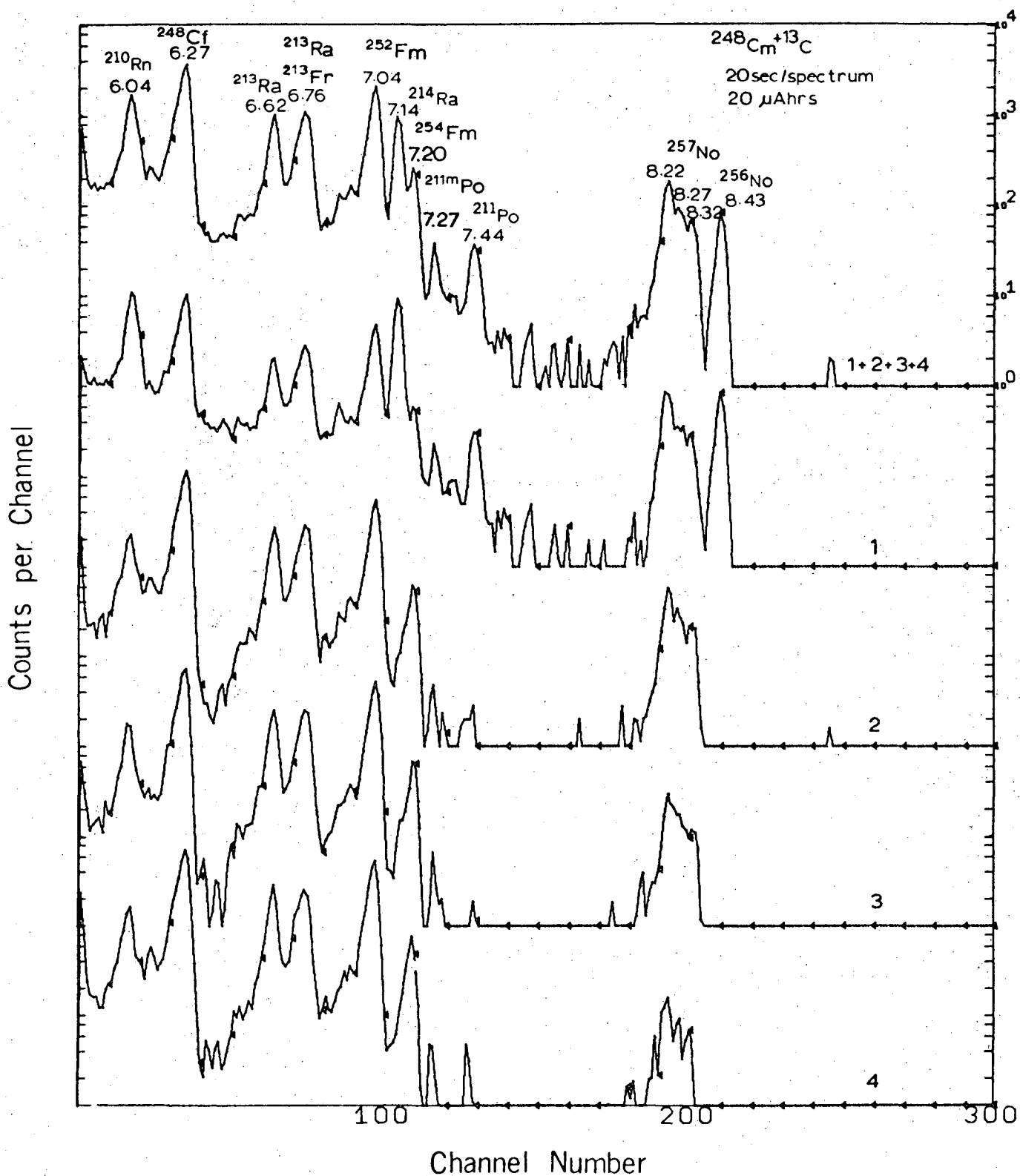


Fig. 12. Alpha spectra of ^{256}No and ^{257}No produced by $^{248}\text{Cm}(^{13}\text{C}, \text{xn})$ reactions. Four crystal-position system at 20 seconds per position.

old 4 crystal-position system. We found the ^{257}No activity decaying with a half-life of 26 seconds with the alpha energies indicated here, 8.22 to 8.32 MeV. The 3-second ^{256}No is also produced.

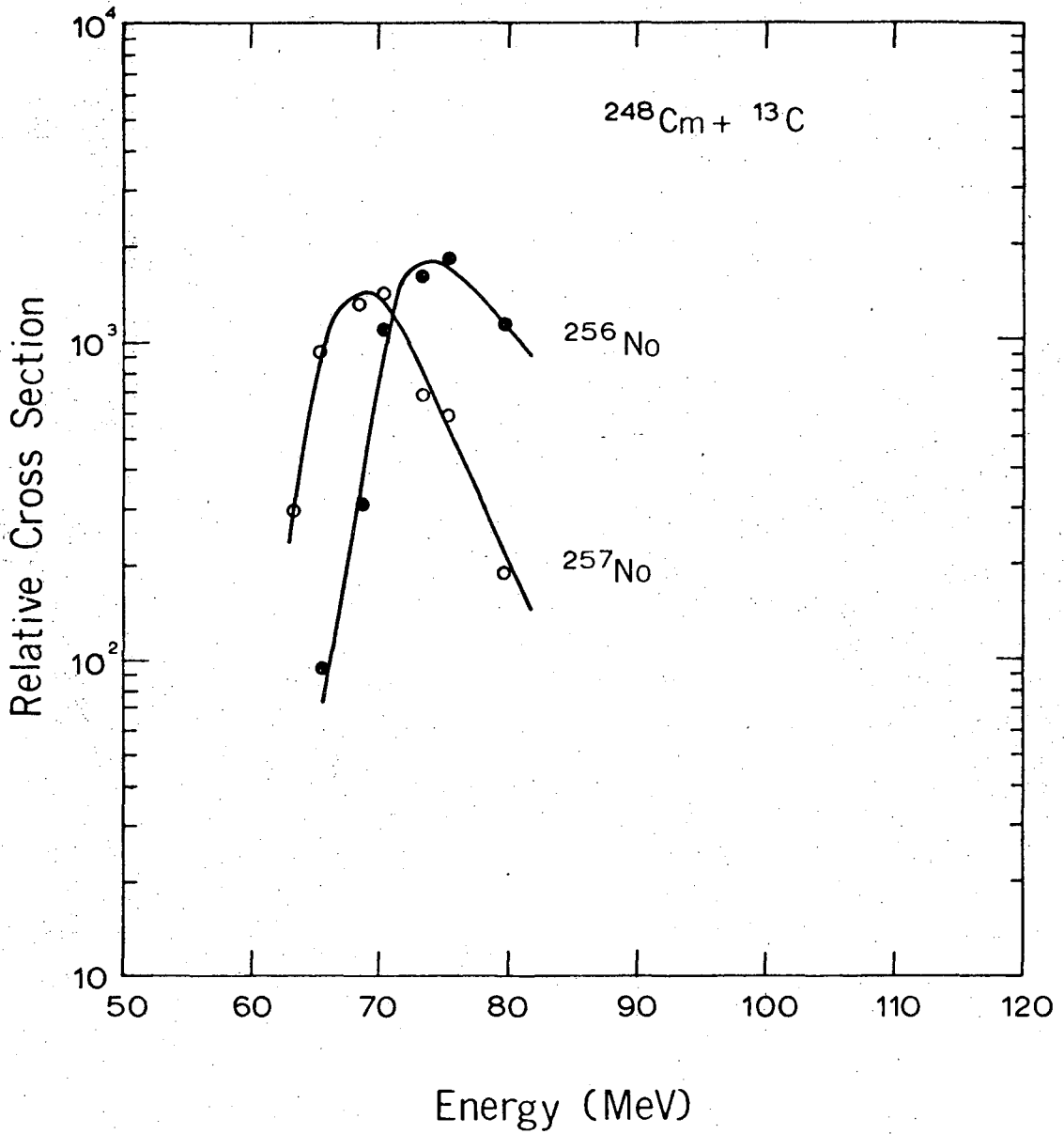
Figure 13 - Excitation functions for ^{256}No and ^{257}No made in $^{248}\text{Cm} + ^{13}\text{C}$ reactions.

The next transparency shows the excitation functions for these two activities. These are consistent with the mass assignments of ^{257}No and ^{256}No assuming they were produced by the evaporation of four and five neutrons, respectively.

Figure 14 - Crystal-shuttle system for alpha-recoil milking experiments.

The next step was to show the genetic relationship between $^{261}\text{104}$ and ^{257}No by making use of the physical separation of mother and daughter that takes place due to recoil when the mother atom undergoes alpha decay. This slide indicates the method that we used. The atoms on the wheel, which we will call the mother atoms, can either decay by emitting an alpha particle outward, which is then detected by the crystal, or it can shoot the alpha particle into the wheel in which case the daughter atom leaves the wheel in exactly the opposite direction and lodges inside the crystal. Obviously there should be as many daughter atoms in the crystal as there are mother alpha particles detected. All that is necessary then is to periodically move the crystal which has looked at the wheel over to a position where it is away from the interfering activity on the wheel.

To increase the geometry for detecting these daughters events, one simply places another crystal next to it so that there is a chance at high geometry to observe the alpha-particle decay of these daughter atoms. By keeping track of the decays in the two positions, it is possible to measure the number of mother events occurring /on the wheel and the corresponding daughter events



XBL 698 4882

Fig. 13. Excitation functions for ^{256}No and ^{257}No made in $^{248}\text{Cm} + ^{13}\text{C}$ reactions.

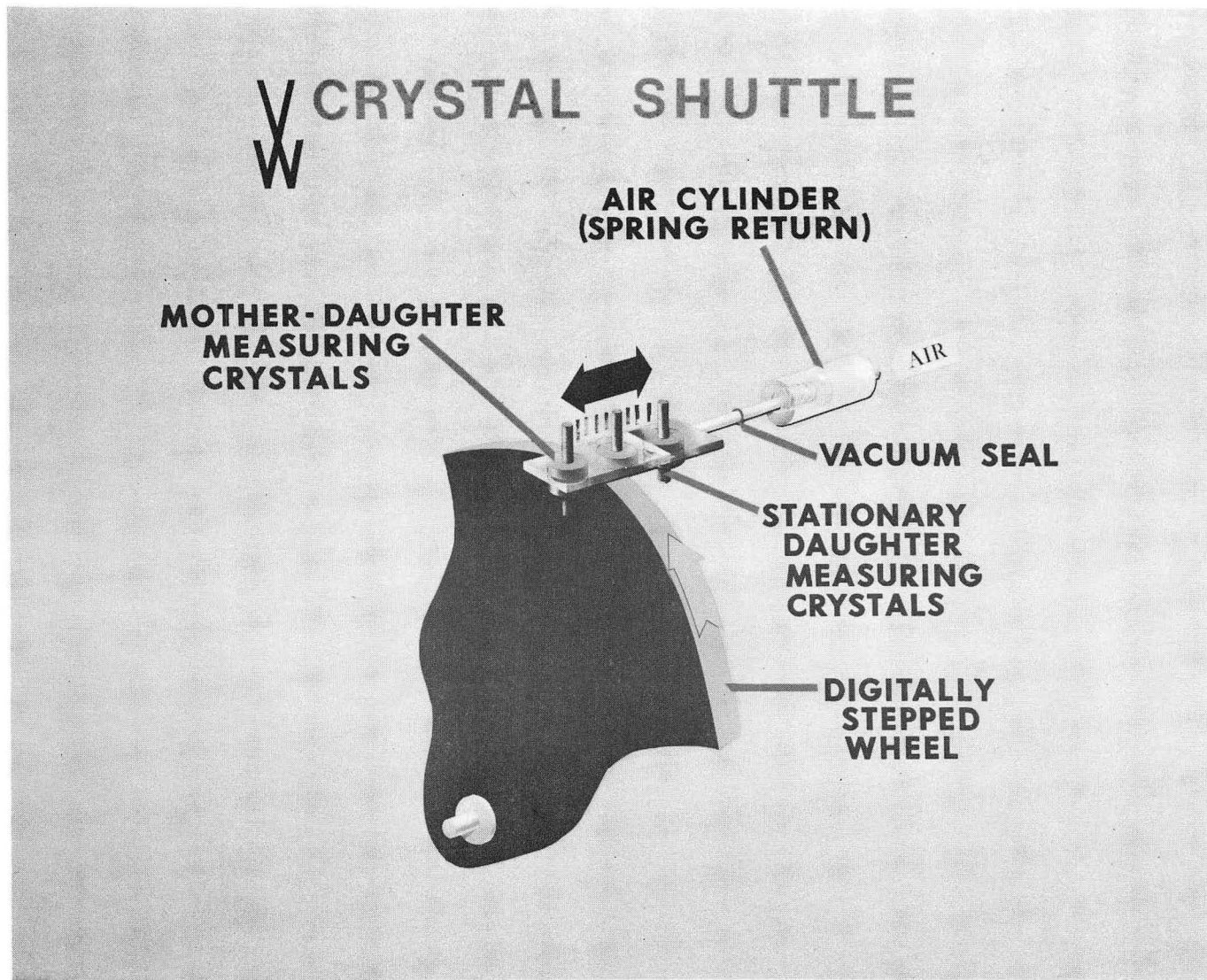


Fig. 14. Crystal-shuttle system for alpha-recoil milking experiments. Schematic representation of the daughter-milking method used with the five crystal-position apparatus. At each position there are four crystals.

