

Lawrence Berkeley National Laboratory

Recent Work

Title

ELEMENT 102, A TEN YEAR HISTORY OF CONFLICTING CLAIMS

Permalink

<https://escholarship.org/uc/item/4xt5916m>

Authors

Ghiorso, Albert
Sikkeland, Torbjorn.

Publication Date

1967-03-01

UCRL-17427

University of California
Ernest O. Lawrence
Radiation Laboratory

ELEMENT 102, A TEN YEAR HISTORY OF CONFLICTING CLAIMS

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*

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.

Submitted to Science

UCRL-17427
Preprint

UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
Berkeley, California

AEC Contract No. W-7405-eng-48

ELEMENT 102, A TEN YEAR HISTORY OF CONFLICTING CLAIMS

Albert Ghiorso and Torbjorn Sikkeland

March 1967

ELEMENT 102, A TEN YEAR HISTORY OF CONFLICTING CLAIMS

Recent experiments at Berkeley have made it possible to clarify the complex history of the discovery of element 102.

Albert Ghiorso and Torbjorn Sikkeland

Author note:

The authors are with the Nuclear Chemistry Division, Lawrence Radiation Laboratory, University of California, Berkeley, California.

Introduction

The synthesis and identification of the chemical elements with atomic numbers greater than 100 (fermium) have been accomplished with extremely small amounts, i.e., literally on a one-atom-at-a-time basis. These elements can only be formed at the present time by the use of accelerators of ions ranging from neon to helium to bombard targets ranging from uranium to einsteinium. Because of their miniscule transmutation cross sections, such experiments are extremely difficult to perform and the results have often been subject to uncertainty of interpretation and, not unexpectedly, occasionally have been in error.

The work on the synthesis of element number 102, started about ten years ago, was particularly fraught with difficulties and pitfalls. It is only recently that experimental results have given sufficient information to make it possible to place the whole picture in historical perspective. The intent of this paper is to present a comprehensive chronological review of the investigations concerning this element.

In a sense the story of element 102 begins with the discovery (1) of element 101 (mendelevium), which occurred in 1955 at the Lawrence Radiation Laboratory of the University of California at Berkeley. Element 101 was the first to be discovered on an atom-by-atom basis and the techniques which made this possible were applied to the work on element 102. Chief among these were the "recoil technique" and

the method of chemical identification. In the first case the momentum imparted by the impinging projectiles to the product nuclei is used to eject them from a thin target layer and thus effect an immediate physical separation from the highly radioactive target. In the second case, for the chemical separation of the actinide elements heavier than plutonium, a cation-exchange adsorption-elution resin column is used with ammonium α -hydroxy-isobutyrate as eluant. (2).

Early Work at the Nobel Institute

The first published attempt (3) to synthesize and identify element 102 was made in 1957 by an international team of scientists from ^{the}Argonne National Laboratory in Chicago, ^{the}Harwell Atomic Energy Research Establishment in England and the Nobel Institute for Physics in Stockholm. In the cyclotron at the Nobel Institute, these investigators used a rather small internal beam of C^{13} ions with a very wide energy spread to bombard Cm^{244} (containing 4% Cm^{246} , 1% Cm^{245} and much smaller amounts of Cm^{247} and Cm^{248}). They made use of the recoil technique to separate any possible element 102 nuclei from the thin curium target and under extremely difficult experimental conditions involving many runs they observed by the use of a Frisch grid ionization chamber a few alpha particles with an energy 8.5 ± 0.1 MeV and a half-life of about 10 minutes. They performed an ion-exchange experiment in which this activity was washed from the column 2 to 3 drops after the passage of one free-column volume of eluant. This was interpreted as an indication that the four detected atoms had the

atomic number 102 since this element would be expected to have an early elution position in front of the lighter actinides. It appears now that there would have been an inadequate separation from the element thorium under the conditions used.

On the basis of these experiments the group suggested for element 102 the name nobelium (symbol No) to honor Alfred Nobel, whose name was given to the Institute where the work was done, and who made such a significant contribution to the advancement of science through his Nobel Prizes. This name was accepted almost immediately by the Commission on Atomic Weights of the International Union of Pure and Applied Chemistry. Judged in its entirety now it must be realized that this initial work, performed under adverse circumstances, was only capable of yielding marginal results. At that time there were many uncertainties so that even in 1959 a re-evaluation (4) of the 1957 data was not able to significantly clarify the early experiments. To our knowledge no attempts to repeat this work have been made by these investigators during the intervening years to the present.

Early Work at Berkeley

Such an attempt (5) was made very soon thereafter, in 1958, by our group at the Lawrence Radiation Laboratory at Berkeley. With great persistence we tried to reproduce the results of the Stockholm work by using the more intense, monoenergetic C^{13} (and C^{12}) ion beams available from the then-new heavy ion linear accelerator (HILAC) to bombard an external curium target having a similar isotopic composition to that used at the Nobel Institute. In scores of experiments conducted with the same

and varied techniques over a period of many months we were unable to produce any nuclei of element 102 with the reported properties (8.5 MeV α particles and 10 minutes half-life). Under the superior conditions of these repeated experiments it was estimated that we should have produced and observed at least 100 such nuclei in each experiment if the Nobel Institute findings were correct; this estimate was made on the basis of the measured yield in the Berkeley work of well-known products which had also been produced in the Stockholm experiment.

These negative experiments were immediately followed in 1958 by experiments of a different character which were successful in producing and identifying an isotope of element 102 (6). The discovery team of Ghiorso, Sikkeland, J. R. Walton, and G. T. Seaborg, utilized a radically new method, the "double-recoil technique." The HILAC was used to bombard the thin target of curium (95% Cm²⁴⁴ and 4.5% Cm²⁴⁶) with C¹² ions to produce 102²⁵⁴ according to the reaction Cm²⁴⁶(C¹², 4n). The product atoms, recoiling out of the curium target in an atmosphere of helium, were attracted by a moving electrically-charged metallic belt; in a second recoil action approximately half of the 30-min Fm²⁵⁰ daughter atoms from the ^{alpha} decay of 102²⁵⁴, recoiling from the surface of the belt, were captured by a negatively-charged catcher foil. The key experiments upon which the discovery of element 102 was based, consisted of the chemical identification of these daughter atoms by elution from an ion exchange column in the well-known elution position of fermium. For these crucial experiments it was necessary to use all of the transferred daughter atoms in each experiment in order to make the chemical identification possible and thus no half-life information was obtained in these milkings.

