Presented at the 3rd International
Conference on Nuclei Far From Stability,
Cargese, Corsica, France, May 19 - 26, 1976

LBL-5037
c.1

FINAL RESOLUTION OF THE ELEMENT 104 QUESTION

Albert Ghiorso

May 1976

Prepared for the U. S. Energy Research and
Development Administration under Contract W-7405-ENG-48

DISCLAIMER

FINAL RESOLUTION OF THE ELEMENT 104 QUESTION

by
Albert Ghiorso
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720, U.S.A.

Paper to be delivered at the
3rd International Conference on Nuclei far from Stability
Cargèse, Corsica (France)
May 26, 1976

This report was done with support from the United States Energy Research and Development Administration. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the United States Energy Research and Development Administration.

00004504459

FINAL RESOLUTION OF THE ELEMENT 104 QUESTION

A. Ghiorso

Lawrence Berkeley Laboratory, University of California, Berkeley, California, USA

Abstract

A history of the long controversy between Dubna and Berkeley concerning the discovery of element 104 is given. In particular we review the difficulties associated with the use of spontaneous fission in detecting new elements and contrast this method with the use of alpha particle activities to define the atomic number. Recent experiments at Dubna and Berkeley concerned with bombardments of ^{246}Cm with ^{18}O, ^{248}Cm with ^{16}O, and ^{249}Bk with ^{15}N are compared in detail. The categorical conclusion is drawn that the Dubna claim that the isotope ^{260}104 decays by spontaneous fission with a half life of 0.1 second is not valid.

* * *

There has long been a debate between Dubna and Berkeley concerning the properties of the isotopes of the element with atomic number 104. At Berkeley we have just completed experiments which we believe can only be interpreted as disproving the Dubna work concerning this element from 1964 to the present moment. I will present the details of these experiments later in this paper after reviewing the history of the work of both laboratories on this element. To place this material in context I will first make some comments on the basic methods that have been used to detect and identify nuclides of the heaviest elements.

The two major methods that have been used in this work up to now are concerned with the information conveyed by spontaneous fission decay and by alpha particle decay, and each method has its inherent advantages and disadvantages. The disintegration of a heavy nucleus by spontaneous fission releases a large amount of energy, which principally manifests itself in the kinetic energy of two roughly equal fragments emitted from the original nucleus in opposite directions. This large energy (ca. 100 MeV per fragment) is easily detected in hostile environments because it is released in traversing a thickness of only a few milligrams per cm^2. Particle counters of various types and dielectric track detectors (with subsequent chemical etching to make the fragment tracks visible optically) are the usual means of detection. The efficiency of detection is usually high--30% to 100% being easily attainable with complete discrimination against lighter particles. Background effects are usually small but can become important in certain cases, such as in the use of track detectors in an apparatus set up inside an accelerator where a high neutron field exists. If the dielectric material contains even minute amounts of uranium or thorium, then a background from fast-neutron-induced fission can be observed which is indistinguishable from the spontaneous fission effect that is the object of search.

It is this non-specificity of fission that makes for a major disadvantage of its use as a tool for the identification of the heavy elements. When the nucleus disintegrates it does so with a total energy which varies considerably in how it is distributed among its fission products. Other than half life no simple way has yet been demonstrated to show the physical difference between one spontaneous fission activity and another except perhaps by the careful detection of many thousands of events. A determination of half life is the only method used to characterize a spontaneous fission activity but, because of the fact that all spontaneous fissions essentially look alike, this can be almost impossible if several activities are present with similar half lives or if backgrounds are present due to stray neutrons or other causes. Even the seemingly trivial question of determining the presence or absence of a fission activity with a given half life in an observed time distribution of events can be very difficult.

This limits the use of the technique of cross bombardments, a method which has been used with some success to make specific identifications. This method, though valuable, has its difficulties when used in regions where ambiguities can prevail or where cross sections are small.

Another method commonly utilized--excitation functions--can also present serious problems in complicated cases. The analysis of excitation curves, i.e., the variation in yield of an activity as a function of the energy of the bombarding particle, necessitates a quantitative measurement of the amount of an activity with a given half life in mixtures with drastically varying amounts of other activities and backgrounds. Such an analysis will work well for light projectiles but can give very misleading results when the projectile is a heavy ion and when the cross section is very small.

Finally, a technique that has been employed with partial success makes use of the fact that compound nucleus reaction products tend to have angular distributions which are peaked sharply forward. The Dubna experimenters have used special honeycomb collimators next to their targets to enhance the yield of such C.N.-products relative to the yield of those products made by transfer reactions. Unfortunately, little or no discrimination is obtained against those reactions of the type (HI,pxn) or (HI,αxn).

All of these difficulties become even more irksome when the activity that is under study is accessible only at a very low production rate. In our laboratory we have never felt a great confidence in this type of experiment unless it incorporated an additional identifying parameter such as that provided by chemical separations.

The alternate method of detection and identification of the heaviest elements makes use of the fact that alpha particle disintegration is usually the most prominent mode of decay; as a consequence, we at Berkeley have gone to great pains to use and improve techniques involving alpha particle measurements. In contrast to spontaneous fission, alpha decay is very specific. The decay by such emission is by precise amounts of energy appearing as several narrow spectral lines. The alpha particles can be readily measured with an accuracy of better than 30 keV even at miniscule counting rates. Alpha particle energies vary in a systematic way which is now well understood and this fact makes it possible to predict the energy to be expected from an unknown nuclide. In effect every nuclide can be labelled with a fair degree of certainty and since an alpha energy can be coupled with a corresponding half life this labelling can be almost definitive by itself.

Background problems can exist, however, because there are certain nuclides such as ^{211m}Po which undergo alpha decay in the same general energy region. If such background activities are made from target impurities at a level that swamps the heavy element activity, the only ready cure is to remove the impurities and this is the approach that we have adopted in the past.

Alpha particle decay has another inherent advantage in that it is usually possible in the very heavy element region to genetically link one alpha emitter with another. In this case the coupling of one species with an almost-unique energy and half life with another nuclide with a known almost-unique energy and half life provides an unambiguous assignment in mass and atomic number. Important also is the fact that the genetic linkage can be established by two independent techniques: alpha-recoil milking and time correlation, and both methods have been employed at Berkeley.

Another technique, which up to now has been used only at Oak Ridge National Laboratory, is to measure the characteristic x-rays following alpha decay to define the atomic number. This beautiful method suffers somewhat in sensitivity compared to the preceding genetic linkage experiments.