CBB 698-5321

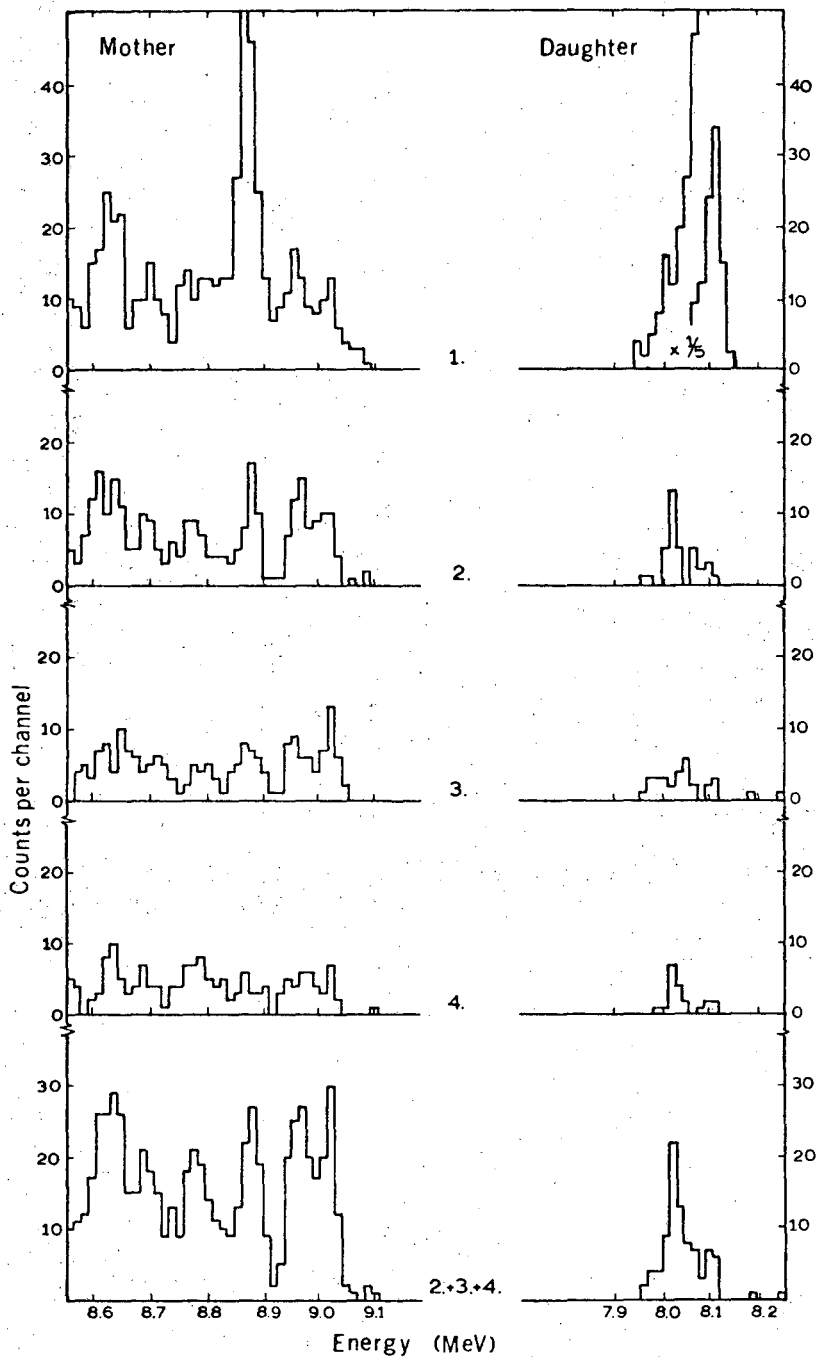
off the wheel.

With only a single crystal being moved off the wheel periodically to measure the recoil daughter, one loses a factor of two because of the time that the detector is away from the wheel waiting for the daughters to decay. With a double-crystal system, one crystal being examined off-line for its daughter atoms while the other crystal is looking at the mother atoms on the wheel, and then vice versa, this lost sensitivity is recovered. There is also the very important double check of one set of crystals versus the other by this redundancy. The data from the 20-detector system are processed and stored by a PDP-9 computer and ancillary units.

Figure 15 - $^{257}_{104}$ \longrightarrow $^{253}_{\text{No}}$ No milking experiment with 8-crystal system.²

To show the strength of the genetic milking experiments, this slide shows those performed with $^{257}_{104}$ decaying to $^{253}_{\text{No}}$ using the old 8-crystal system; that is, four crystals looking at the wheel with four daughter-measuring crystals looking at these same crystals in the off-wheel position. Though our sensitivity was low with this system, nonetheless, we did observe the 105-sec 8.01-MeV $^{253}_{\text{No}}$ daughter activities in the crystals when they were off the wheel.

In the first crystal there was a considerable amount of 55-sec 8.10-MeV $^{254}_{\text{No}}$ which, as I mentioned, was transferred by itself, not by alpha recoil. Fortunately, it only happened at the first crystal position because the transfer half-life was less than a second and the wheel-cycle rate was 3 seconds per position, so that it was possible to detect the alpha-emitting daughter completely free from interference in the succeeding crystal positions. Plotted in the figure is the sum of activities in the second, third and fourth positions. This activity had the proper half-life and energy to be $^{253}_{\text{No}}$ and



XBL 694 4813

Fig. 15. $^{257}_{104} \rightarrow ^{253}$ No milking experiment with the 8-crystal system.

the number of decays that we observed was consistent with the number of alpha-particle decays of $^{257}_{104}$ that we observed on the wheel.*

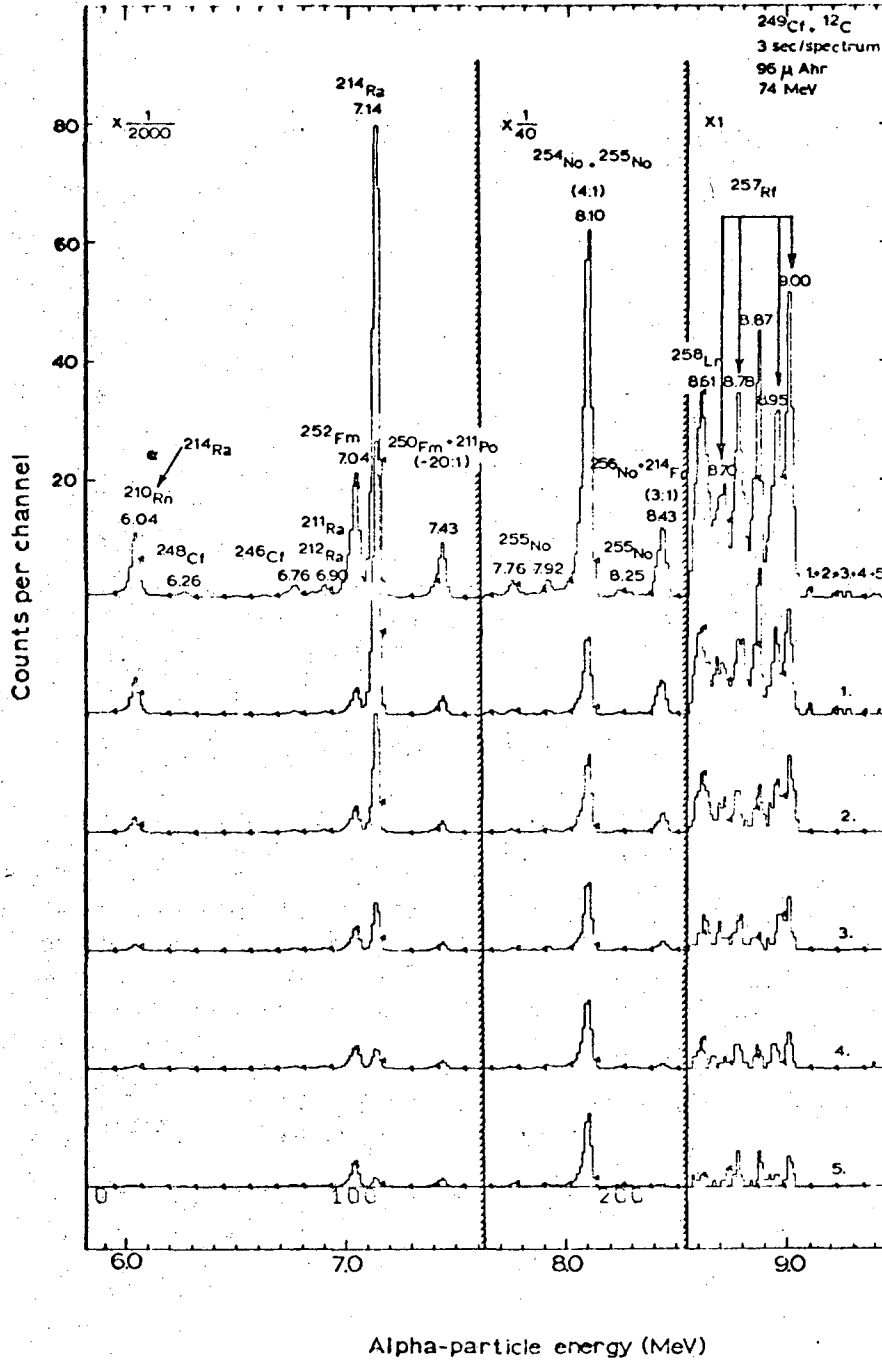
Figure 19 - $^{261}_{104}$ \longrightarrow $^{257}_{104}$ No milking experiment with 20-crystal system.

This slide shows the results of the milking experiment in which $^{257}_{104}$ No was observed as the daughter of $^{261}_{104}$. In this work we used the more efficient 20-detector system. We have plotted the number of daughter events in the interval 8.2 to 8.4 MeV and the results from several independent runs are combined. The number of daughter events observed at each of the five positions is determined by the half-life of the $^{261}_{104}$ mother atoms and we find a value of about 70 seconds. The half-life of the daughter atoms was determined by combining these same events according to the time interval in which they were observed after the shuttle operation began each daughter-measuring sequence. The value found was consistent with twenty-five seconds. I believe we have proven here in the best possible way the genetic sequence of $^{261}_{104}$ decaying into $^{257}_{104}$ No.

Figure 20 - Alpha particle energy versus neutron number with the new element 104 values.