The half-life of the parent was deduced by a completely physical method. From the speed of the belt and the variation in the distribution of the Fm^{250} alpha activity along the catcher foil, information was obtained that was consistent with a half-life of approximately 3 seconds for the 102 parent of this alpha emitter (6). A year later, in experiments in which the element 102 atoms were electrostatically attracted to a stationary metallized tape which was periodically moved into a counting position within a gridded ionization chamber, an alpha activity of 8.3 MeV energy and half-life also about 3 sec was found (7,8,9). Surprisingly, spontaneous fissions were also observed with the same half-life with an abundance of about 30%. Because of the sharply peaked excitation functions to produce these activities and because of the half-life it was quite naturally assumed that they were both most likely associated with 102^{254} , the isotope which had been identified as described above through the chemical identification of its daughter isotope Fm^{250} .

Before this work could be followed to a satisfactory conclusion it was abruptly halted by a disastrous incident. A helium-cooled window interposed between the curium target and the HILAC beam tube was explosively ruptured by an accidental momentary overpressure. We had always considered that the system was "fail-safe" in this regard because it was assumed that the thin vacuum-isolating window on the HILAC side would break rather than an identical window on the target side which had an atmosphere of helium on each side. (To protect the HILAC a very fast valve had been installed upstream.) For reasons unknown, this 14.7 psi "fuse" did not function and when the foil ruptured on the target side only, a large amount of helium impinged directly against the very active and very fragile curium

target. The curium oxide, emitting $\sim 10^{12}$ α dis/min, was broken into micron-size particles and carried out into the experimental room. Since at that time there was no secondary containment in this area, only heavy lumber shielding with many cracks, the dangerous alpha-active dust was quickly carried throughout the building to completely contaminate everything! Fortunately a hasty evacuation of the building kept anyone from ingesting an alarming amount of curium. Three weeks later, after a tremendous effort by Health Chemistry personnel, it was feasible to work in the building again but many years went by in which curium continued to be found in obscure out-of-the-way places.

The accident not only disposed of all of the precious curium available to us at that time; it also quite naturally made us "gun-shy" in the use of highly alpha-active targets. It has only been recently that we have been willing to undertake such experiments and, of course, now they are conducted with elaborate precautions.

We could have turned our attention to the use of plutonium targets and oxygen ions to form element 102, but it was known that the combination of smaller cross sections and weaker HILAC beams would probably make such experiments very unfruitful. Instead we decided to push on and try to produce element number 103 by bombarding the small amounts of californium available at that time with boron ions. In 1961 in the course of this work which did successfully lead to the identification (10) of an isotope of element 103, Ghiorso, Sikkeland, A. E. Larsh, and R. M. Latimer also reported a new isotope of element 102. This activity, produced by bombarding a mixture of californium isotopes (Cf^{250} , Cf^{251} and Cf^{252}) with boron ions at the HILAC, decayed by the emission of

8.2 MeV alpha particles with a half-life of approximately 15 sec. We assigned this activity to 102^{255} on the basis of the comparative yields of this and other product activities and the variation of its yield with energy of the bombarding boron projectiles. This mass assignment was not definite as the excitation function was very broad, both because it was the product of a (B^{11}, pxn) reaction and because the target was not monoisotopic.

Early Work in the USSR

Work directed toward the production and identification of element 102 commenced in the Soviet Union as early as autumn of 1957. After the publication of the Berkeley element 102 experiments G. N. Flerov at the Second Geneva Conference reported/on some work (11) carried out in his laboratory in Moscow at the Kurchatov Institute and a more complete description (12) of this work was published in 1960. The Soviet scientists bombarded targets containing 100-200 $\mu\text{gm}/\text{cm}^2$ of Pu^{241} with intense currents of O^{16} ions using the internal beam in a 150 cm cyclotron. The product atoms were caught in vacuum in an aluminum catcher foil which was cyclically shuttled a distance of 200 cm in a time of 2 sec and placed next to a nuclear emulsion. Alpha particles of energy 8.8 ± 0.5 MeV were detected and attributed to an isotope of element 102 (with probably mass number 253 or 254) having a half-life in the range of 2-40 secs. There was present in the plutonium target a tiny amount of lead impurity which also produced high-energy alpha activity in the same energy range and, consequently, it was necessary to make large background corrections to the observed results. Figure 1, taken from Ref. 12, shows the gross alpha-particle spectrum obtained from 10 three-hour bombardments. Years

later the source of much of the background was determined (13) to be from a reaction with a large cross section, $\text{Pb}^{208}(\text{O}^{16}, 3\alpha n)$ to produce 25-sec Po^{211m} . Most of the alpha decay of this isomer occurs at 7.14 MeV but 7% of it occurs with an energy of 8.87 MeV and it is this group which accounted for much of the activity that was observed. It should be noted that this 8.87 MeV group also serves as a convenient internal energy standard for a present-day evaluation of this early work. In 1963 the paper of Donets et al (13), though devoted primarily to the isotope 102^{256} , contained a final re-evaluation of the 1957-1958 Russian experiments. It stated that after making all corrections necessary, the energy of the alpha particles observed was found to be 8.9 ± 0.4 MeV and that the total background amounted to about 50% of this activity.

Soviet experimenters, working at the relatively new Dubna Laboratory, started a new series of experiments which culminated with the successful production and identification of a different isotope of element 102 in 1963. They used the internal beam of the new 300 cm cyclotron to bombard U^{238} with Ne^{22} ions, producing 102^{256} by the reaction $\text{U}^{238}(\text{Ne}^{22}, 4n)(13)$. The method was based on the Berkeley double-recoil technique, with the chemical identification in this case of the alpha-emitting daughter 25-hr Fm^{252} . The double-recoil system in this case used a rotating drum and the change in daughter activity on the second (the catcher) foil made it possible to deduce a half-life of about 8 sec for 102^{256} . This was followed by the production of 102^{256} by another reaction, $\text{Pu}^{242}(\text{O}^{18}, 4n)$, with identification by the same method (14).