In summary we believe that it is fair to say that the use of spontaneous fission as a means to detect unknown nuclides is a very sensitive technique and a very important one. However, it is also true that the very nature of the fission process means that the detection of such events is non-specific and cannot in itself provide an atomic number or mass identification. On the other hand, the use of alpha particle spectroscopy can readily achieve this goal.

We must expect that reports pertaining to such an important matter as the discovery of a new chemical element should fulfill the usual criteria for papers accepted for journal publication. In this context the two most important criteria are, first, that the experiments be carefully conducted and reproducible, and second, that the proof of the atomic number be demonstrated. We suggest that it is proper to carefully evaluate the work of the Dubna and Berkeley groups against these criteria.

A. Dubna Work on Element 104

1. Phase I--"The 0.3-sec spontaneous fission activity

In a paper submitted August 29, 1964, Flerov et al.[1] reported "the synthesis and physical identification of the isotope with mass number 260 of element 104." They bombarded a target consisting of 97% ^{242}Pu, 1.5% ^{240}Pu and 1.5% ^{238}Pu with ^{22}Ne ions, and reported that "with bombarding particles of energy 113-115 MeV an isotope is formed which undergoes SF with lifetime $\sim$0.3 sec and cross section $\sim 2.10^{-34}$ cm^2." The decay curve obtained is shown in Fig. 1. Notice that the error bars seem to indicate that little or no background has been subtracted from each point. They also determined the excitation curve shown in Fig. 2 and reported that "the excitation function of the isotope with $T_{sf} = 0.3$ sec was found to have a maximum at $E_{Ne} = 114$ MeV and a half-width of $\sim$10 MeV--which corresponds to the evaporation reaction (^{22}Ne,4n)."

In order to "verify that the observed effect ($T_{sf} = 0.3$ sec) was due to $^{260}104$," cross bombardments were also carried out. "It turned out that in the reactions $^{238}U + {}^{22}Ne$, $^{242}Pu + {}^{20}Ne$, and $^{242}Pu + {}^{18}O$, no isotope with half life 0.3 sec was formed."

The experimenters concluded that "the shape of the excitation function, the value of the cross section at the maximum, and the absence of the observed effect in control experiments with other particles and targets give good grounds for supposing that the

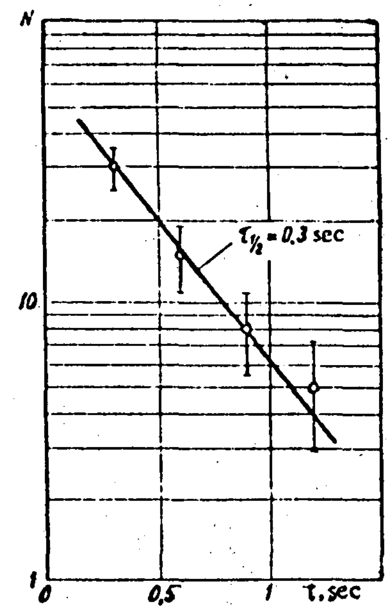


Fig. 1. Decay of spontaneously fissioning product formed by interaction of accelerated ^{22}Ne ions with ^{242}Pu.[1]

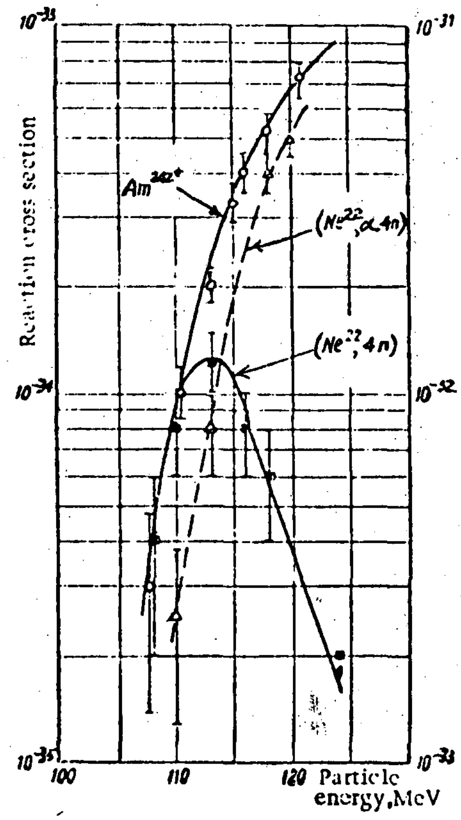


Fig. 2. Energy dependence of formation cross section for spontaneously fissioning products of the reaction $^{242}Pu + {}^{22}Ne$ (the right-hand scale refers to synthesis of ^{242m}Am). Experimental points: ■--synthesis of isotopes with $T_{sf} = 0.3$ sec; △--synthesis of isotope $^{256}102$; ○--synthesis of isotope ^{242m}Am.[1]

reaction $^{242}Pu(^{22}Ne,4n)$ gives rise to the isotope of mass 260 of element 104, which undergoes SF with half life of 0.3 ± 0.1 sec." They then suggested that the name kurchatovium be given to the element.

In a paper submitted May 18, 1966, Zvara et al.[2] reported a comparison of the "properties of the chlorides of Cm, Cf, Hf and the isotope ^{260}104." Using the same kind of ^{242}Pu target as in (1), the experimenters subjected the nuclear-reaction products to chlorination by means of a $NbCl_5$-$ZrCl_4$ vapor admixture in the nitrogen carrier gas. The gas was passed through a long heated duct that was lined with dielectric detectors.

In the first series of experiments the duct was kept at 200°-250°C; the time it took the gas to reach the detectors was 0.2 sec and the time it took to pass the detector area, 1.2 sec. The system was found to "trap" isotopes of Cm, Cf and Sc with a purification factor of 50 while there were "no losses of Hf in the chemical process."

Four fission events were recorded in this series with a total fluence of 4×10^{18} particles through the targets. This was "about 10 times lower than the effect expected for the isotope ^{260}104."

In the second series the temperature of the duct was raised to 300°-350°C; the time for the carrier gas to pass the detector area was reduced from 1.2 sec to 0.7 sec. In this series a fluence of 6×10^{17} particles through the target produced eight fission events "which corresponds to the expected effect. The coefficient of purification from Cm, Cf and Sc remained at the previous level."

These experiments were described in more detail by the same authors in another publication submitted in 1968.[3] It contains information about the conditions of the various experiments and the time distribution of the fission events observed.