Let me put on the transparency again--the same as shown earlier--but now with the alpha energies from these three isotopes of element 104. Plotted is the alpha energy of the most abundant group; the ground states could be a little higher. Qualitatively, you can see that this is not quite what we predicted four years ago. I understand, however, that these values are actually more consistent with some recent calculations based on the up-to-date

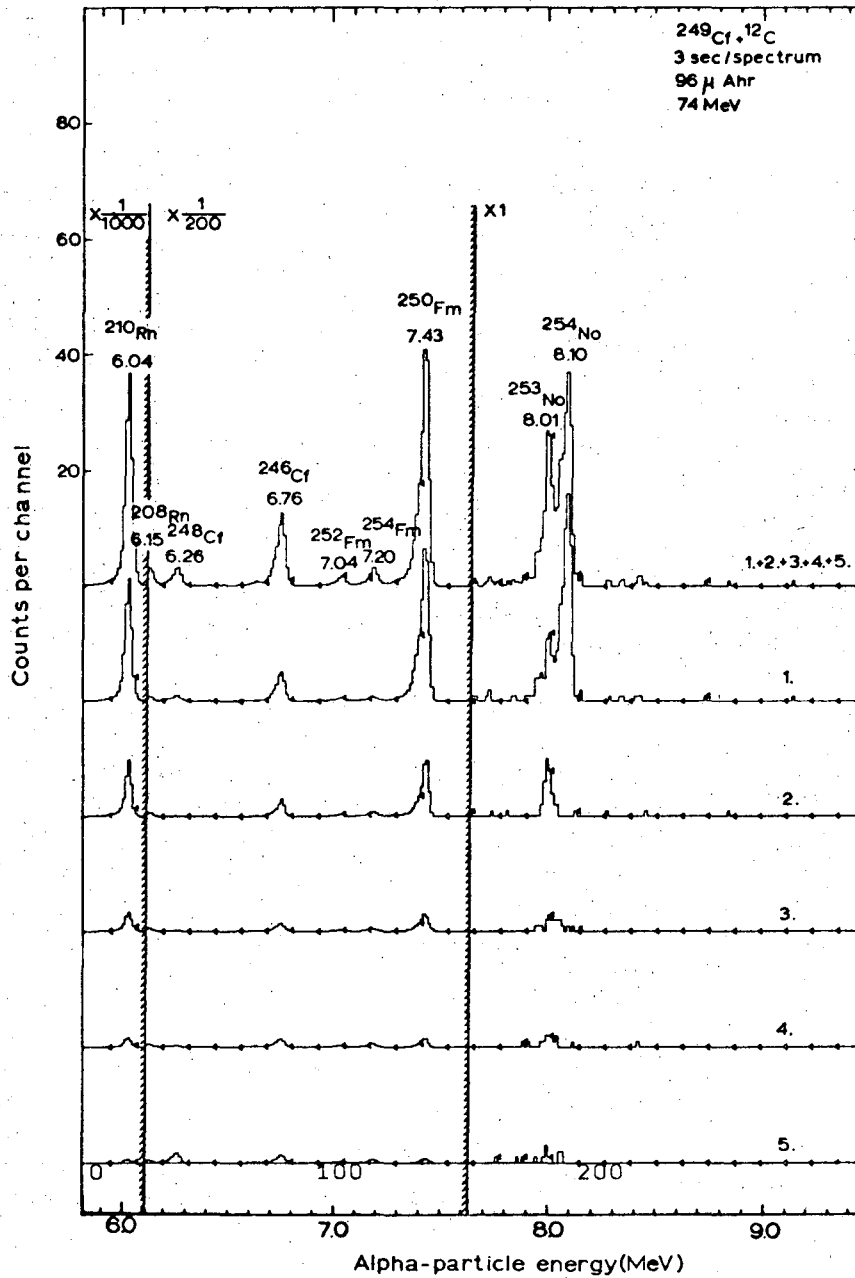
* Since the conference we have repeated this experiment using the much more sensitive 20-crystal system. The new experiments benefited also by an increase in resolution and counting yield due to the incorporation of other improvements. The results are substantially the same as before and are shown in Figs. 16 and 17. Analysis of the data indicates a half-life of 4.8 ± 0.5 seconds for $^{257}_{104}$ as measured directly by its alpha-particle decay; a value of 4.4 ± 1.0 seconds is obtained by measuring the amount of $^{253}_{104}$ No recoiling at each crystal position. Figure 18 shows comparable spectra with Pb + ^{12}C under similar conditions.



XBL 702 6141

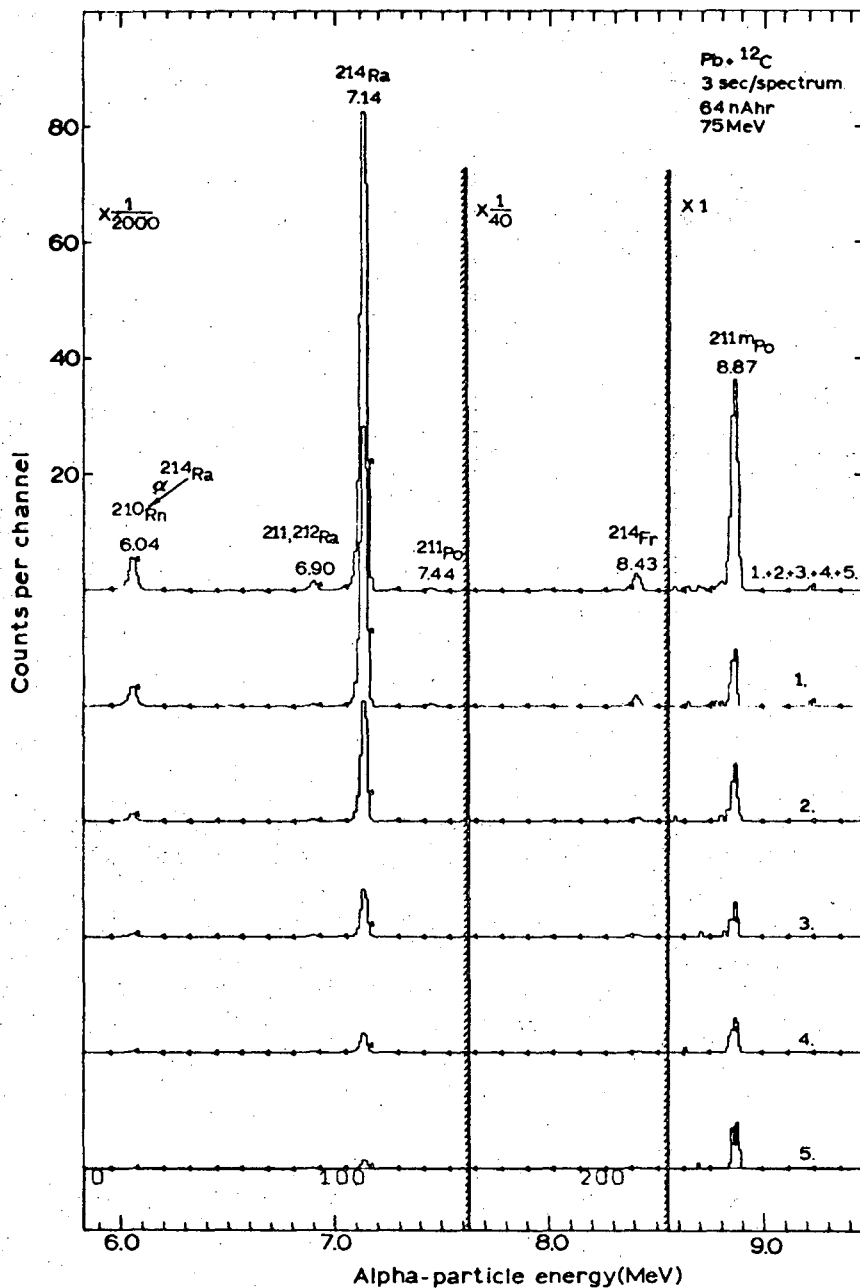
Fig. 16. A series of alpha spectra of the activities produced by recent bombardments of ^{249}Cf target with 74-MeV ^{12}C ions. The experiment was done with the 20-crystal system and the spectra shown are from crystals in the on-wheel position. The wheel-cycle rate was 3 seconds per crystal position. Again some of the labeled peaks are due to a lead impurity in the target. The 6.04-MeV ^{210}Rn peak has the apparent half-life of its mother, ^{214}Ra , because the observed events are due to the decay of ^{210}Rn atoms recoiled to the crystals as a result of the alpha decay of ^{214}Ra atoms.

(Note inproof: The 8.87-MeV peak is believed to be due to an 0.7-sec isomer of ^{258}Lr (or possibly ^{257}Lr) plus the 25-sec ^{211}mPo)



XBL 702 6140

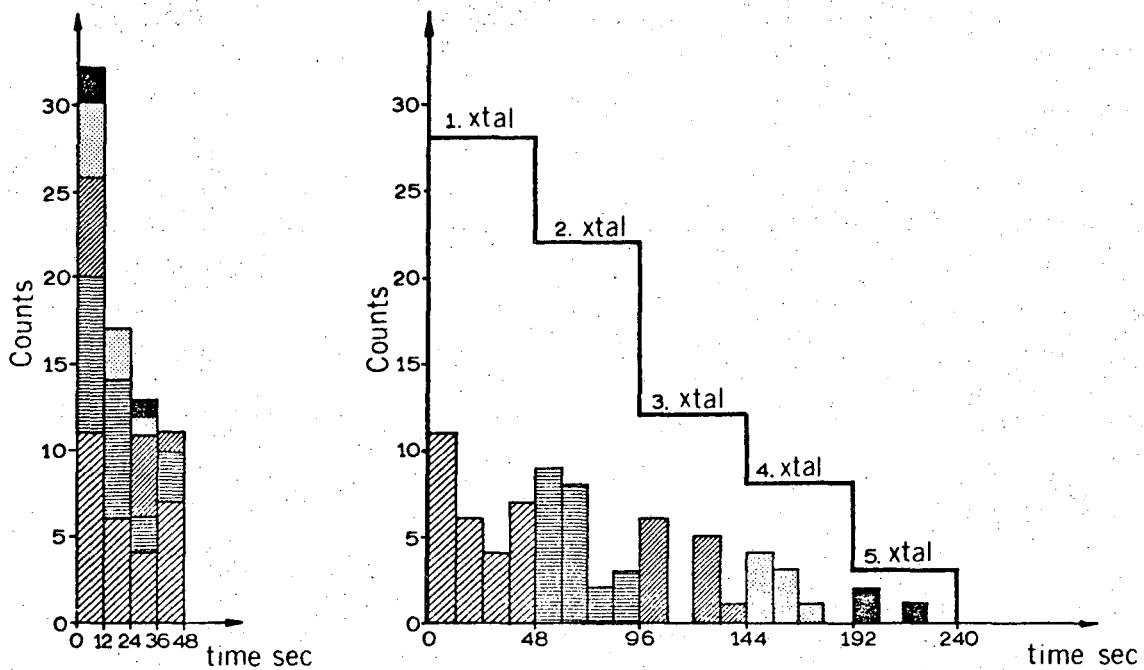
Fig. 17. A series of alpha spectra from the same experiment as in Fig. 16 but recorded by the crystals in off-wheel position. Each spectrum is thus a sum of events from four crystals in the same crystal position. The on-wheel-off-wheel cycle was 100 seconds, while the wheel-cycle rate was 3 seconds.