Recent Dubna Research

In December 1965 work was published by the Soviet groups in which they identified 102^{254} , the same isotope produced in 1958 in the discovery experiment at Berkeley. One research team, Donets et al, (15) continued to use the double-recoil technique, this time separating Fm^{250} from 102^{254} , and came up with the surprising conclusion that the half-life of 102^{254} was not 3 sec but 50 ± 10 sec. They used two methods of production, $\text{U}^{238}(\text{Ne}^{22}, 6n)102^{254}$ and $\text{Am}^{243}(\text{N}^{15}, 4n)102^{254}$, to show quite conclusively that the Berkeley half-life must be greatly in error. Although the statistics of the Am^{243} experiment were poor relative to those obtained with the U^{238} target they were sufficient to show that the 102^{254} half-life must be much greater than 3 sec.

Another group, Zager et al, (16) used the external beam of the 150 cm cyclotron, which had been moved to Dubna, to make measurements of the alpha-particle energy of 102^{254} produced via the $\text{Am}^{243}(\text{N}^{15}, 4n)$ reaction. For this experiment they made use of the "gas jet" technique to transport the recoil atoms of element 102 to a collector. This method, though it originated in our laboratory (17) and had been used for the study of alpha-particle emitters in the rare earth and lead regions, (18) had not yet been adapted for use with the heaviest elements because of the difficulties presented by extremely radioactive targets such as curium. As used by the Dubna experimenters the method consisted of stopping the recoils in an atmosphere of helium gas which was continuously pumped through an orifice into an evacuated space where the heavy atoms could impinge on one arm of a collector shaped like a maltese cross. After an interval of bombardment, the cross was periodically rotated 90 deg to put the collected atoms in front of a

solid state alpha-particle detector while simultaneously another arm was placed to collect a new batch of jet-borne atoms from the target chamber. By the use of a 2-parameter analyzer the decay periods of the activities could be measured by noting the time of arrival of each decay event after each rotation of the cross. With this arrangement these experimenters were able to observe an alpha activity of 8.10 ± 0.05 MeV which was due in part to 102^{254} . Their measurement of 30-40 sec for its half-life was complicated by the detection of alpha particles with the same energy from the alpha decay of Em^{213} . This nuclide appeared as the electron-capture daughter of 34-sec Ra^{213} which was formed by interaction of the N^{15} ions with a minute lead impurity in the Am^{243} target. In order to prove the assignment of the 8.10 MeV alpha particles of 102^{254} , the recoiling Em^{250} daughter nuclei which reached the face of the crystal were analyzed by themselves by shielding the detector from the collector arm. Two detectors were used to allow both types of experiments to be conducted simultaneously.

The next series of experiments, published in August 1966 by Mikheev et al, (19) were devoted to confirming the preceding work on 102^{254} and to measuring the properties of 102^{253} and 102^{252} . This group of researchers, using the same kind of apparatus, bombarded a target containing 5% Pu^{242} with O^{16} ions to form 102^{254} . They found an alpha-particle energy of 8.11 ± 0.03 MeV with a half-life of 75 ± 15 sec and an excitation function to form the activity in good agreement with that expected for a reaction involving the evaporation of four neutrons. At a higher O^{16} bombarding energy they discovered a longer-lived activity which they attributed to 102^{253} . Its characteristics were 8.01 ± 0.03 MeV alpha

particles with a half-life of 95 ± 10 sec. To confirm that this activity was due to 102^{253} they analyzed the recoiling daughter nuclei by the method mentioned previously and were able to marginally identify the 25-min 7.52 MeV alpha particles emitted by Fm^{249} . To supplement this proof they turned to production of the same isotope via a $\text{Pu}^{239}(\text{O}^{18}, 4n)102^{253}$ reaction. In this experiment they found an alpha energy of 8.02 ± 0.03 MeV and a half-life of 95 ± 20 sec with an excitation function of the proper shape and energy range. At a higher bombarding energy the isotope 102^{252} was produced with the Pu^{239} target and O^{18} ions by the evaporation of five neutrons. They reported an energy of 8.41 ± 0.03 MeV for the alpha particles with a half-life of 4.5 ± 1.5 sec, and, as in the other milking experiments, they detected a few events that could be attributed to the daughter isotope Fm^{248} , a 36-sec 7.85 MeV alpha particle emitter. They did not report a spontaneous fission branching decay.

In September 1966 the discovery of the isotope 102^{255} was published by Akap'ev et al (20). This Dubna group detected alpha particles with an energy of 8.08 MeV with a half-life of approximately 2 min by bombarding U^{238} with Ne^{22} ions. From the peak energy of the excitation function they deduced that they had observed the product of a 5 neutron-out reaction and suggested that the 1961 Berkeley work attributing its 8.2 MeV 15-sec alpha activity to 102^{255} must be in error. With a five times smaller yield they also observed a small number of 8.41 MeV alpha particles that were probably due to the isotope 102^{254} made in a $(\text{Ne}^{22}, 4n)$ reaction. It was not possible to measure the half-lives accurately in these experiments.

In October 1966 at the International Conference on Heavy Ion Physics held at Dubna all of the Russian work on element 102 was reviewed by

various speakers. A table was circulated to the delegates which summarized the latest data found by their groups concerning the nuclear properties of the isotopes of element 102 from mass numbers 252 to 256. These figures are included in Table 1 along with recent Berkeley measurements on the same isotopes as described below.

Recent Berkeley Work

Responding to the challenge of the Soviet results, which seemed to be at variance with all of the earlier Berkeley research on element 102, our group set out in November 1966 to repeat the old experiments with a view to clearing up the discrepancies. Using a greatly improved HILAC with more intense beams of heavy ions we were able to produce and identify all of the isotopes of element 102 from mass numbers 251 to 257, inclusively. (21). In addition meaningful limits were set regarding the properties of 102^{258} .

To span the range of these isotopes we bombarded essentially mono-isotopic targets of Cm^{244} , Cm^{246} , and Cm^{248} with concentrated beams of C^{12} and C^{13} ions. Compound nucleus reactions made by these combinations have the distinct advantage of cross sections that in many instances are larger by nearly one order of magnitude to produce the same nuclides when compared with the combinations used by the Dubna groups. This enabled us to make what we believe to be substantially more accurate measurements of alpha half-lives and energies and more definitive observations of spontaneous fission branching.