The following statements in this paper are especially noteworthy:

a. The half life of 0.3 sec reported in (1) is stated to be based on 150 recorded events which seemingly would allow a statistical accuracy better than 10% if it was a pure activity.

b. The distribution is stated to show "positively that the effect was not caused to an appreciable extent by the decay of nuclides undergoing spontaneous fission with half lives of 0.014 sec and 3.7 sec." (Emphasis ours.) "All that has been stated above confirms that the fission acts recorded were due to the decay of the 0.3-second isotope." (Ref. 3, p. 167.)

2. Phase II--"The 0.1-sec spontaneous fission activity"

A significant change in the position of the Dubna group took place late in 1969 after the publication of our work on the discovery of the alpha-particle-emitting isotopes of element 104. At the Heidelberg Conference in August[4] Druin still stated that "the independent chemical technique has proved the assignment of the emitter with $T_{sf} = 0.3$ sec to element 104." However, at the Welch Conference in November, Zvara[5] presented the decay curve shown in Fig. 3 which shows no trace of a 0.3-sec activity! To illustrate the severity of the dilemma we show the earlier "0.3-sec" decay data together with the new data in Fig. 4.

Actually the half life shown by the straight

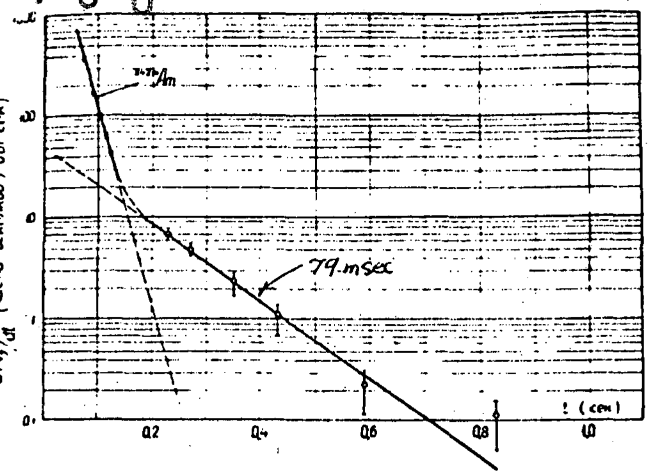


Fig. 3. The complex decay curve of spontaneously fissioning nuclides from the bombardment of a ^{242}Pu target with ^{22}Ne ions. The counting rate is given in counts per 0.01 sec interval.

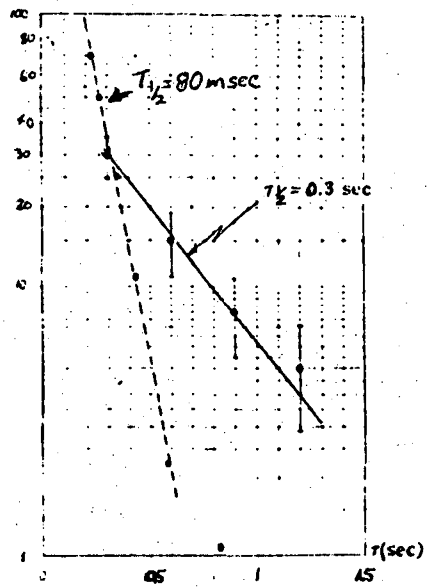


Fig. 4. The decay of the spontaneously fissioning product formed in the interaction of accelerated ^{22}Ne ions with ^{242}Pu. (Zero intercept adjusted to correspond with claimed cross section.)

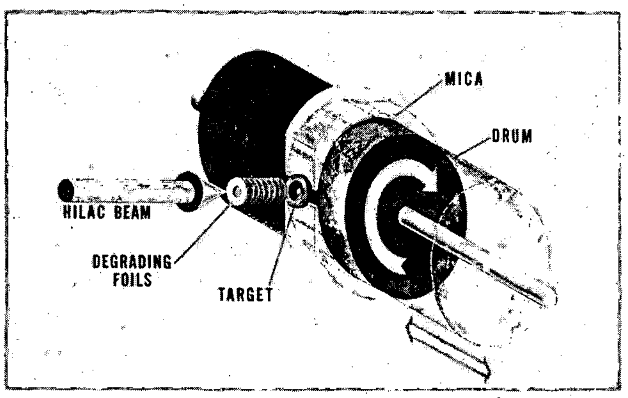
line in Fig. 3 is not 0.1 sec, but 79 msec! This, of course, makes the task of identifying it with the "0.3-second" activity still more onerous. A report issued in December 1969[6] states that the 0.3 second $T_{1/2}$ was caused by fast neutron background effects. How, then, does this explain the excitation function peak that was obtained (see Fig. 2)? Surely the fast neutron effects would increase as the energy was raised.

As if this had not been enough, it was also realized that the retention time of the "kurchatovium" atoms in the chemical experiments was of the order of one second before they reached the mica detectors.[7] In order to save the significance of the chemical experiments of Phase I it was thus necessary to invoke a longer-lived isotope of "kurchatovium"--but the longer-lived activity seen in the early experiments had already been positively ascribed to ^{256}No.[8] Nevertheless, they assigned it as a small fission branching of an isotope of element 104 that we had just discovered, the 3-sec alpha emitter, $^{259}104$. It is well to remember that there is no independent proof that this nuclide has the SF branching ratio that they claim.

B. Berkeley Attempts to Reproduce the 0.1 sec (née 0.3-sec) Spontaneous Fission Activity

In 1964 we were inclined to accept the Soviet work on element 104 since it sounded convincing but our doubts were aroused a little later when we discovered the nuclide ^{258}No (which has the same number of neutrons as $^{260}104$) and found that it had an SF $T_{1/2}$ of only a millisecond in agreement with a prediction by our empirical systematics (Ref. 9, p. 143).

We decided to try and produce the same nuclide that Dubna had made in their bombardments of ^{242}Pu with ^{22}Ne by bombarding curium isotopes with oxygen ions to take advantage of the larger cross sections that were known to prevail. The reactions attempted were ^{246}Cm(^{18}O,4n) and ^{248}Cm(^{16}O,4n) to make $^{260}104$. The apparatus was a rotating wheel device which carried the reaction recoils in front of mica dielectric track detectors (see Fig. 5). We were not successful even though our sensitivity, as judged by the known cross sections for making similar reactions in this region, was such that we should have observed the SF activity if its half life was as long as a few hundred milliseconds. These experiments were reported at the Welch Conference in 1969.[9]



CBB 689-5714

Fig. 5. Drum-Mica Assembly (used in earlier experiments).