XBL 702 6139

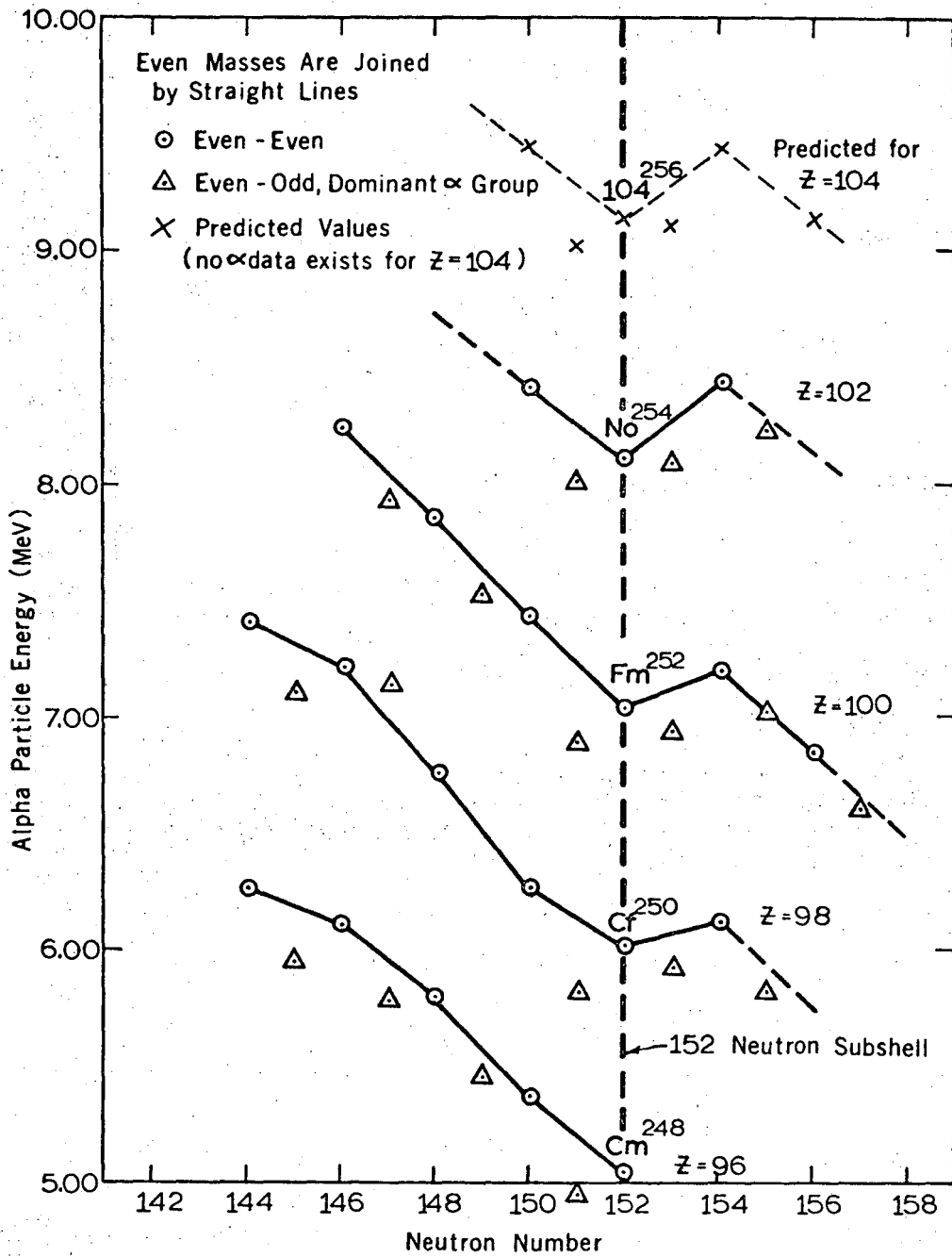
Fig. 18. A series of alpha spectra of lead plus ^{12}C taken under similar conditions to those in Fig. 16. The ratio of $^{210}\text{Rn}/^{214}\text{Ra}$ is smaller than in Fig. 16 because the decay of ^{210}Rn was not followed equally long after the bombardment. Because of the differences in target thickness and beam intensity, the ratio of $^{211\text{m}}\text{Po}(8.87 \text{ MeV})/^{214}\text{Ra}(7.14 \text{ MeV})$ is higher in the lead spectrum.

$^{248}\text{Cm} + ^{18}\text{O}$ $122 \mu\text{A hrs}$
Observed daughter counts at 8.2 - 8.4 MeV energy



XBL 7016105

Fig. 19. $^{261}_{104} \rightarrow ^{257}$ No milking experiment with the "20-crystal" system.



XBL6732028

Fig. 20. Alpha-particle energy versus neutron number with the new element 104 values.

masses so that this data may not really represent any great irregularity. It does not seem to bother any of the theoreticians.

D. FIRST AQUEOUS CHEMISTRY OF ELEMENT 104

Figure 21 - Chemical periodic chart.

Our next step was to try to do chemical experiments with this well-established isotope of element 104. With a 70-second half-life it seemed that we might be able to do meaningful chemistry. In conversation with Dr. Seaborg I indicated that this was possible with some difficulty. Did he think it was really worth the effort? He did not surprise me very much by indicating that he thought it was highly worthwhile and we took this as a command. That was just six weeks ago. We proceeded rapidly to set up a fairly complicated experiment which, I feel, turned out very successfully. I have a movie showing some of the details of this experiment which you might be amused to see.

You will remember from Dr. Seaborg's periodic table that element 104 should be like hafnium if it behaves the way it is supposed to. That means it has four valence electrons and a similar ionic radius and thus a system can be used that can separate it uniquely from most of the other elements. I have portrayed the method in a little sketch here.

Figure 22 - Sketch of chemical method used for separating element 104.

The simplicity of the method depends upon its chemistry being so much different from that of the actinide elements that it will not be adsorbed readily on a cation-exchange column. Using the alpha-hydroxy-isobutyrate eluant that we typically use, the eka-hafnium atoms should be washed straight through the column. Bob Silva, Jim Harris, and Matti Nurmi worked out the necessary

CONVENTIONAL FORM OF PERIODIC TABLE SHOWING PREDICTED LOCATIONS OF NEW ELEMENTS

| | | | | | | | | | | | | | | | | | |
|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|
| 1 H | | | | | | | | | | | | | | | | | 2 He |
| 3 Li | 4 Be | | | | | | | | | | | 5 B | 6 C | 7 N | 8 O | 9 F | 10 Ne |
| 11 Na | 12 Mg | | | | | | | | | | | 13 Al | 14 Si | 15 P | 16 S | 17 Cl | 18 Ar |
| 19 K | 20 Ca | 21 Sc | 22 Ti | 23 V | 24 Cr | 25 Mn | 26 Fe | 27 Co | 28 Ni | 29 Cu | 30 Zn | 31 Ga | 32 Ge | 33 As | 34 Se | 35 Br | 36 Kr |
| 37 Rb | 38 Sr | 39 Y | 40 Zr | 41 Nb | 42 Mo | 43 Tc | 44 Ru | 45 Rh | 46 Pd | 47 Ag | 48 Cd | 49 In | 50 Sn | 51 Sb | 52 Te | 53 I | 54 Xe |
| 55 Cs | 56 Ba | 57 La | 72 Hf | 73 Ta | 74 W | 75 Re | 76 Os | 77 Ir | 78 Pt | 79 Au | 80 Hg | 81 Tl | 82 Pb | 83 Bi | 84 Po | 85 At | 86 Rn |
| 87 Fr | 88 Ra | 89 Ac | (104) | (105) | (106) | (107) | (108) | (109) | (110) | (111) | (112) | (113) | (114) | (115) | (116) | (117) | (118) |
| (119) | (120) | (121) | (154) | (155) | (156) | (157) | (158) | (159) | (160) | (161) | (162) | (163) | (164) | (165) | (166) | (167) | (168) |

LANTHANIDES

| | | | | | | | | | | | | | |
|----|----|----|----|----|----|----|----|----|----|----|----|----|----|
| Ce | Pr | Nd | Pm | Sm | Eu | Gd | Tb | Dy | Ho | Er | Tm | Yb | Lu |
| 58 | 59 | 60 | 61 | 62 | 63 | 64 | 65 | 66 | 67 | 68 | 69 | 70 | 71 |

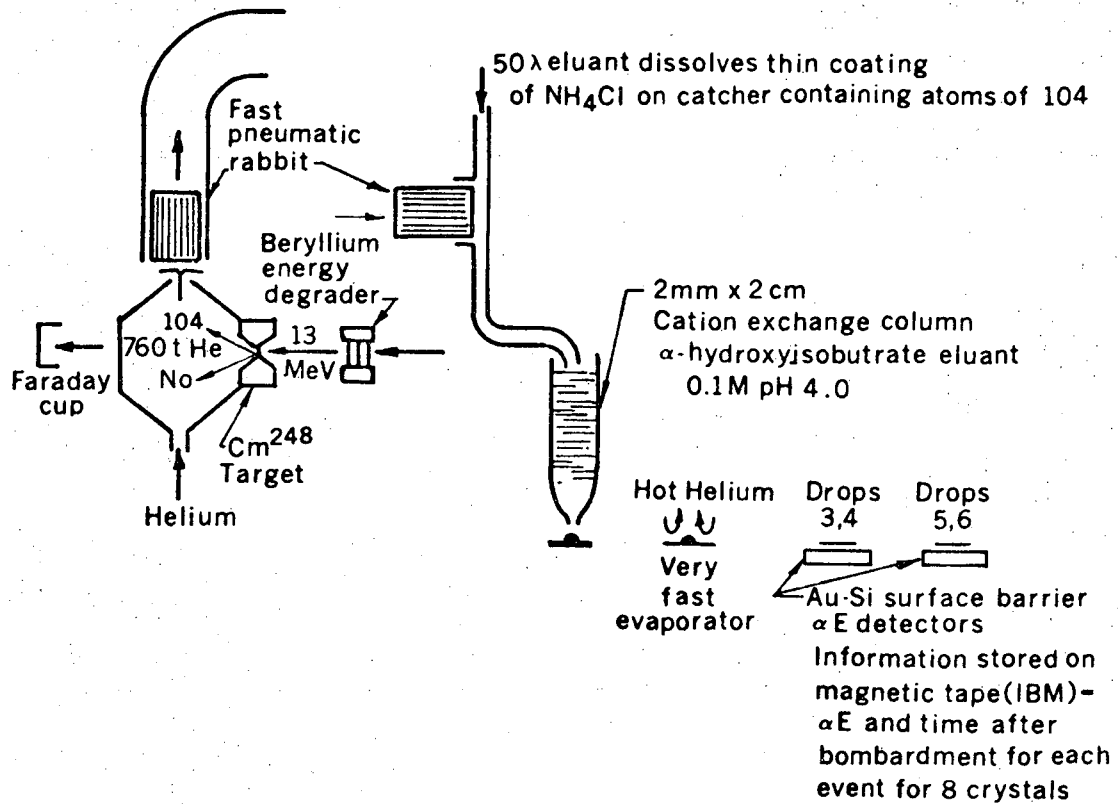
ACTINIDES

| | | | | | | | | | | | | | |
|----|----|----|----|----|----|----|----|----|----|-----|-----|-----|-----|
| Th | Pa | U | Np | Pu | Am | Cm | Bk | Cf | Es | Fm | Md | No | Lr |
| 90 | 91 | 92 | 93 | 94 | 95 | 96 | 97 | 98 | 99 | 100 | 101 | 102 | 103 |

SUPER- ACTINIDES

| | | | | | | | | | | | | | |
|-------|-------|-------|--|--|--|--|--|--|--|--|--|--|-------|
| (122) | (123) | (124) | | | | | | | | | | | (153) |
|-------|-------|-------|--|--|--|--|--|--|--|--|--|--|-------|

Fig. 21. Chemical periodic chart.



XBL 702-6151

Fig. 22. Sketch of chemical method used for separating element 104.

conditions for performing this separation very quickly. They found that it was possible to elute hafnium off the column with the necessary speed so that the problem became simply a matter of logistics, i.e., getting enough atoms in a series of experiments to give meaningful results.

As it turned out, by a factor of two we obtained somewhat fewer atoms than we expected. We think that the probable reason was mechanical losses, which is not too surprising under the circumstances. When you see the movie I think you will appreciate some of the difficulties. We detected an average of one atom in about twenty experiments with each experiment taking about five minutes altogether. This was not as bad as it might seem because, fortunately, the experimental background was essentially zero.

The technique is quite simple. The ^{248}Cm target is bombarded in a recoil chamber just as in the other cases. The recoil atoms are entrained in the gas stream which hits the bottom of a rabbit. On the bottom of the rabbit there is a platinum plate with a thin coating of ammonium chloride previously evaporated there so that the atoms of element 104 can be dissolved readily.

The rabbit is shot very quickly to the chemistry area (in a matter of a second or two), and manually plugged into a holder where automatically the eluant can be rinsed across the face of the catcher. A 50-lambda portion of the solution dissolves the material and puts it on the column. The column is then pressurized to force the drops of eluant through at the rate of three or four seconds per drop. The first two drops are thrown away. The next four drops are where hafnium comes off.

Drops three through six were counted either singly or in pairs in separate counters. The computer keeps track of the time when each bombardment

stops, when each sample is placed in the counter, and when each alpha decay occurs along with its energy. In this manner the data can be summed up from all the bombardments properly.