For most of these experiments a system was used in which a helium gas jet transported the recoiling atoms into vacuum to impinge upon the

periphery of a wheel. The wheel was periodically rotated in a digital fashion to position the atoms in turn in front of each of four silicon (Figs. 2,3). alpha-particle detectors/ By analysis of the decay events with a multi-channel electronic apparatus, it was possible to measure energies and half-lives with good accuracy. Recoiling alpha-daughter activities were identified wherever possible. In some cases mica detectors located just outside the circumference of the wheel were used for unambiguous spontaneous fission analysis (22). It was usually possible to make the same isotope by more than one reaction and thus clearly define its mass number.

One of the first observations to be made in this work was that the half-life of 102^{254} was indeed not 3 sec as claimed in our papers of 1958 and 1959. A value of 55 ± 5 sec was determined in good agreement with the 1965 work of Donets et al (15). However, the cross section for formation of 102^{254} via a $\text{Cm}^{246}(\text{C}^{12}, 4n)$ reaction was found to be about a microbarn which is consistent with the value found in our 1958 experiments in which we milked Fm^{250} from 102^{254} and identified it chemically. Clearly there was something wrong with the milking experiments which implied a 3-sec half-life by the physical identification of " Fm^{250} ."

The next step in attempting to unravel the puzzle was the identification of 102^{252} as having a 2.3 ± 0.3 sec half-life and an alpha energy of 8.41 ± 0.02 MeV (the Dubna values are 4.5 ± 1.5 sec and 8.41 ± 0.03 MeV). In addition to the alpha activity we found a spontaneous fission branching to the extent of about 30% in close agreement with the 1959 work in which we used a grid chamber for analysis. The alpha-energy measurement of

8.3 MeV made at that time can now be corrected to 8.4 MeV if we use as an internal standard the latest value for the energy of Fm^{248} , and the old 3-sec half-life measurement is well within the standard deviations of the two experiments. We are still not certain that the spontaneous fissions observed are actually due to 102^{252} . The fact remains, however, that we have not been able to change appreciably the observed alpha-to-fission ratio by excitation function or recoil range variations, and the spontaneous fissions are not observed when B^{11} ions are used to bombard the Cm^{244} target. Whatever their origin they certainly are produced simultaneously with the 102^{252} alpha emitter and accordingly they serve as an excellent tracer to compare the early experiments with the new.

As a consequence of this work on 102^{254} and 102^{252} we believe the following statements can be made about our earliest experiments without qualification: (1) Fm^{250} was milked from 102^{254} in 1958 thus giving for the first time positive proof for the presence of element 102 and, (2) the isotope 102^{252} was discovered for the first time in 1959 although extenuating circumstances caused us to mislabel it as 102^{254} . It is impossible to be certain as to how the physical milking experiments went awry and gave a 3-sec half-life for 102^{254} in 1958. We can only offer the following hypothesis as being entirely reasonable and consistent with all of the facts as they are known today.

Our curium target contained 95% Cm^{244} so that by the reaction $\text{Cm}^{244}(\text{C}^{12}, 4n)$ the isotope 102^{252} would have been produced with a yield about three times greater than that for 102^{254} formed via the $\text{Cm}^{246}(\text{C}^{12}, 4n)$ reaction. The nuclide 102^{252} would have led to the alpha emitting granddaughter 25-min 7.22 MeV Cf^{244} through the decay chain

$102^{252} \xrightarrow[\alpha]{2.3s} \text{Fm}^{248} \xrightarrow[\alpha]{36s} \text{Cf}^{244}$. The Cf^{244} produced in this way could have been confused with the alpha emitting 30-min 7.43 MeV Fm^{250} when one considers the relatively poor energy resolution and drift problems that had to be contended with in the multi-chamber system used to analyze the small amounts of activity available at that time. Fm^{248} would also have been produced directly by a (C^{12} , α 4 n) reaction causing a 36-sec component in addition to the 2.3-sec one due to 102^{252} . However, most of the experiments for the half-life measurement were performed at the 70 MeV peak for the (C^{12} , 4n) reaction, where we know now that the cross sections for the production of 102^{252} and Fm^{248} are comparable. Since the belt speed was about 3 sec per catcher, it would have discriminated against the 36-sec component by a factor of ~ 15 in the first catcher and so the apparent half-life would have been minimally affected. In the chemical milking experiments there was no attempt to make a half-life measurement of the element 102 parent, since in each case all of the catchers were used for the chemical separation; there was also no resolution problem, since any Cf^{244} would have been separated away by the cation exchange column. For these reasons we think it quite likely that our half-life results were due mostly to the isotope 102^{252} . The amount formed is consistent with its formation cross section as known today.

In our recent work we have found an activity which we attribute to 102^{257} . This has two prominent alpha groups of 8.23 and 8.27 MeV and a half-life of 23 ± 2 seconds. This corresponds reasonably well with the 15-sec 8.2 MeV alpha activity observed in 1961 in the lawrencium discovery experiments and attributed to 102^{255} . This isotope thus is the third one to be found at Berkeley before the 1963 experiments of the

Dubna groups in which for the first time in their laboratory they positively identified an isotope of element 102 by milking Fm^{252} from 102^{256} .

We have essentially confirmed the Soviet findings on the isotope, 102^{256} , except that our half-life of 3.2 ± 0.2 sec is much shorter than their latest values, 6 ± 2 and 9 ± 3 sec. We have carefully repeated this experiment several times and feel that there must be some systematic error in the Dubna work, since they have also reported an 8-sec value for its observed spontaneous-fission decay rate (23). On the other hand, we find very good agreement for the characteristics of 102^{253} and 102^{255} which were also first discovered by the Russian experimenters.

We have been able to add 102^{251} to the list of isotopes of element 102. We find that it decays by the emission of a prominent group of alpha particles of 8.60 MeV (80%) and possibly a higher energy group at 8.68 MeV (20%) with a half-life of 0.8 ± 0.3 sec. We have carefully searched for the isotope 102^{258} without reward. With a predicted alpha half-life of about a minute (see Fig. 5) and a predicted cross section of 10^{-31} cm² for its formation via a $\text{Cm}^{248}(\text{C}^{13}, 3n)$ reaction, we should have been able to observe its presence. Not finding alpha particles or spontaneous fissions in the seconds to minutes interval, we have extended the search to look for spontaneous fissions in the half-life region much less than a second but so far we have not been successful. The properties of 102^{258} are very important to determine, since predictions of the nuclear properties of heavier nuclides are very much in doubt at the present time.