More recently, in 1975, we again made an attempt to find a 0.1-sec $^{260}104$ by bombarding ^{249}Bk with ^{15}N ions. The calculated σ at that time was a minimum of 14 nb, a value based on the measured cross section to make $^{260}105$ via the (^{15}N,4n) reaction on ^{249}Cf and hence thought to be reasonably accurate. We calculated that we should have observed 200 SF events of the hypothetical 0.1-sec activity in the short 2 μahr bombardment. We observed only 20 events and these were distributed in time such that they must have been due to the well-known 2.7-hr ^{256}Fm nuclide. We set a limit of <0.7 nb for the production of the 80 msec activity and this was reported at the Dubna School-Seminar in September 1975.

C. The Berkeley Discovery of the Element 104 Alpha Particle Emitters

Early in 1968 enough ^{249}Cf had become available to allow us to bombard it effectively with ^{12}C ions and allow a search for short-lived alpha emitters. After some initial experiments which disclosed the existence of a 3-5 sec complex alpha spectra from 8.7 to 9.0 MeV, we began the development of a system which used a large wheel as a conveyor for helium-jet-deposited atoms (see Fig. 6). The periphery was used to place successive deposits in front of a series of solid state alpha particle detectors mounted around the rim of the wheel. In addition, we equipped it with auxiliary detectors to allow us to identify the element 102 daughter products simultaneously with the identification of the element 104 mother atoms.

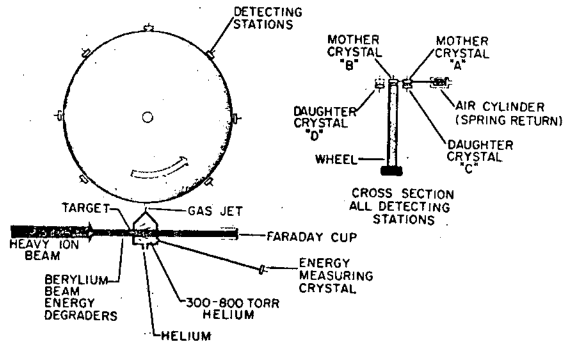


Fig. 6. Vertical-Wheel Helium-Jet System.

We felt that the definitive proof of the identity of an element 104 alpha emitter would be the establishment of a mother-daughter relationship to a known isotope of element 102, nobelium. Such mother-daughter measurements were accomplished in the following way. Advantage was taken of the physical separation of daughter from mother afforded by the recoil energy imparted by alpha particle decay. Periodically the detecting crystals which had been "looking at" the alpha particles emitted by the mother atoms on the conveyor wheel were moved to positions off the wheel. We were thus able to measure by itself the daughter alpha activity coming from those atoms which had been transferred to the crystals as a result of alpha recoil. To increase the likelihood of detection of these daughter atoms, the shuttled-positions of the crystals were next to a similar set of detectors to establish almost a 4π geometry. Ultimately we employed seven detecting stations around the wheel. To avoid the loss of detection of element 104 atoms while measuring the recoil daughters, we used a duplicate system so that we could measure the mothers and daughters simultaneously. The final complex system used 28 detectors and was found to perform with commendable accuracy and stability.

By early 1969 we had identified without ambiguity the isotope $^{257}104$ in ^{12}C bombardments of ^{249}Cf. It was found to have a complex alpha spectrum with lines ranging from 8.70 to 9.00 MeV and a half life of 4.5 ± 1.0 sec. Its known daughter, ^{253}No, was found to be transferred by alpha recoil as expected.

The ^{249}Cf target was soon bombarded by ^{13}C ions at Berkeley to produce another isotope of element 104, this one with mass 259 having two prominent groups at 8.77 and 8.86 MeV and a shorter half life of 3 seconds. Again the recoil daughter was identified, this time ^{255}No. We deduced that this nuclide must undergo branching electron-capture decay to the extent of ca. 50% because of the ratio of mother to detected-daughter atoms. This interesting sidelight has been confirmed by the Oak Ridge group with an independent experiment on ^{255}No. Our work was published in April 1969.[10]

Somewhat later we performed fast rotating drum experiments with mica detectors to detect SF products from ^{13}C and ^{12}C bombardments of ^{249}Cf and discovered an 11 msec* SF emitter which we deduced was probably ^{258}104. Although we produced thousands of events, we were not able to prove to our satisfaction that our assignment was unambiguous; and indeed at a high bombarding energy in the ^{12}C bombardments, we observed a 5 msec SF activity! This may be from an unknown isomer in a lower Z element, but this speculation has not been investigated as yet.

About a year later, in bombardments of ^{248}Cm with ^{18}O ions, a much longer-lived isotope of element 104 was found,[11] ^{261}104, with a half life of 65 ± 10 seconds and an alpha energy of 8.3 MeV. This time the known recoil daughter, ^{257}No, was shown to be transferred into the detecting crystals as they viewed the wheel. This discovery of ^{261}104 ruled out the dim possibility that the Dubna 0.3-sec SF activity could be assigned to that isotope.

In 1969 the first aqueous chemistry was performed with element 104 using the 65-sec ^{261}104. Several hundred individual experiments were made using a cation-exchange column with the aim of showing that element 104 could be separated from the actinide elements.[12] Seventeen events were observed in elution position indicating that element 104 was similar to its predicted homologue hafnium.

D. The Oak Ridge Confirmation of the Berkeley Work on ^{257}104

In 1973, Bemis et al.[13] at the Oak Ridge National Laboratory undertook a different type of experiment to see whether or not they could confirm the Berkeley work on the element 104 alpha emitters. They also used the helium gas jet technique in bombardments of ^{249}Cf by ^{12}C ions but in their case the activities were deposited onto the surface of a mechanical shuttle. This "rabbit" was used to transport the atoms to a shielded remote area where α-x-ray coincidences could be observed with high efficiency. By measuring the $K\alpha_1$, $K\alpha_2$, $K\beta_1$, $K\beta_2$ characteristic x-rays of element 102, which were in coincidence with alpha particles from element 104, they were able to make an independent confirmation of the atomic number of the isotope, 4.5-sec ^{257}104.

E. Recent Work on the 0.1-sec SF Activity at Dubna

The next step in the course of action designed by the Dubna scientists to defend their claim to the discovery of element 104 occurred late last year. At the School-Seminar held at Dubna in September 1975 they announced new work which contradicted the Berkeley findings (or rather non-findings!). Their experiment consisted of the bombardment of ^{246}Cm with ^{18}O ions to make the same 0.1-sec SF activity that they had observed in ^{242}Pu bombardments by ^{22}Ne ions years earlier. The curium target was 16 mm in diameter, had a surface density of 0.8 mg/cm^2, and was bombarded with 94 MeV ^{18}O ions with an intensity of 3 microamperes. The recoil products knocked out of the target traveled 50 mm through helium at about 10 torr pressure to lodge in a Ni tape. The 10 micron thick tape was 25 mm wide and 800 meters long. The tape was run at three different speeds, 150, 300 and 600 msec/plate, alternately in each direction to the end of the tape to carry the recoils next to a series of phosphate glass SF detectors. The observed fission tracks in each plate were combined in number for the corresponding plate positions for the two tape directions to give a spontaneous fission decay curve. The first plate on each side of the target was 40 mm (2/3 of a plate length) from the area where the recoils were collected, so that at their highest tape speed there was a factor of two decay for a 0.1-sec component.