A very important hurdle that had to be overcome in this experiment is the necessity that each drop had to be evaporated very quickly. You will find that it is very frustrating to take a drop of this material and try to evaporate it fast. It simply is impossible to do by ordinary techniques. Finally we evolved a system which made use of a hollow cylinder of hot helium blowing down around the drop. This made it possible to evaporate the drop in about 5 seconds instead of 30 and--very important--to keep it centered on the counting disk.

Let us go on to the movie now and I think you will understand better the whole experiment. I will try to narrate it as we go along.

Movie - First aqueous chemistry of element 104.

This movie was made just two weeks ago so we have not had time to be fancy. This is the target--60 micrograms of ^{248}Cm . Like the californium target, it was electroplated onto a $100 \text{ microgram/cm}^2$ surface of palladium which had been sputtered upon a 2.4 mg/cm^2 beryllium foil. Matti Nurmia's hands are installing the target in the recoil chamber. You can see the gas-jet orifice on the top of the chamber. Now he is fastening on the degrader-foil holder which is liquid-cooled. The coolant we use is alcohol at -20°C . The beam will go through the degraders to be reduced to an energy of 94 MeV. The recoils will be swept out of the chamber by the helium gas and will emerge from this little orifice here to hit the bottom of the rabbit which he will put in next.

The whole system is in a rough vacuum provided by a large Heraeus blower pump. The pressure depends upon the helium flow rate used--typically, it is in the range from 100 to 300 microns.

This is the rabbit system itself. It is a cylinder which houses a piston with a hole for a vertically-mounted rabbit. When the piston is pneumatically shuttled, the rabbit is moved to a position where it can be blown out into a connecting transit tube to the laboratory bench.

To make certain that the degraded energy is correct, a solid-state detector monitors the scattered ^{18}O beam emerging through a thin window in the target chamber.

Now, Jim Harris and Bob Silva are getting ready to do the chemistry. Here comes the rabbit. Jim plugs it into the side of the column and the liquid flows across washing the ammonium chloride off the rabbit. It then flows off down through the small tube onto the top of the resin column. The column is pressurized and the drops come out. The first two are thrown away. There is the fast evaporator. The stream of hot helium gas is coming out through little holes in the bottom of a copper annulus.

Now, here are drops 3 and 4. These will be evaporated down very quickly while the next drops are coming off. As soon as the evaporation is concluded the platinum plate is flamed with an induction heater and placed upside down on top of a solid-state detector. Now the sequence is repeated so that you can see the details again.

The counting system has eight Si(Au) detectors, each in its own vacuum housing. As soon as a sample is placed next to a detector, a switch is thrown which tells the computer that the counting interval has started. Since

all eight detectors are not usually needed for a single experiment, a new experiment can start while the counting is continuing on previous experiments.

These are the preamplifiers, amplifiers, and discriminators, modular units that were designed and built in our laboratory. The pulses are combined and sent to the computer with proper identification signals. The processed information for each alpha event is stored on IBM magnetic tape together with the time of each event. Energy information between the limits of 6 and 12 MeV is stored in 512 channel spectra.

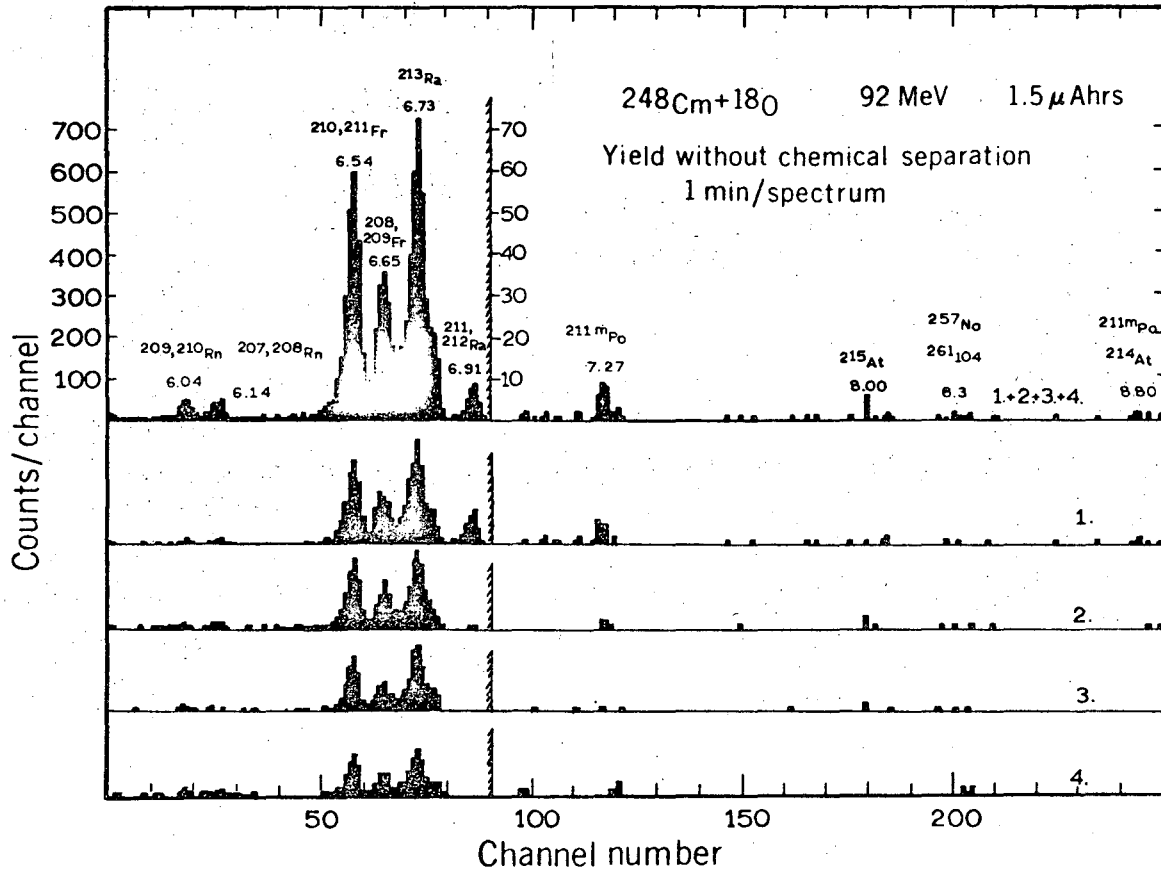
End of film.

One of the unnerving aspects of this experiment was that it was difficult to know when the procedures were working correctly. We did not have a suitable internal tracer available and at best we did not expect more than one countable atom of $^{261}_{104}$ or its daughter, $^{257}_{90}$ No, in 10 experiments. At the same time it was necessary to continue making improvements in the techniques employed. Fortunately, after a few days in which many dozens of experiments had been carried out, we could see that we were probably going to succeed even though our yield was lower than expected--all that was necessary was to perform the task several hundred times. This burden fell largely to Silva and Harris and they did very well indeed.

We intend to go further with this experiment, but to do it entirely automatically--it is just too hard to do with people. We believe that we can automate the whole system and do meaningful chemistry with an even smaller rate of production than one count in every 20 experiments.

Figure 23 - Alpha spectra of the catcher foil without chemical separation.

These are the results. This is the sum of the alpha spectra from



XBL 701 6104

Fig. 23. Alpha spectra of the catcher foil without chemical separation.

many detector analyses of the catcher foil without chemistry. The identified activities below 7 MeV are made from known lead and mercury impurities in the target. In the 8.3 MeV region we find the alphas from $^{261}_{104}$ and its daughter $^{257}_{\text{No}}$. Some of the $^{257}_{\text{No}}$ is made directly. The decay of this peak indicates that about half of it is due to nobelium but fortunately nobelium is held up entirely on the column. It would not come off until perhaps theoretical-drop 3,000 or so. The first actinide element to elute from the column should be lawrencium at about drop 200. As long as $^{257}_{\text{No}}$ is held up by the column there is no interfering activity at this energy to obscure the detection of the $^{261}_{104}$ atoms.

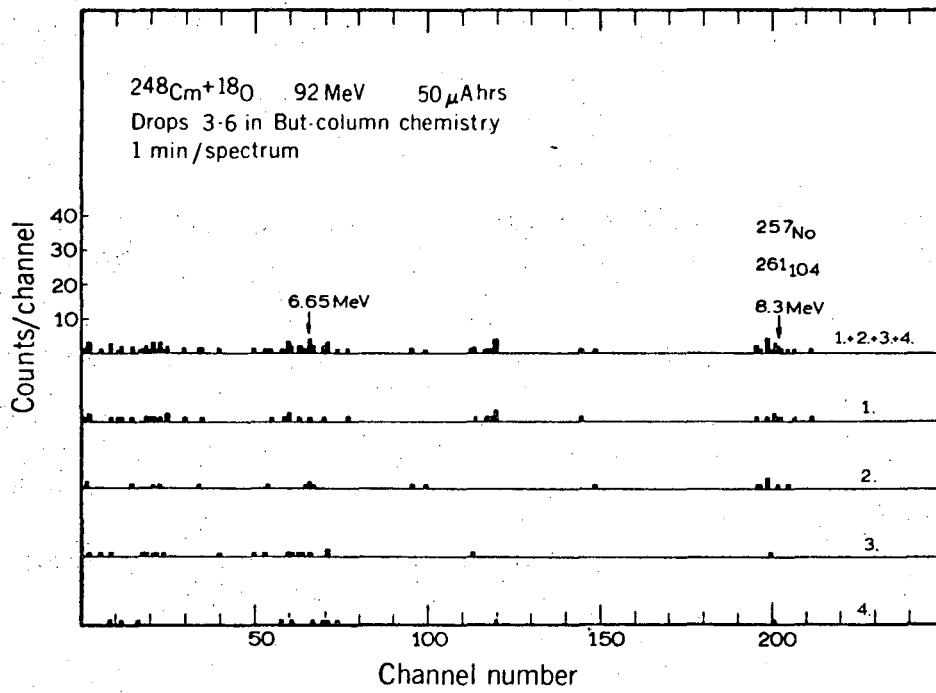
Figure 24 - Element 104 fraction from But column (drops 3-6).

The combined spectrum of drops 3-6 that were washed through the column is shown here. There was a total of seventeen countable atoms which were separated chemically by the resin column and as you can see there was nothing else in this early fraction. These decay with a half-life of about a minute. The next six time channels which are not shown have no counts in them in this energy region.

These results, I believe, are completely beyond question and show that element 104 has an aqueous chemistry different from that of all the other transuranium elements.

These seventeen events remind me that just fourteen years ago the first chemistry on element 101 was done with just seventeen atoms!

(Subsequent to the conference, the final computer analysis of the results of all of the chemical experiments became available. It shows that of the seventeen events reported there were two double events; that is, in two



XBL 701 6103

Fig. 24. Element 104 fraction from But column (drops 3-6).