Conclusions

Table 1 summarizes all the data from both groups for detailed

comparison. It should be noted again that both the Russian and the American efforts now completely rule out the possibility of any isotope of 102 having alpha radiation in the vicinity of 8.5 MeV with a 10-min half-life. We suggest that the most likely explanation for the production of such an activity in the early Nobel Institute experiments is that it was due to 8-min Th^{225} made by a multi-nucleon transfer reaction known to occur with a measureable cross section. Th^{225} decays through an alpha decay chain to 8.34 MeV alpha particle emitting Po^{213} which might easily have been confused in energy at that time.

The earliest experiments which might have identified an isotope of element 102 were those performed at the Kurchatov Institute in 1957. Unfortunately this work was marred by backgrounds so that, though 8.4 MeV 102^{252} alpha particles may have been present in the nuclear emulsions used at that time, it appears that their detection would have been obscured by range straggling of the 8.87 MeV alpha radiation from Po^{211m} and by the 8.43 MeV alpha radiation from Fr^{214} . We believe that the Russian assignment at that time of 8.9 ± 0.4 MeV alpha radiation with half-life from 2-40 sec to an isotope of element 102 is too indefinite to support a claim to discovery of this element.

Table 2 recapitulates in a simple manner the early history of the key isotopes 102^{252} , 102^{254} , and 102^{257} . We feel that the conclusion is clear that the original discovery of an isotope of element 102 at Berkeley in 1958 with supporting work in 1959 has been confirmed by subsequent work.

A number of additional isotopes, all rather short-lived, have been produced and identified, thus contributing substantially to the knowledge

of the nuclear properties of this element. In particular the dramatic effects of the subshell at 152 neutrons (24) have been further demonstrated. To show striking similarities with the other heavy elements we have plotted the new data for alpha energy, alpha half-life and spontaneous fission half-life vs neutron number for the even Z elements (Fig. 4,5,6). The stabilizing effects of the 152 neutron subshell are very apparent for element 102 and this observation strongly suggests that the rate of decrease in spontaneous fission half-life with increasing atomic number is larger than expected, since this effect has been greatly modified by the neutron shell in the lower Z elements. We have extrapolated these curves to element 104 and find that the nuclear half-lives predicted for isotopes of this element must be revised radically downward from former prognostications.

A Name for Element 102

It has been often suggested to us that, in view of our right according to tradition, we should suggest a name for element 102 to replace the hastily given one, nobelium. We and our collaborators have given this matter a great deal of thought. In view of the passage of time and the degree of use of this honorable name in numerous text books and scientific writings throughout the world, we wish to suggest that nobelium be retained as the name for this element along with its symbol, No.

References and Notes

1. A. Ghiorso, B. G. Harvey, G. R. Choppin, S. G. Thompson, and G. T. Seaborg, Phys. Rev. 98, 1518 (1955).
2. G. R. Choppin, B. G. Harvey, and S. G. Thompson, J. Inorg. Nucl. Chem. 2, 66 (1956).
3. P. R. Fields, A. M. Friedman, J. Milsted, H. Atterling, W. Forsling, L. W. Holm, and B. Åström, Phys. Rev. 107, 1460 (1957).
4. P. R. Fields, A. M. Friedman, J. Milsted, H. Atterling, W. Forsling, L. W. Holm, and B. Åström, Arkiv. Fysik 15, 225 (1959).
5. A. Ghiorso, T. Sikkeland, J. R. Walton, and G. T. Seaborg, Phys. Rev. Letters 1, 17 (1958).
6. A. Ghiorso, T. Sikkeland, J. R. Walton, and G. T. Seaborg, Phys. Rev. Letters 1, 18 (1958).
7. A. Ghiorso, paper delivered at Eighth Mendeleev Conf. on Chem., Moscow, Mar. 1959, Lawrence Radiation Laboratory Report UCRL-8714, Mar. 1959 (unpublished).
8. A. Ghiorso, Atomnaya Energiya 7, 338 (1959).
9. A. Ghiorso, Proc. Second Conf. on Reactions Between Complex Nuclei, Gatlinburg, Tennessee, May 1960 (Wiley & Sons, New York, 1960), p. 195.
10. A. Ghiorso, T. Sikkeland, A. E. Larsh, and R. M. Latimer, Phys. Rev. Letters 6, 473 (1961).
11. G. N. Flerov, Proc. Second International Conf. on Peaceful Uses of At. Energy, Geneva, 1958 (United Nations, New York, 1958) 14, 151.
12. G. N. Flerov, S. M. Polikanov, A. S. Karamyan, A. S. Pasyuk, D. M. Parfanovich, N. I. Tarantin, V. A. Karnaukov, V. A. Druin, V. V. Volkov, A. M. Semchinova, Yu. Ts. Oganesyanyan, V. I. Khalisev,

- G. I. Khlebnikov, B. F. Myasoedov, and K. A. Gavrilov, Zh. Eksperim. i Teor. Fiz. (English transl.: Soviet Phys.--JETP) 38, 82 (1960).
See also an earlier version of this paper by the same authors in Doklady 3, 546 (1958).
13. E. D. Donets, V. A. Shchegolev, and V. A. Ermakov, Atomnaya Energiya 16, 195 (1964).
 14. V. A. Druin, N. K. Skobelev, B. V. Fefilov, and G. N. Flerov, Dubna Preprint JINR-P 1580 (1964).
 15. E. D. Donets, V. A. Shchegolev, and V. A. Ermakov, Atomnaya Energiya 20, 223 (1966).
 16. B. A. Zager, M. B. Miller, V. L. Mikheev, S. M. Polikanov, A. M. Sukhov, G. N. Flerov, and L. P. Chelnokov, Atomnaya Energiya 20, 230 (1966).
 17. A. Ghiorso, unpublished work, 1959.
 18. R. D. Griffioen and R. D. Macfarlane, Phys. Rev. 133, 1373 (1964).
 19. V. L. Mikheev, V. I. Ilyushchenko, M. B. Miller, S. M. Polikanov, G. N. Flerov, and Yu. P. Kharitonov, Dubna Preprint JINR-P 2839 (1966) submitted to Atomnaya Energiya.
 20. G. N. Akap'ev, A. G. Demin, V. A. Druin, Yu. V. Lobanov, B. V. Fefilov, G. N. Flerov, and L. P. Chelnokov, Dubna Preprint JINR-P 2938 (1966) submitted to Atomnaya Energiya.
 21. A. Ghiorso, T. Sikkeland, and M. J. Nurmi, Phys. Rev. Letters 18, 11 (1967), p. 401.
 22. P. B. Price and R. M. Walker, Phys. Rev. Letters 3, 113 (1962).
 23. V. I. Kuznetsov, Yu. V. Lobanov, and V. P. Pereygin, Dubna Preprint JINR-P 2525 (1965).
 24. A. Ghiorso, S. G. Thompson, G. H. Higgins, B. G. Harvey, and G. T. Seaborg, Phys. Rev. 95, 293 (1954).
 25. The work discussed in this article was done under the auspices of the U.S. Atomic Energy Commission.