The Dubna group claimed that the data from these experiments proved conclusively that their 0.1-sec activity was formed with a cross section of 1.5 nb; they also claimed that a comparable amount of a 1.1-sec SF activity was formed which was most likely due to a new nuclide, ^{262}104, formed by an ^{18}O,4n reaction with ^{248}Cm, present in their target with a 10% abundance.

---

* A recent re-evaluation of this old data gives a better value of 13 ± 2 msec. (Private communication, M. J. Nurmia.)

During the School-Seminar a discussion was held between Dubna and Berkeley scientists as to the best procedure that might resolve the disparate findings of the two laboratories. We agreed to repeat our bombardments of ^{249}Bk with ^{15}N once more, this time with a representative from their laboratory present when the experiments were done. Bombardments of ^{246}Cm with ^{18}O and ^{248}Cm with ^{16}O were also contemplated if necessary, but they were deemed of lesser importance since the cross sections for producing ^{260}104 are substantially smaller in those cases compared to the use of ^{249}Bk.

F. Recent Work on the 0.1-sec SF Activity at Berkeley

Back in Berkeley we set about to prepare for the new set of experiments. It was decided to use the 88" Cyclotron instead of the SuperHILAC because the latter machine was going to be shut down for the next several months for major alterations. We also decided to build a new apparatus so that our new experiments would be more sensitive.

In the meantime, we went ahead with some preliminary bombardments of curium with oxygen ions using our old rotating drum equipment. A bombardment of ^{246}Cm by ^{16}O ions produced 13-msec ^{258}104 with a σ of about 4 nb in agreement with calculation, and bombardments of ^{246}Cm by ^{18}O and ^{248}Cm by ^{16}O revealed an ∿20 msec SF activity just as they had in 1968 (reported at the 1969 Welch Conference as a 10-30 msec activity), but no 0.1-sec nuclide showed up. More will be said about these results later.

We had invited scientists from three other laboratories to join us in the new experiments to make sure that the results obtained would have the most careful scrutiny possible. Collaborating on this work were the following: from Lawrence Livermore Laboratory, E. K. Hulet, R. W. Lougheed, J. F. Wild and J. H. Landrum; from Oak Ridge National Laboratory, R. J. Silva and C. E. Bemis; from Los Alamos Scientific Laboratory, M. Fowler; and from Lawrence Berkeley Laboratory, P. A. Baisden, I. Binder, E. Leber, D. Lee, D. Morrissey, J. M. Nitschke, M. J. Nurmia, R. J. Otto, L. P. Somerville, K. Thomas and K. E. Williams.

The new rotating drum equipment in principle was basically the same as that used in our old experiments. An important difference was the use of a longer drum to spread out the long-lived SF background activity from ^{256}Fm produced in the bombardments from its e.c.-mother, ^{256}Md. The ^{249}Bk target used consisted of ca. 85 μgms of the element (480 μgm/cm^2) vacuum evaporated as BkF_3 onto 2.08 mg/cm^2 Be in a circle 4.8 mm in diameter. The target was placed 21 mm from the surface of a 254 mm diameter water-cooled drum which was rotated typically at 60 revolutions per minute. The nuclear reaction recoils passed in vacuum through an opening 20 mm square to strike the drum surface. The 360-mm long drum was scanned axially, usually at a speed of 7.7 mm/sec The mica fission track detectors, 31 mm wide and 50 mm long, were held in a curved holder about 0.8 mm away from the drum surface and completely surrounded the drum in a 31 mm slice.

The ^{15}N ion energies used were obtained by three settings of the cyclotron and the use of Be foil degraders. The nominal values of the energies out of the cyclotron, calculated from the cyclotron frequencies, were 91.6, 93.6 and 105 MeV, thought to be accurate to better than 0.5 MeV. In the 93.6 MeV case a special analysis was made by D. Hendrie using the large beam analyzer magnet and the energy was found to be 93.74 ± 0.01 MeV. The Be degraders used to obtain lower energies were weighed and the final energies at the 0.5 mg/cm^2 ^{249}Bk target were obtained by calculation. The drop in energy of the ^{15}N ions through the Bk itself was about 0.5 MeV. The early experiments showed that the beam profile attained a needle-sharp focus when tuned for maximum beam current and indeed we melted small holes through two old targets with currents in the vicinity of 3-5 charge microamperes. A 3-phase 60 Hz wobbler was installed about 5 meters upstream following the last beam quadrupole magnet. The wobbler was simply the stator of an induction motor and succeeded in moving the beam in a small circle at a 60 Hz rate with the loss of about 15% of the beam intensity. With this arrangement we were able to use beam currents as high as 4 μa, but normally we limited the beam to about 3 μa to have a margin of safety. An infrared detection system was installed to guard against any unusual excursions of the beam and was similar to the one used in our element 106 experiment.[14]

The beam current was measured by recording the current to the insulated drum. Normally this did not give us any difficulty because a weak magnetic field near the surface of the drum trapped secondary electrons. Checks of the beam that penetrated through a test hole in the old drum gave the same reading into a standard magnetically protected faraday cup. In the new equipment, however, no test hole had been provided, and we had no such check available. When a thin foil was placed over the target we found that the beam reading was substantially reduced, ostensibly because some secondary electrons emitted from the foil (which was about 5 mm closer to the drum) were able to reach the drum. A last minute solution to the problem was afforded by temporarily placing a suitable faraday cup between the target and the drum and calibrating the misleading low drum reading. An upstream cup which read the total beam was used as a monitor to derive the calibration ratios when necessary. Normally during the drum runs the faraday cup was removed and the target was then moved closer to the drum.

We found it advantageous to put a very thin (26 μgm/cm^2) aluminum foil between the target and the drum to prevent any of the target material from being knocked out by the beam into the drum surface and thus increasing the spontaneous fission background. This also served as a rough monitor of the beam current since as much as 5% of the ^{256}Md that was produced stopped in these foils.