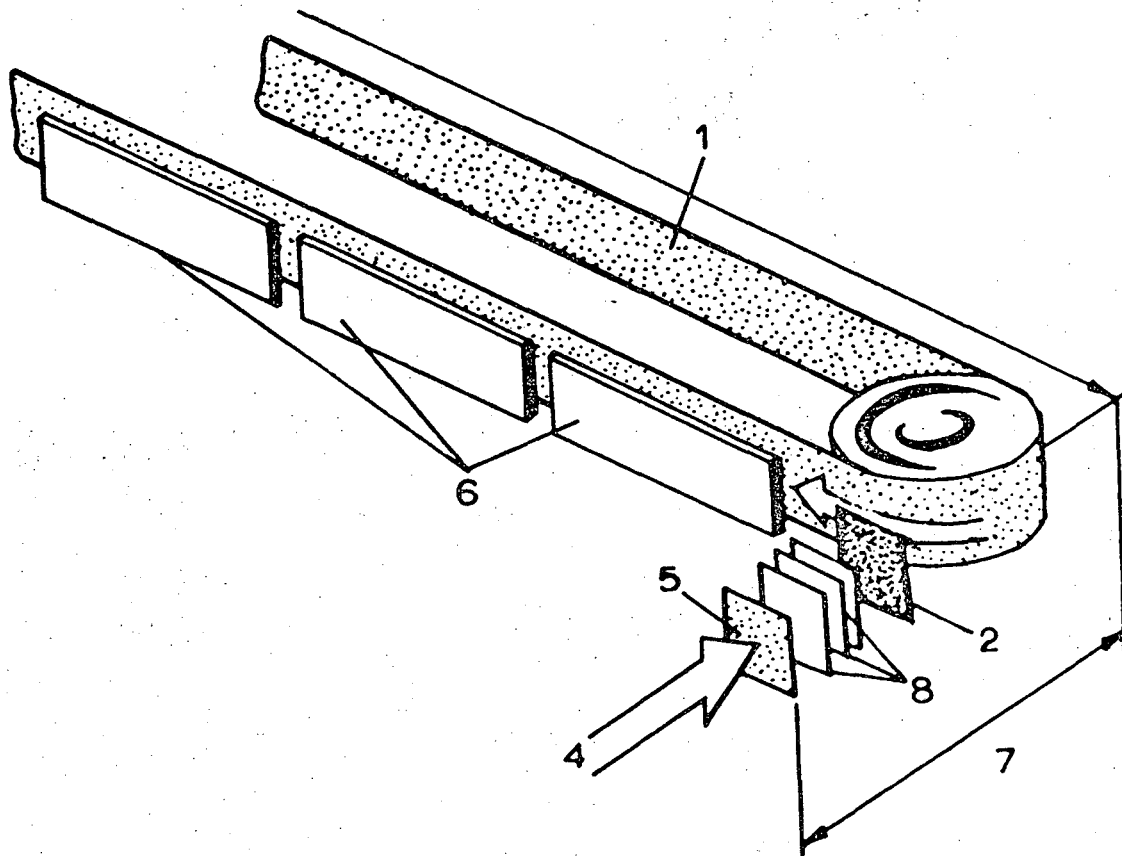
experiments there was one 8.3 MeV alpha followed by another within a minute. Since the counting was performed at about 40% geometry, it was expected from a statistical standpoint that there would be about three such cases in which we would first detect the alpha from a 10^4 atom and then the alpha from its nobelium daughter. Roughly one half of the element 10^4 atoms that were separated chemically would be detected as nobelium atoms since as element 10^4 they either would have decayed before reaching the counter or would have decayed in the counter into the platinum plate on which they were mounted. Correcting for the double decay observed, there were thus only 15 countable atoms of element 10^4 separated by the column. On the other hand correcting for geometry there were 35 atoms of element 10^4 separated, so perhaps we exceeded the 1955 results for mendelevium after all!)

E. ATTEMPTS TO FIND THE REPORTED FRACTIONAL-SECOND SPONTANEOUS-FISSIONING ISOTOPE OF ELEMENT 10^4 .

Now, I would like to go on to the spontaneous-fission emitters and some of the problems that we have had.

Figure 25 - Dubna spontaneous-fission experiment for detecting element 10^4 (1953-1969).

The next slide is one taken from a Russian publication on work done six or seven years ago at Dubna and published first in 1964.³ In this experiment reaction products were knocked out of a ^{242}Pu target by ^{22}Ne ions and caught in an endless belt which passed in front of glass detectors. Spontaneous-fission events were detected as etchable tracks in the glass. They found a spontaneous-fission activity with a reported half-life of three-tenths of a second. They mentioned backgrounds with half-lives of 14 milliseconds (due to $^{242\text{m}}\text{Am}$, as it later turned out) and 8 seconds (attributed to ^{256}No fission



XBL 701-236

Fig. 25. Dubna spontaneous-fission experiment for detecting element 104 (1963-1969).

branching decay, although our best half-life for ^{256}No is close to 3 seconds), along with a neutron-induced fission background. This experiment was done initially inside the 3-meter cyclotron, so it was very difficult work. The rate at which they detected the 0.3-second events at that time was about one count every five hours. When they published their work, they assigned the activity as being most likely due to $^{260}_{104}$. Subsequent gas chromatographic experiments seemed to confirm the purely physical experiments.

Figure 26 - Berkeley drum system used to detect spontaneous fission (1967-).

The next slide shows the system that we have used in searching unsuccessfully for this same spontaneous-fission activity. Instead of using an endless tape to catch the recoiling atoms, we use a metal drum. The drum is rotated to carry the transmuted atoms embedded in the drum next to mica or lexan detectors so that fission events can be detected as etchable tracks. The drum also goes back and forth along its axis so that any long-lived backgrounds are spread out over a very large area; we can get a discrimination ratio of the order of a couple hundred to one if we adjust the rotational speed so that the atoms that we wish to detect decay into the first few centimeters of the mica near the target. It is a very simple, very handy method with a counting yield of nearly 100%, depending only on the efficiency of detecting the tracks. With uranium-free detectors it can be a very low background system.

Using this method we found a new spontaneous-fission activity which has a half-life of eleven milliseconds by bombarding our ^{249}Cf target with ^{12}C and ^{13}C ions. The cross section is fairly large (10^{-32}cm^2), so that over a period of time we have had thousands of fission events and yet, oddly enough, we still do not have absolute confidence in their really being due to $^{258}_{104}$, the most obvious assignment. They most likely are due to this nuclide,

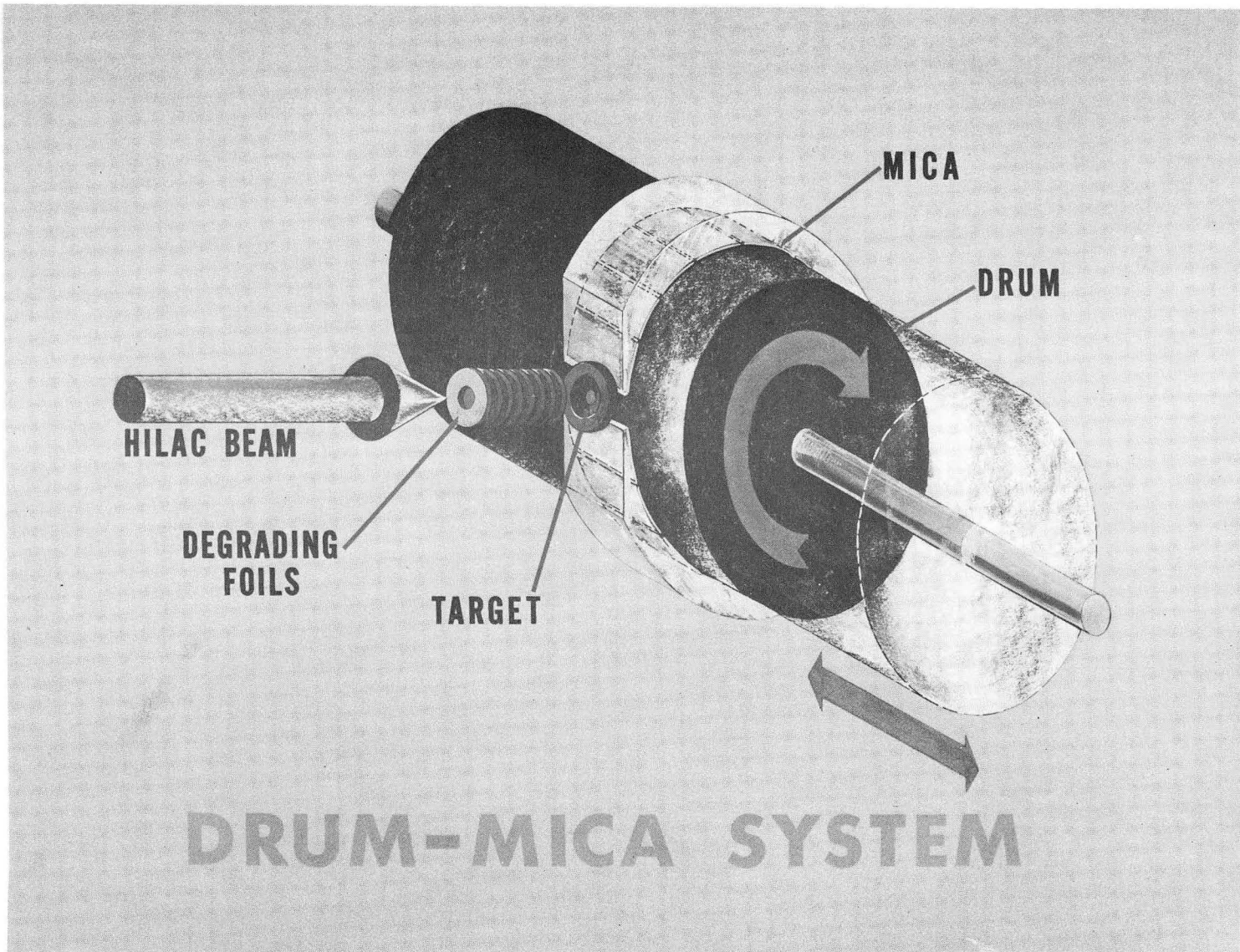


Fig. 26. Berkeley drum system used to detect spontaneous fission (1967 --).

CBB 689-5714

but it is simply hard to be certain. We have done $^{12}\text{C}, 3n$ and $^{13}\text{C}, 4n$ excitation functions and find that they are consistent with one another; yet there is always a doubt since there is no way to tell one spontaneous-fission event from another. One worries about $^{242\text{m}}\text{Am}$ which has a 13.7 millisecond half-life. We have done other experiments which tend to indicate that this nuclide is not made with any significant yield when using such a heavy target with a light projectile. Nonetheless, we still have some doubts.

Figure 27 - Spontaneous-fission half-life versus neutron number.

The next slide shows an empirical plot of spontaneous-fission half-life versus neutron number. It has no physical meaning that we know of but it is a very convenient way of systematizing the available fission data. When plotted in this manner it appears that at the 152-neutron subshell there is a sharp increase in half-life. One way of interpreting this effect is to say that the very slight change in mass near the subshell is enough to increase the spontaneous-fission half-lives above their normal, very much shorter values. In other words, if it were not for the 152-neutron subshell, perhaps the heavier elements would not exist in the laboratory at all!

Plotted here is the Dubna point which is claimed to be $^{260}_{104}$. We would say that the point is too high by orders of magnitude because our $^{258}_{104}$ point seems to fit very well with a simple empirical extrapolation from the nearby, lighter elements.

Seeming to reinforce this interpretation are three other important points-- ^{258}No , ^{246}Fm , and ^{244}Fm . We have produced these three nuclides with a fair degree of confidence, and yet since there is no way of making this mode of decay as specific as alpha-particle emission, there still remains some uncertainty that our assignments are correct.