Table 1. Production schemes and decay properties of various isotopes of element 102.^a

UCRL-17427
Ghiorso 22

Isotope	Reaction used	Peak cross section ^c (10 ⁻³⁰ cm ²)	Half-life (sec)	α E (MeV) (±.02)	SF/α ratio	Reaction used	Half-life (sec)	α E (MeV) (±.03)	SF/α ratio
102 ²⁵¹	Cm ²⁴⁴ (C ¹² ,5n)	0.09	0.8±0.3	8.60(80%) 8.68(20%)		Not reported			
102 ²⁵²	Cm ²⁴⁴ (C ¹² ,4n)	0.13	2.3±0.3			8.41	1/2 ^d	Pu ²³⁹ (O ¹⁸ ,5n)	4.5±1.5
	Cm ²⁴⁴ (C ¹³ ,5n)	0.096	~2.5 ^b	8.41	1/2 ^d				
102 ²⁵³	Cm ²⁴⁴ (C ¹³ ,4n)	0.29	105±20	8.01		Pu ²⁴² (O ¹⁶ ,5n)	95±10	8.01	
	Cm ²⁴⁶ (C ¹² ,5n)	0.25	~100 ^b	8.01		Pu ²³⁹ (O ¹⁸ ,4n)			
102 ²⁵⁴	Cm ²⁴⁶ (C ¹² ,4n)	0.89	55±5	8.10		Am ²⁴³ (N ¹⁵ ,4n)	—	8.11	≤1/1800
	Cm ²⁴⁶ (C ¹³ ,5n)	0.54	~50 ^b	8.10		U ²³⁸ (Ne ²² ,6n)	50±10		
	Cm ²⁴⁴ (C ¹³ ,3n)	0.096	~50 ^b	8.10		Pu ²⁴² (O ¹⁶ ,4n)	75±15		
102 ²⁵⁵	Cm ²⁴⁶ (C ¹³ ,4n)	0.47	185±20	8.11		U ²³⁸ (Ne ²² ,5n)	~120	8.08	
	Cm ²⁴⁸ (C ¹² ,5n)	0.38	~180 ^b	8.11		Pu ²⁴² (O ¹⁸ ,5n)	180±10	8.09	
102 ²⁵⁶	Cm ²⁴⁸ (C ¹² ,4n)	0.74	2.9±0.5	8.43	~1/200	U ²³⁸ (Ne ²² ,4n)	6±2	8.41	1/200
	Cm ²⁴⁸ (C ¹³ ,5n)	0.75	3.2±0.2	8.43		Pu ²⁴² (O ¹⁸ ,4n)	9±3	8.42	
	Cm ²⁴⁶ (C ¹³ ,3n)	0.09		8.43					
102 ²⁵⁷	Cm ²⁴⁸ (C ¹³ ,4n)	1.1	23±2	8.23(50%) 8.27(50%)		Not reported			
	Cm ²⁴⁸ (C ¹² ,3n)	0.08	~20 ^b			8.25			

^aData on the left are from the recent Berkeley work; those on the right are from a summary by the Dubna groups published at the Joint Conf. on Heavy Ion Physics, Dubna, USSR, Oct. 1966.

^bNo error given due to rather poor statistics.

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

^dThe mass assignment of the SF emitter is not yet conclusive.

TABLE 2. Recapitulation of the early
Berkeley work and its subsequent confirmation.

1958-59 Discovery Experiments
and 1961 Experiment

1966 Confirmatory Experiments

1958-59 Discovery Experiments and 1961 Experiment			1966 Confirmatory Experiments				
Description of Experiment	Half- Life	Radiation (Energy, MeV)	Old Isotopic Assignment	Description of Experiment	Half- Life	Radiation (Energy, MeV)	Present Isotopic Assignmen
1. 1958. Double recoil, chem. identification of daughter Fm^{250} (~0.5 hr., ~7.4 MeV α).	Not deter- mined	Not deter- mined	102^{254}	Direct Measurement of Radiation	55 sec.	α , 8.11	102^{254}
2. 1958. Double recoil, moving belt, observa- tion of α daughter (~0.5 hr., ~7.4 MeV, actually Cf^{244}).	~3 sec.	Not deter- mined	Assumed 102^{254}	Direct Measurement of Radiation	2.3 sec.	α , 8.41 S.F. (30%)	102^{252}
3. 1959. Direct measure- ment of radiation.	~3 sec.	α , 8.3 (Corrected to 8.4) S.F. (30%)	Assumed 102^{254}	Direct Measurement of Radiation	2.3 sec.	α , 8.41 S.F. (30%)	102^{252}
4. 1961. Direct measure- ment of radiation.	~15 sec.	α , 8.2	Assumed 102^{255}	Direct Measurement of Radiation	23 sec.	α , 8.23, 8.27	102^{257}

Figure Legends

Fig. 1. Spectrum of alpha particles in emulsion with energies above 7.5 MeV obtained by the Dubna group from April to September 1958 in the bombardment of Pu^{241} with O^{16} . Alpha particles with ranges 45-51 μ have a mean range of 8.8 MeV. After correcting for energy loss in the catcher, the alpha-particle group of interest has the energy 8.9 ± 0.4 MeV. It was concluded that perhaps half of the events could have been caused by an isotope of element 102. The half-life was somewhere between 2 and 40 sec. The series of experiments, from which this spectrum was taken, represent an improved version of the initial ones on which the Dubna group based a claim to have observed an isotope of 102 in the autumn of 1957.