In addition to the regular drum experiments we made measurements of the long-lived recoil products directly. This was accomplished with a special holder mounted next to the target which could accommodate as many as four foils. In these experiments the external faraday cup mentioned previously was used to give accurate beam readings. An excitation function for ^{256}Md is shown in Fig. 8 and was obtained by

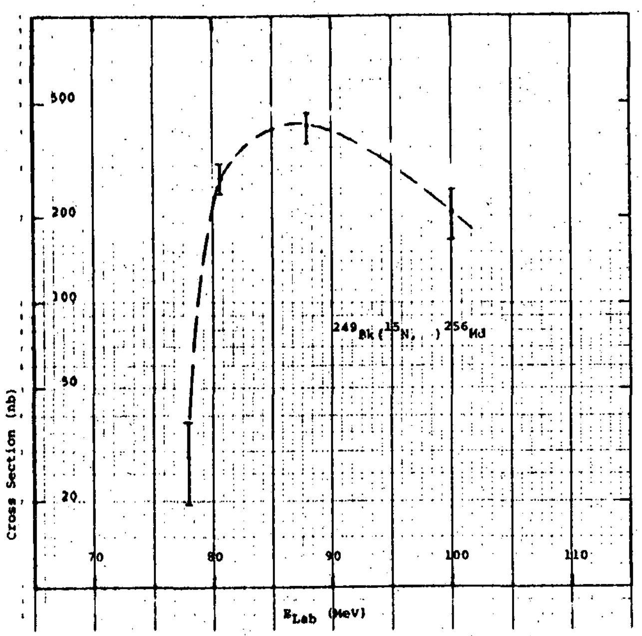


Fig. 8. Excitation function for ^{256}Md.

stopping its recoils in 1.0 mg/cm^2 Al and counting the foils directly in a fission counter. The growth and decay in one of these runs, a 1.65 μahr bombardment at 88 MeV for 2.0 hrs shown in Fig. 9, shows

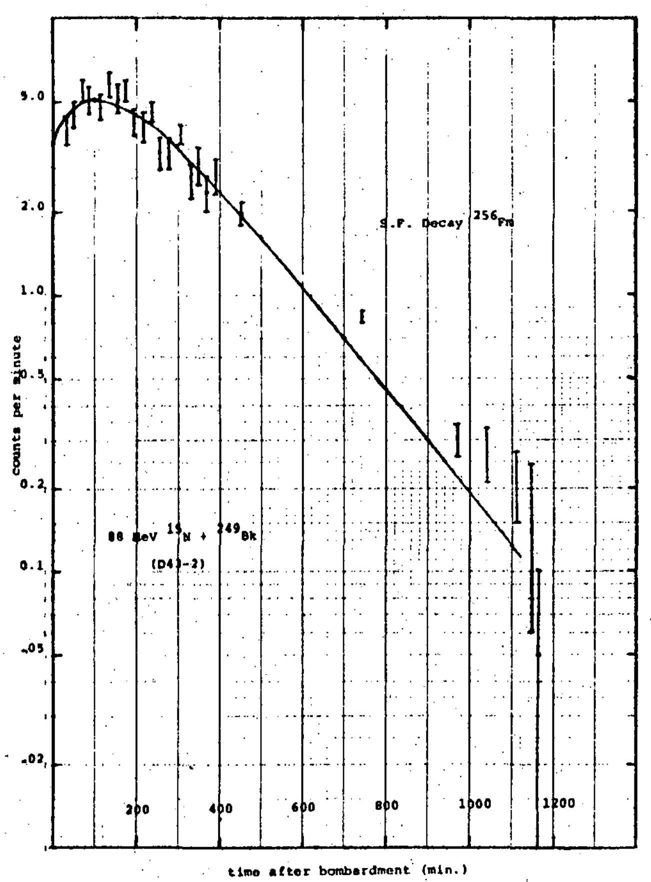


Fig. 9. Growth and decay curve of ^{256}Fm (from ^{256}Md)

quite clearly that very little ^{256}Fm is produced directly since the data can be fitted by a curve, assuming that only ^{256}Md is produced.

Dr. Ivo Zvara from Dubna arrived a few days before the berkelium bombardments were scheduled to begin, and we discussed the latest developments. By far the most important was the news that the Dubna team had bombarded a small 50 μgm/cm^2 ^{249}Bk target with ^{15}N ions and was in the process of scanning their track detectors for signs of their 0.1-sec SF emitter. By telephone Zvara was informed that they had succeeded in finding some 60 tracks in about 20 μahrs which they felt were due to this activity and that the cross section for its production was roughly 5-10 nanobarns. With this information it became clear that the berkelium bombardments should take precedence over those with curium targets. Approximately a hundred hours of beam time were used during the first week in May 1976 for this purpose. The berkelium target was prepared at the Lawrence Livermore Laboratory after being chemically separated from its californium daughter just a few days before the first bombardment. This timing of the separation is important because the half life of ^{249}Bk is only 314 days and it very soon becomes contaminated by its 360-yr daughter.

The series of bombardments were then carried out with the chief objective being the attempt to see whether the latest Dubna work could be confirmed. It was not and we decided to make as many cross checks as we could so that there would be no question of the validity of our attempts.

In comparing the experiments in the two laboratories one should keep in mind the relative advantages and disadvantages of the two methods used. First, the Berkeley experiments made use of a target which was ten times as thick as that used at Dubna. Second, at Berkeley our geometry registered SF tracks within 5 msec from the time that atoms to be detected were made whereas Dubna had a delay of about 75 msec, one half life of the activity in question. Third, we employed mica detectors with an inherent efficiency of about 90% compared to 30% for the Dubna glass detectors. The product of these three advantages is about 30 so that our production rate was that many times greater for the same beam current, and I believe that we did use roughly the same current, ∼3 μa measured as N^{7+}. On the other hand, the one serious disadvantage that we had to cope with was the fact that we spread out the $^{256}Md \rightarrow {}^{256}Fm$ on an area of about 3000 cm^2, whereas Dubna used a tape that was 500 meters long. This disadvantage cost us a factor of ∼20 when one considers the details of the experiment and it turns out to be the element that limits our final sensitivity; however, we feel that the cross section limit that we can quote is low enough to rule out the claim that ^{260}104 has a $T_{1/2}$ of 80 msec.

Bombardments were made with 78, 82, 86, 88 and 100 MeV ^{15}N ions entering the target. There is general agreement that the cross section for the ^{249}B(^{15}N,4n)^{260}104 reaction should peak at about 82 MeV so this series adequately covers this range. Approximately 40 μahrs of beam were devoted at each of these energies in the search for the 80 msec activity. The results of the bombardment at 82 MeV are shown in the decay curve in Fig. 10. A calculated computer fit is also plotted. As can be seen, the maximum amount of the 80-msec activity that can be extracted from these data is very small. It corresponds to a cross section of less than 0.5 nanobarns if an excursion 2 σ higher than observed is allowed.