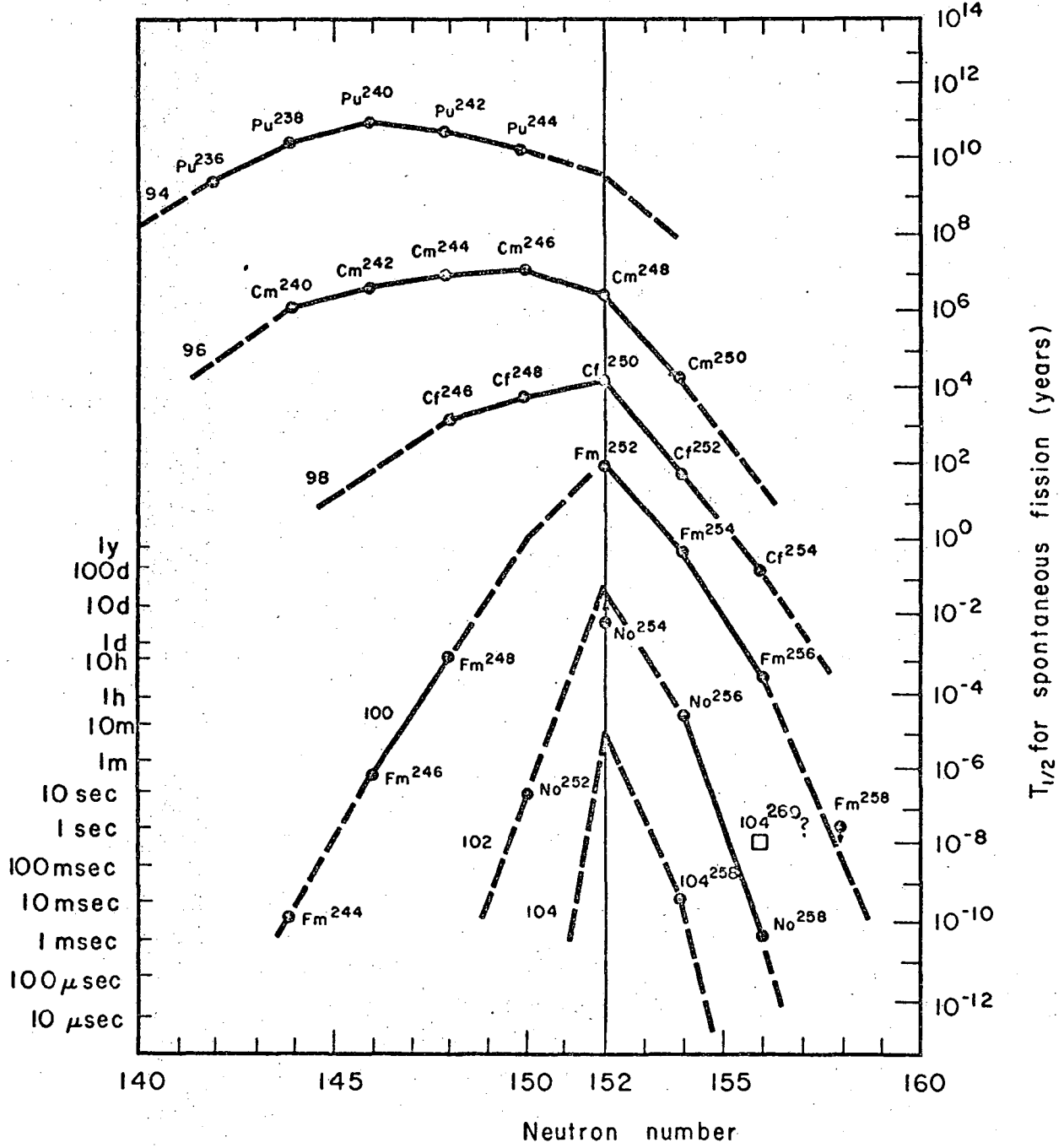


Fig. 27. Spontaneous-fission half-life versus neutron number.

XBL693-2168

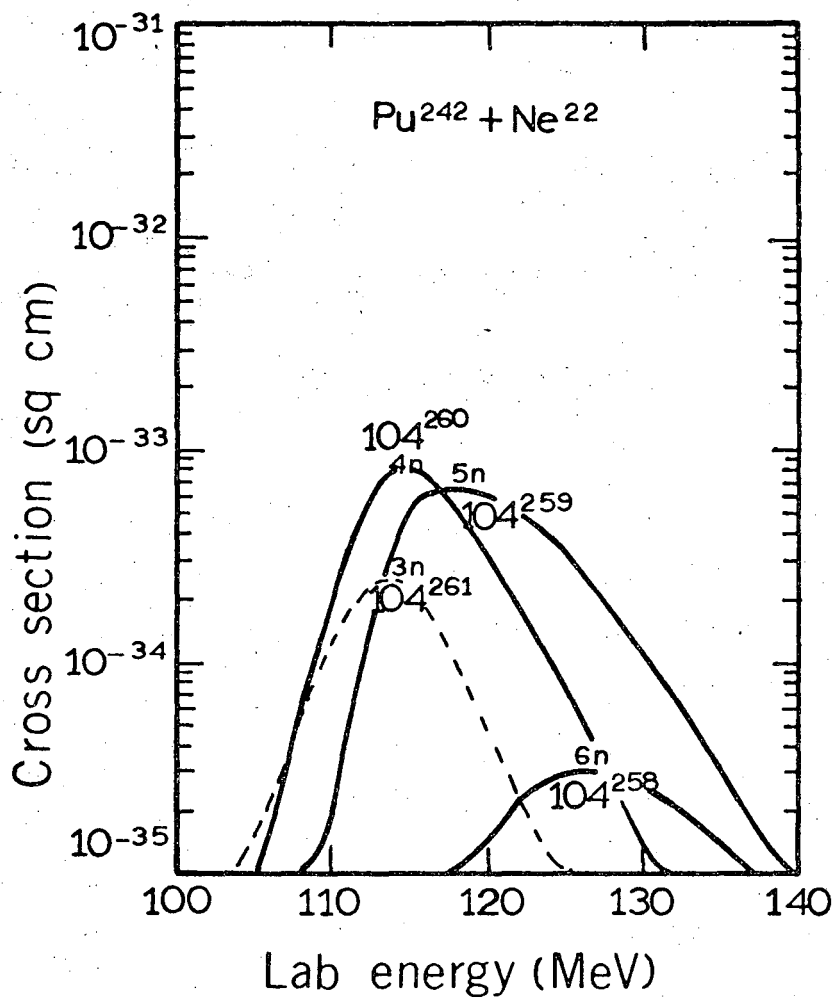
Consistent with this spontaneous-fission half-life systematics picture is the upper limit set by various research groups on the half-life of ^{258}Fm . It still has not been found and the spontaneous-fission half-life limit is getting shorter and shorter (now 100 ms). This fact alone would not seem to be consistent with $^{260}_{104}$ being as long-lived as 300 milliseconds.

The next question to be faced is whether the 0.3-second spontaneous-fission emitter can be assigned to element 104. Some months ago we were told that new experiments performed since our announcement of the new element-104 alpha emitters showed that this half-life is more like 0.1 second,⁵ so we examined our earlier data to see whether this lower value could also be excluded. We decided that our sensitivity was lowered somewhat and it would be well to repeat these experiments using an improved drum system.

These new experiments have also not been successful in finding an isotope of element 104 with a spontaneous-fission half-life in the 0.1-0.3 sec region. In order to illustrate what our new cross section limits for various reactions mean, I will indicate them as values relative to calculated excitation functions for these reactions. These functions are due to Torbjørn Sikkeland⁶ who pioneered in the use of the Jackson-type calculations empirically fitted to dozens of experimental curves obtained at Berkeley and Dubna. These calculated functions are extremely useful, and in general we find that they are not off in the worst case by more than an order of magnitude in absolute cross section. These errors can be corrected by adjusting the parameters, $\Gamma_n/(\Gamma_n + \Gamma_f)$ and R_0 , by using our latest data.

Figure 28 - Calculated excitation functions for $^{242}\text{Pu} + ^{22}\text{Ne}$.

The $^{242}\text{Pu} + ^{22}\text{Ne}$ excitation function should look like this. As



XBL702 6148

Fig. 28. Calculated excitation function for $^{242}\text{Pu} + ^{22}\text{Ne}$.

you can see, the cross sections to make $^{260}_{104}$ or $^{259}_{104}$ are somewhat less than a nanobarn (10^{-33} cm^2). That for $^{261}_{104}$ is indicated to be perhaps 0.2 nanobarn, but it is likely to be a factor of 3 less than this judging by a comparison of our other data in this region with the calculated functions at energies near the coulomb barrier.

I understand that the Dubna value of the cross section for the production of the 0.1-second activity is approximately 0.8 nanobarns at 114 MeV. This relatively large cross section would seem to rule out $^{261}_{104}$ as being responsible for the activity, in agreement with our finding that it is a 70-sec alpha emitter. An assignment to $^{259}_{104}$ might be made on the basis of cross section, but since we have already found a 3-second alpha emitter corresponding to this nuclide it would then be necessary to attribute the 0.1-second activity to a fission isomer of this mass. All other isomers of this type that have been discovered to date have been produced with very much lower cross sections than the ground-state activities, so for its production cross section to be as high as the calculated value is already very surprising. These observations seem to us to limit the mass assignment of this activity, if indeed it is due to element 104, to 260, or less likely to 259.

In our attempts to reproduce the 0.3 (now 0.1) second fission activity we have made the very reasonable assumption that, if it is due to element 104, we can make it more readily by using a higher-Z target and a lower-Z projectile than by bombarding ^{242}Pu with ^{22}Ne ions. We obtain two advantages by this approach: larger cross sections and fewer complicating side reactions. This assumption would appear to be still valid even if we are attempting to make a fission isomer of element 104.

Figure 29 - Calculated excitation functions for $^{246}\text{Cm} + ^{18}\text{O}$.

By using the reaction, $^{246}\text{Cm}(^{18}\text{O},4n)$, we should make $^{260}\text{104}$ with what we consider to be a sizeable cross section. There is some evidence to believe that the $^{18}\text{O},4n$ cross section might be actually somewhat larger than that which has been calculated. The bar indicates our present limit for production of the 0.1-second activity. This conservative limit is more than an order of magnitude below the expected cross section.

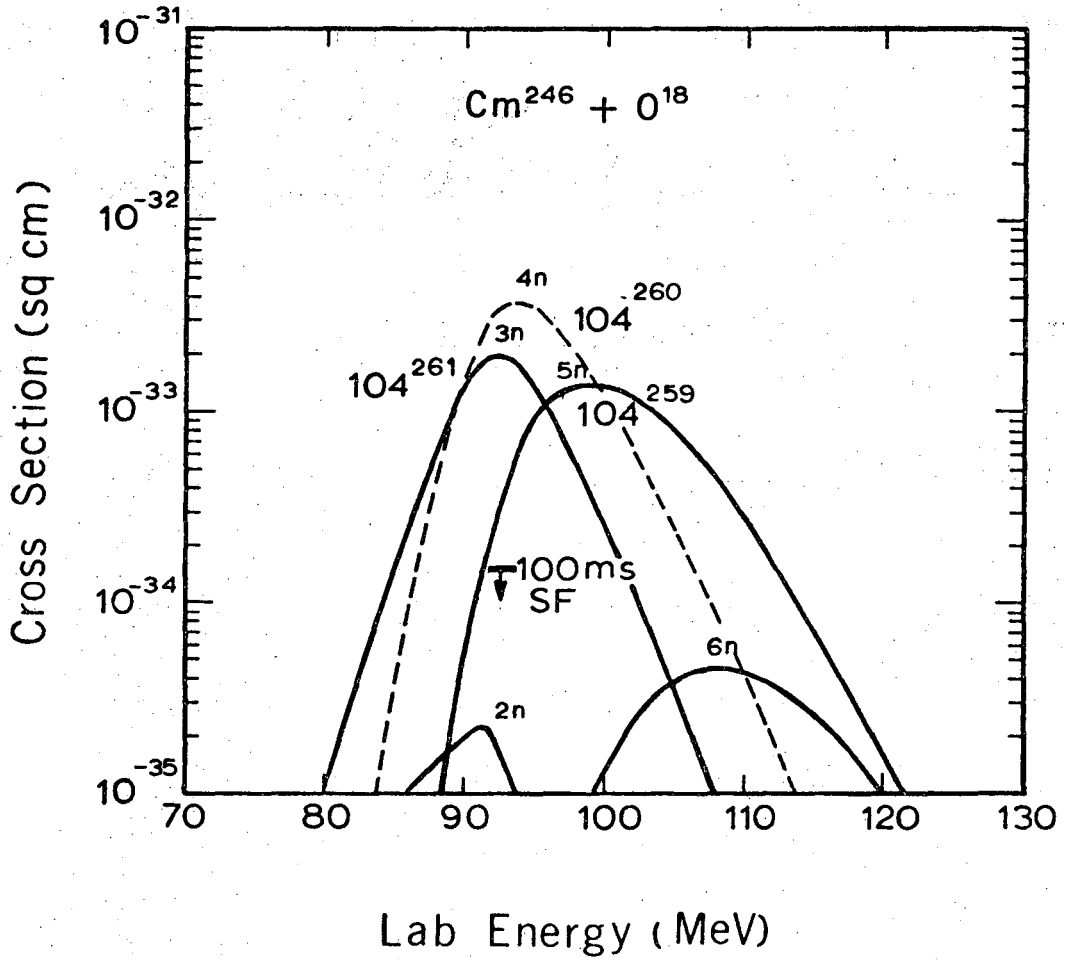
Figure 30 - Calculated excitation functions for $^{248}\text{Cm} + ^{16}\text{O}$.

We have bombarded ^{248}Cm with ^{16}O and again we have not found the 0.1-second activity. In this case the limit was only a factor of 4 below the calculated cross section. On the other hand the measured cross section for the 3-second $^{259}\text{104}$ alpha emitter is above the curve by a factor of 4 which could indicate that the calculated value for the $^{248}\text{Cm}(^{16}\text{O},4n)$ reaction is somewhat low.