Fig. 2. A plan view of the gas jet system used in recent Berkeley experiments on element 102. The beam, entering from the right through a thin Be window, passes through a Be energy degrading assembly to the target chamber in the center and thence out to the left to a beam-monitoring faraday cup. The target chamber operates at approximately an atmosphere of helium and the whole box is continuously pumped to about 0.1 torr. The four surface-barrier Au-Si alpha detectors are shown surrounding the digitally-pulsed wheel.

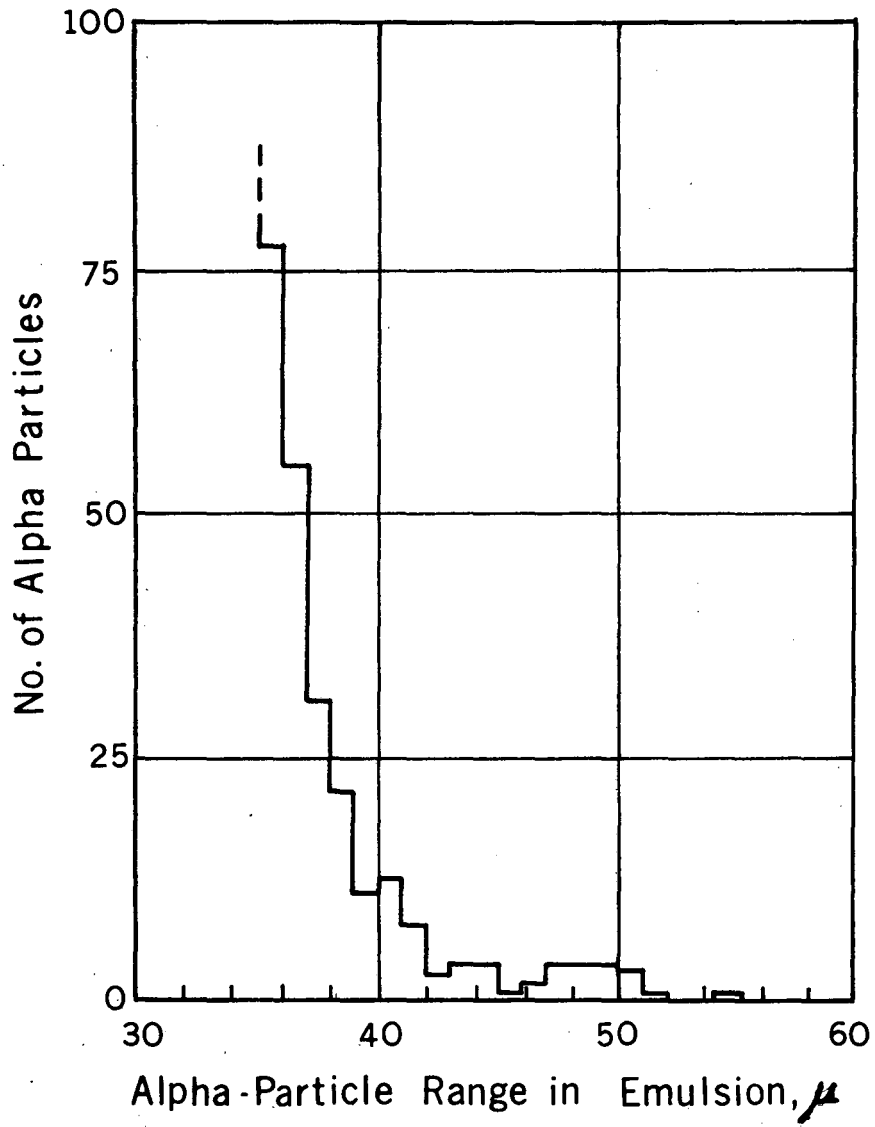
Fig. 3. A close view of the Berkeley gas jet system. The atoms of element 102 are carried by a stream of helium gas to the

periphery of the wheel for analysis by the alpha-particle detectors.

Fig. 4. Alpha-particle energy versus neutron number for trans-amerium elements of even atomic number. Note the very nearly even spacing in energy between nuclides with same neutron number on which the extrapolation to element 104 is based. The effect of the 152 neutron subshell is maintained through element 102.

Fig. 5. Partial alpha half-life versus neutron number for even Z nuclides in the heavy element region. Points representing even-even isotopes are joined by straight lines. The position of the 152 neutron subshell is marked by a vertical line. The extrapolation to element 104 is based on the assumption of a continuation of the systematic variation of the half-life with Z and N.

Fig. 6. Partial spontaneous fission half-life versus neutron number for even-even nuclides. The extrapolation to unknown nuclides is here much more uncertain than it is in the case of alpha half-lives and alpha energies.