These decay curves were extracted from the raw

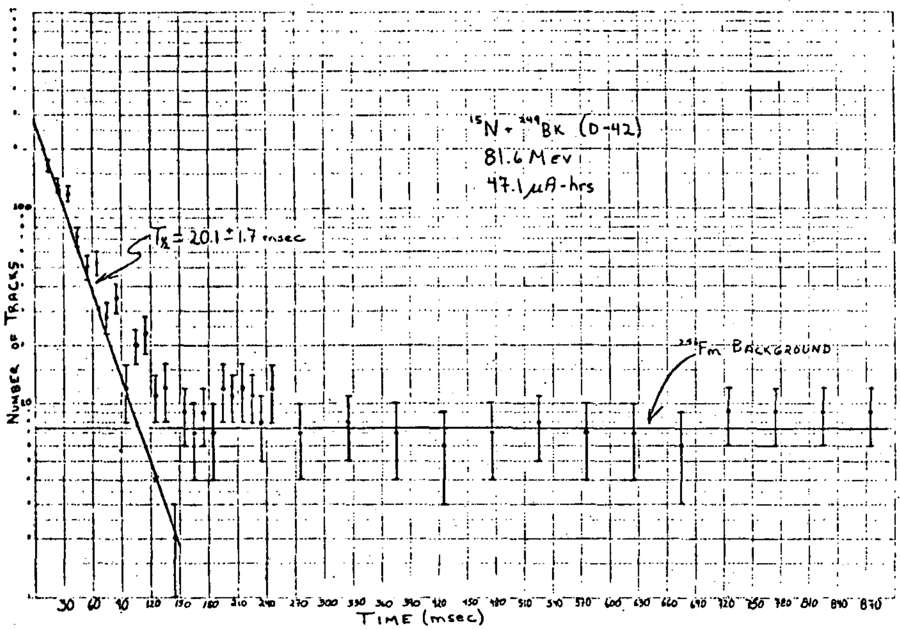


Fig. 10. Decay curve for spontaneous-fission events detected at 81.6 MeV. Solid lines represent computer fit to the data.

scanning data by the following method. An early test run with a thick ^{241}Am target to produce the 2.3-sec ^{252}No via the ^{15}N,4n reaction at 82 MeV gave us an actual situation of recoiling nuclei made by complete fusion. The angular distribution of these recoils is very narrow, being broadened mostly by multiple scattering within the target. When the tracks were summed in slices along the direction of rotation of the drum, the distribution shown in Fig. 11 was obtained; such a distribution indicates that most of the fusion-produced nuclei can be measured by selecting a relatively narrow "window" in the transverse direction. Since in the case of the berkelium bombardments the background activity is the long-lived $^{256}Md \rightarrow ^{256}Fm$ which is spread out on the entire drum surface, we gained about a factor of 2 in signal-to-background ratio by using this method.

To make sure that our track counting efficiency was normal, a few test exposures of our mica were

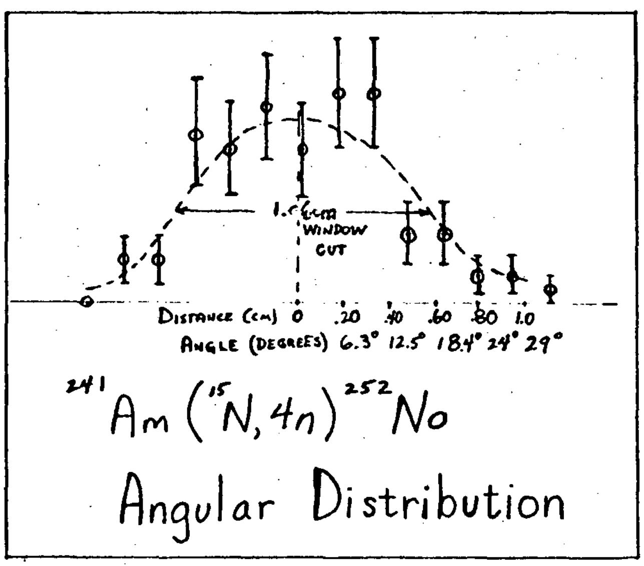


Fig. 11. Transverse distribution of ^{252}No fission tracks from $^{241}Am(^{15}N,4n)$ reaction at 81 MeV.

made from the source with and without an aluminum foil 250 μgm/cm^2 in thickness. Our average efficiency was 90%.

Though our decay curves show no hint of the so-called 80-msec SF activity, they very clearly show one with a half life of ∿20 msec.* This same activity was first reported by us at the Welch Conference in 1969 (Ref. 9, p. 148). Let me quote from that report.

> *"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 $^{260}104$ 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....*

> *"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 $^{260}104$ although it seems that such a half life is much too long. For that matter the 1-millisecond activity could also include $^{260}104$ as well as ^{258}No; however it seems to us more likely that the $^{260}104$ half life is in the microsecond range."*

The maximum cross section for producing the "∿20 msec" activity is about 10 nb when made by ^{15}N ion bombardment of ^{249}Bk and has the excitation function shown in Fig. 12. However, on the basis of this data alone we cannot exclude the possibility that the yields include some fission from the 13.7-msec SF isomer, ^{242m}Am, produced by some sort of transfer or other reaction. The transverse track distribution for the "∿20-msec" activity is

* It is quite possible that the "∿20 msec" activity has both a short and a long component to give an average value of about 20 msec.