Figure 31 - Some experimental excitation functions for $^{248}\text{Cm} + ^{16}\text{O}$.

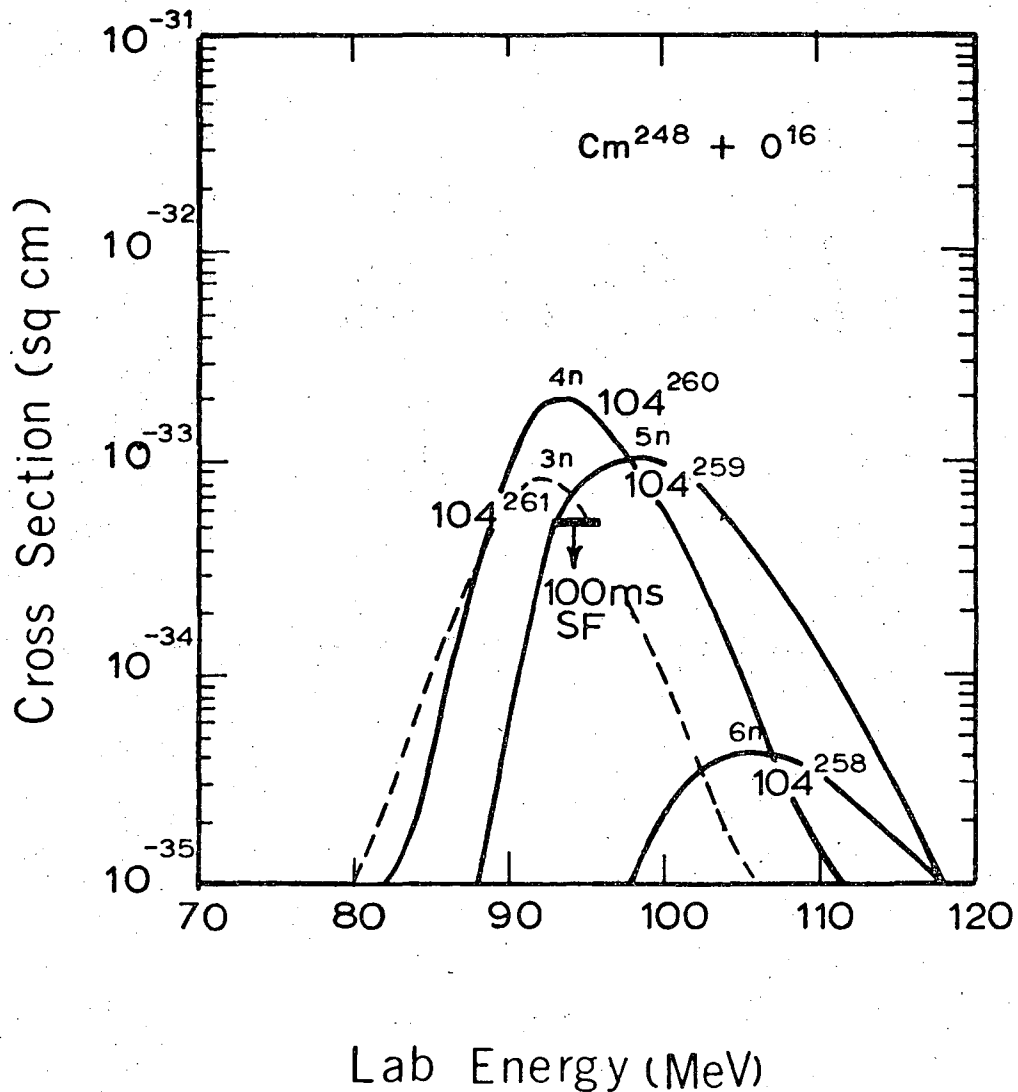
Last week we bombarded the ^{248}Cm target with ^{16}O ions again and ran the drum at two higher speeds in the hope of finding spontaneous-fission activity which might be due to $^{260}\text{104}$ if it had a half-life shorter than 0.1 second. At a very high speed we found a 1-millisecond activity which according to its yield and results of cross-bombardment experiments is probably due to ^{258}No .⁷ Compare the yields to make ^{257}No and ^{256}No .

At a slower rotation we detected an activity with a half-life between 10 and 30 milliseconds but we do not yet have an assignment for it. Of course, it could be due to $^{260}\text{104}$ although it seems that such a half-life is much too long. For that matter the 1-millisecond activity could also include



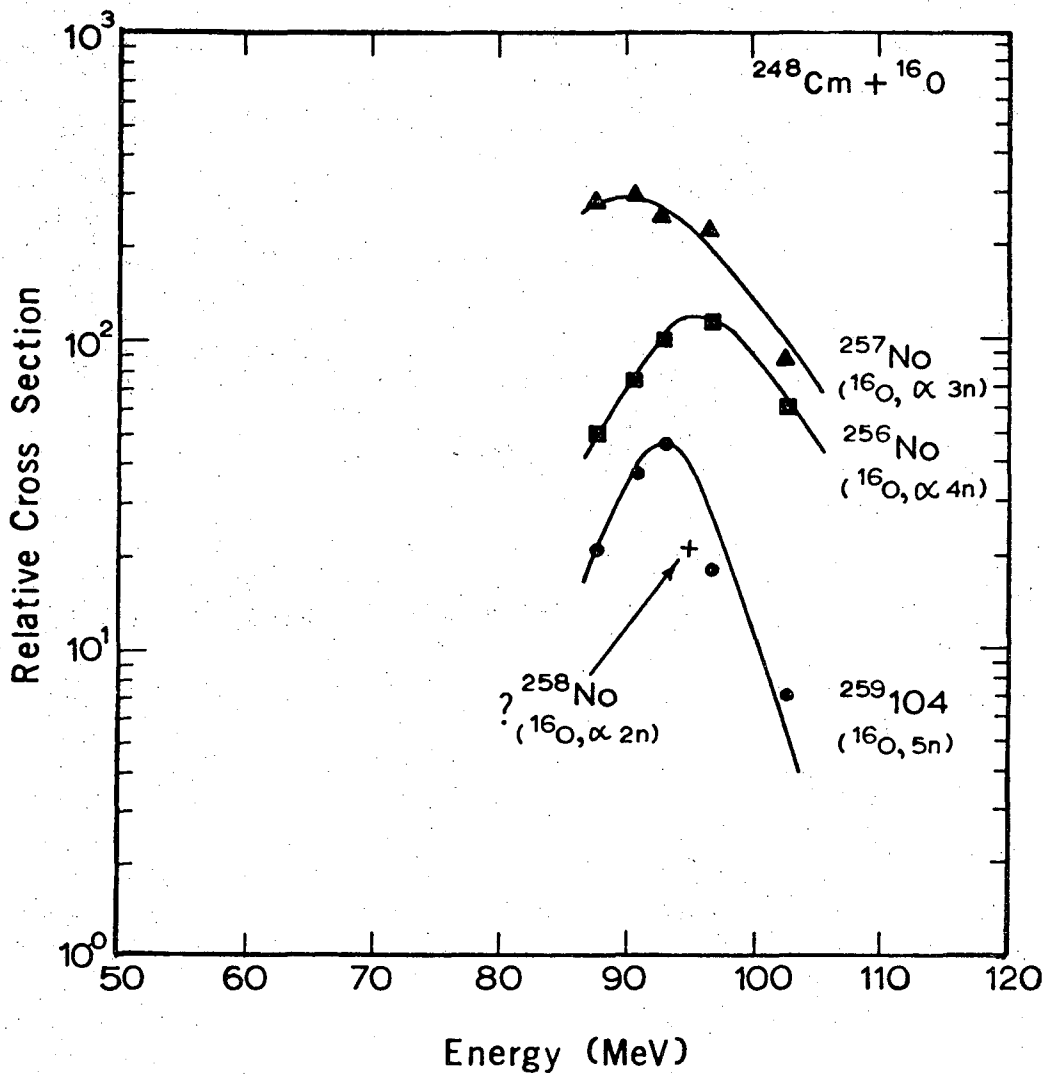
XBL 702 6149

Fig. 29. Calculated excitation function for $^{246}\text{Cm} + ^{18}\text{O}$.



XBL 702 6150

Fig. 30. Calculated excitation function for $^{248}\text{Cm} + ^{16}\text{O}$.



XBL 6984884

Fig. 31. Some experimental excitation functions for $^{248}\text{Cm} + ^{16}\text{O}$.

$^{260}_{104}$ as well as $^{258}_{104}$ No, however it seems to us more likely that the $^{260}_{104}$ half-life is in the microsecond range.

Incidentally, notice ^{that} the $^{248}_{98}\text{Cm}(^{16}_8\text{O}, \alpha 3n)$ reaction makes $^{257}_{104}\text{No}$ very clearly. When we bombard the same target with $^{18}_8\text{O}$ ions, we would expect to make $^{259}_{104}\text{No}$ quite readily. Yet we have not been able to identify it even though we have looked for both alpha particles and spontaneous fissions over a fairly wide half-life range. It makes us wonder whether $^{259}_{104}\text{No}$ is also going to turn up among the missing because of having a shorter half-life than expected.

F. CONCLUSIONS

This, then, is our most recent work on element 104. In summing up the consequences that would seem to follow from our results, I suggest that it is proper for us to draw the following conclusions:

1. We have established with great confidence the alpha-decay properties of the isotopes $^{257}_{104}$, $^{259}_{104}$, and $^{261}_{104}$, and there is a reasonable probability that we have also characterized the spontaneously-fissioning isotope, $^{258}_{104}$.

2. We have chemically separated element 104 by an aqueous method from the other transuranium elements and shown that it behaves like hafnium and zirconium as expected.

3. We have searched diligently without success for an isotope of element 104 that decays by spontaneous fission with a half-life of three-tenths of a second (the original Dubna-postulated value). We have more recently repeated these experiments with higher sensitivities, but find equally negative results for a half-life down to a tenth of a second (their revised value).

4. We are suggesting that element 104 be called rutherfordium, after Lord Rutherford, the great pioneer of nuclear science. If in the course of

further experiments, contrary to our present expectations, we do confirm the earlier findings of the Dubna group of an approximately three-tenths of a second spontaneous-fission activity, we will withdraw our suggested name and accept that proposed by the Soviet group, kurchatovium.

I and my colleagues in these research efforts would like to acknowledge the vital and painstaking assistance furnished to us by all of the members of our laboratory. In particular we are grateful to C. Corum, R. Leres, D. Lebeck, A. Wydler, and A. E. Larsh for mechanical and electrical aspects of the experiments and to F. Grobelch and the Hilac operating crew for the patient and efficient delivery of the diverse heavy-ion beams.

REFERENCES

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

1. A. Ghiorso, B. G. Harvey, G. R. Choppin, S. G. Thompson, G. T. Seaborg, Phys. Rev. 98, 1518 (1955).
2. A. Ghiorso, M. Nurmia, J. Harris, K. Eskola, P. Eskola, Phys. Rev. Letters 22, 1317 (1969).
3. G. N. Flerov, Yu. Ts. Oganessian, Yu. V. Lobanov, V. I. Kuznetsov, V. A. Drulin, V. P. Perelygin, K. A. Gavrilov, S. P. Tretiakova, 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).
4. A. Ghiorso, T. Sikkeland, A. E. Larsh, R. M. Latimer, Phys. Rev. Letters 6, 473 (1961).
5. Yu. Ts. Oganessian, Yu. V. Lobanov, S. P. Tretiakova, Yu. A. Lasarev, I. V. Kolesov, K. A. Gavrilov, V. M. Plotko, Yu. V. Poluboyarinov, Joint Institute for Nuclear Research Preprint JINR-P7-4797 (1969).
6. T. Sikkeland, D. F. Lebeck, unpublished work.
7. M. Nurmia, K. Eskola, P. Eskola, A. Ghiorso, University of California Lawrence Radiation Laboratory Report No. UCRL-18667, 63 (1969).

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

- A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or*
- B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.*

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

TECHNICAL INFORMATION DIVISION
LAWRENCE RADIATION LABORATORY -
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720