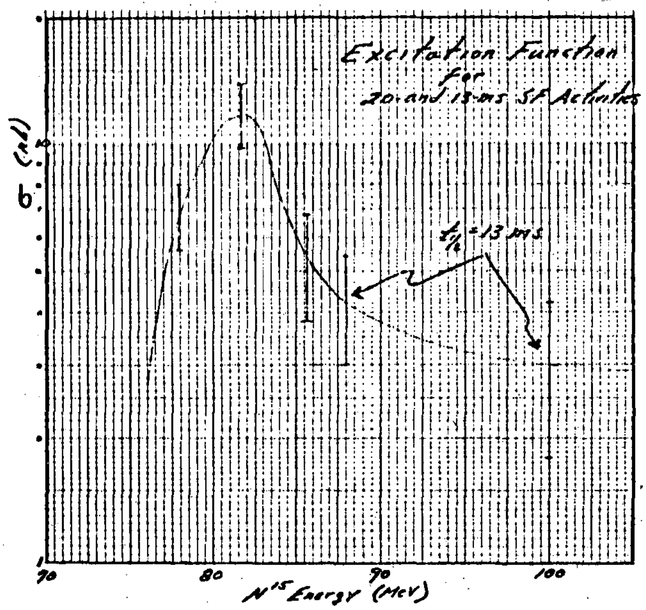


Fig. 12. Excitation function of the "∿20-msec" spontaneous-fission activity.

remarkably narrow at 82 MeV (see Fig. 13). These data would seem to point to a complete fusion reaction except that its production σ is lower than expected (see below). Just prior to undertaking the berkelium bombardments, we bombarded ^{248}Cm with ^{16}O ions and ^{246}Cm with ^{18}O ions at about 93 MeV and once again observed the "∿20-msec" SF activity without observing the hypothetical 80-msec Dubna activity. No excitation function was measured because of lack of time. It can be seen that there is some possibility that the "∿20-msec" activity includes ^{260}104 since it passes some of the tests required. It may take some time before we can state with certainty whether or not we have observed this nuclide, and this situation points up again the great difficulties that are encountered when working with spontaneous fission activities.

Although we set a maximum cross section for the ^{15}N,4n reaction on ^{249}Bk to produce the 80-msec activity which was more than an order of magnitude

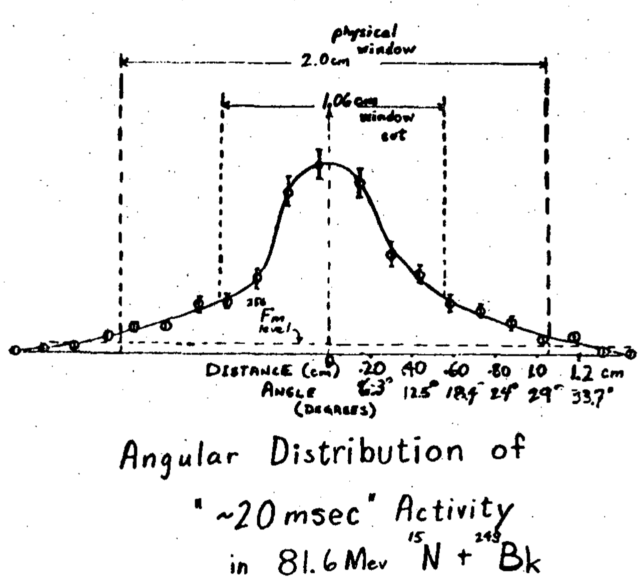


Fig. 13. Transverse track distribution of the "∿20-msec" spontaneous-fission activity at 81.6 MeV.

less than the 5 nb claimed by Dubna, it should be remembered that the actual cross section to produce ^{260}104 by this mechanism is expected to be about 25 nb! The reasoning is as follows: the cross section for the identical reaction to make ^{260}105 in bombardments of ^{249}Cf has been found experimentally to be 12 nb.* Surely the cross section to make ^{260}104 by the same reaction should be larger since the values of $\Gamma_n/\Gamma_n + \Gamma_f$ and R_0 are more favorable. Indeed a calculation adjusted to the ^{260}105 production cross section shows that the production of ^{260}104 will be about twice as large. Our limit on the amount of the 80-msec activity that is made in the ^{249}Bk plus ^{15}N reaction is thus almost two orders of magnitude below that expected if it is due to ^{260}104.

Although this is only a preliminary report (a more complete analysis will be published forthwith), there is no escape from the categorical conclusion that can be drawn on the basis of our data. The isotope of element 104, rutherfordium, does not decay by spontaneous fission with a half life of "0.3 sec," or "0.1 sec," or "80 msec."

It has been 10 years that we have been chasing this will-o-the-wisp and many people-months have been expended in the efforts to confirm or deny the validity of the spontaneous fission results of the Dubna group. We know now that it is a thankless chase and wasteful of scientific talent and we do not intend to pursue this any further with other elements. The claim to the discovery of a new element should stand or fall on the merits of the work itself; any premature claim that cannot stand the scrutiny of the scientists in the field should not be regarded as any claim at all. It is now clear that the mere counting and measurement of a half life of a spontaneous fission activity does not provide the necessary proof of the atomic number of a new element. Other independent confirming evidence must be provided that will stand the test of time.

We extend our thanks to the staff and operations crew of the 88" Cyclotron. Their help was invaluable to the success of these experiments.

I would like to acknowledge with gratitude the concentrated effort by my many colleagues that went into the task of making these latest experiments so definitive. In particular I would like to thank Glenn T. Seaborg for following developments very closely and providing inspirational and scientific guidance.

---

* This new value was obtained by a re-evaluation of our old data by Pirkko Eskola and appears in her recent thesis, University of Helsinki.

References

1. G. N. Flerov et al., Atomnaya Energiya 17, 310 (1964); Sov. J. Atomic Energy 17, 1046 (1964); Phys. Lett. 13, 73 (1964).

2. I. Zvara et al., Atomnaya Energiya 21, 83 (1966); Sov. J. Atomic Energy 21, 709 (1966).

3. I. Zvara et al., Radiokhimiya 11, 163 (1969); Sov. Radiochem. 11, 161 (1969).

4. V. A. Druin, Proc. of Int. Conf. on Nuclear Reactions Induced by Heavy Ions, Heidelberg, July 1969 (p. 663).

5. I. Zvara, Proc. of the Robt. A. Welch Foundation Conf. on Transuranium Elements, Houston, Nov. 1969 (p. 161).

6. Y. Oganesyan et al., Atomnaya Energiya 28, 393 (1970).

7. I. Zvara et al., Sov. Radiochem. 14, 119 (1972).

8. While it was stated in Ref. 1 that the longer-lived component of the spontaneous-fission background was $^{256}102$ (T_{sf} = 1500 sec), this assertion was modified in Ref. 3 to attribute the activity to one of the heavier transuranic elements.

9. A. Ghiorso, Proc. of the Robt. A. Welch Foundation Conf. on Transuranium Elements, Houston, Nov. 1969 (p. 107).

10. A. Ghiorso et al., Phys. Rev. Lett. 22, 1317 (1969).

11. A. Ghiorso, et al., Phys. Lett. 32B, 95 (1970).

12. R. Silva et al., Inorg. Nucl. Chem. Lett. 6, 871 (1970).

13. C. E. Bemis et al., Phys. Rev. Lett. 31, 647 (1973).

14. A. Ghiorso et al., Phys. Rev. Lett. 33, 1490 (1974).