XBL673 2027

Fig. 1

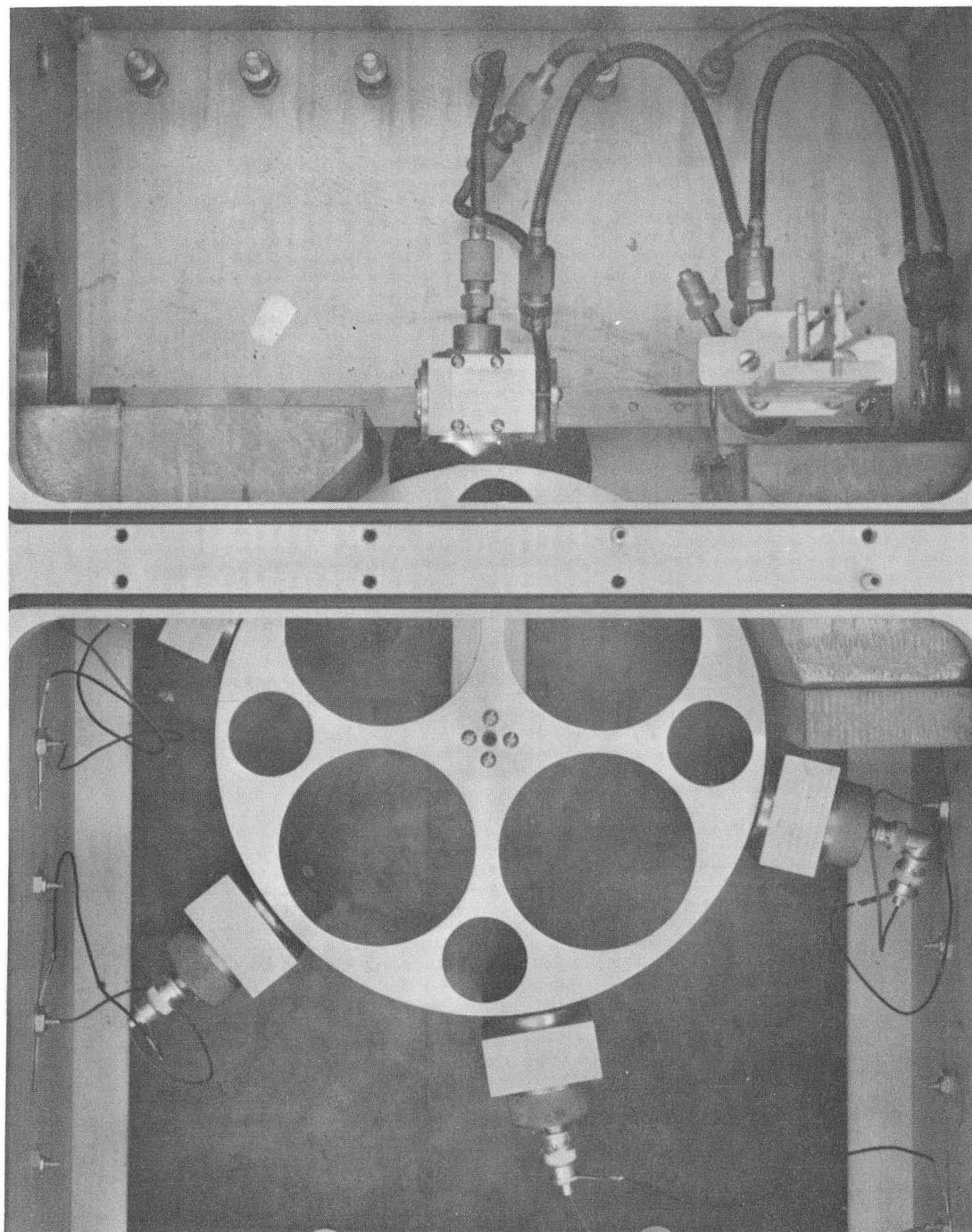


Fig. 2

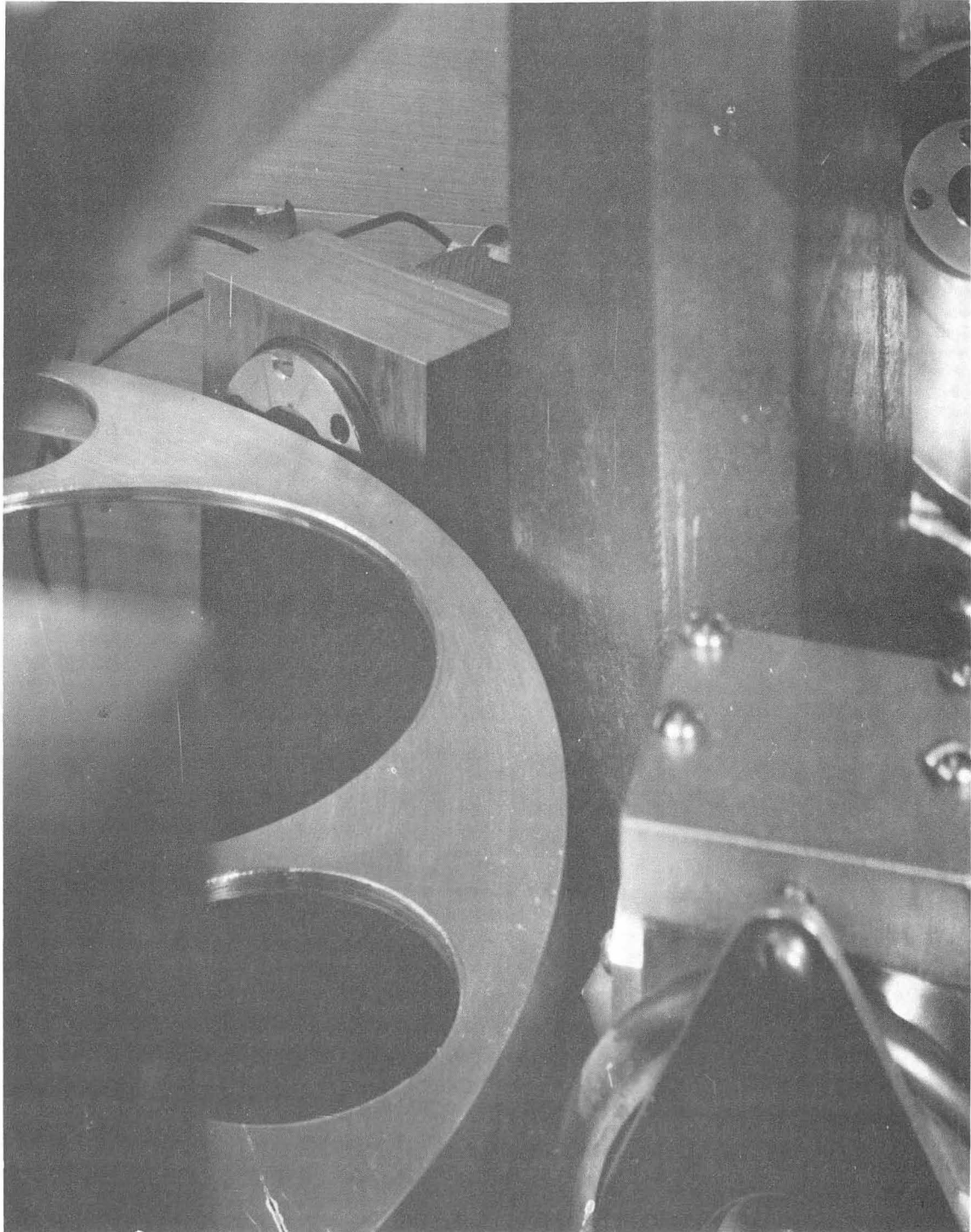
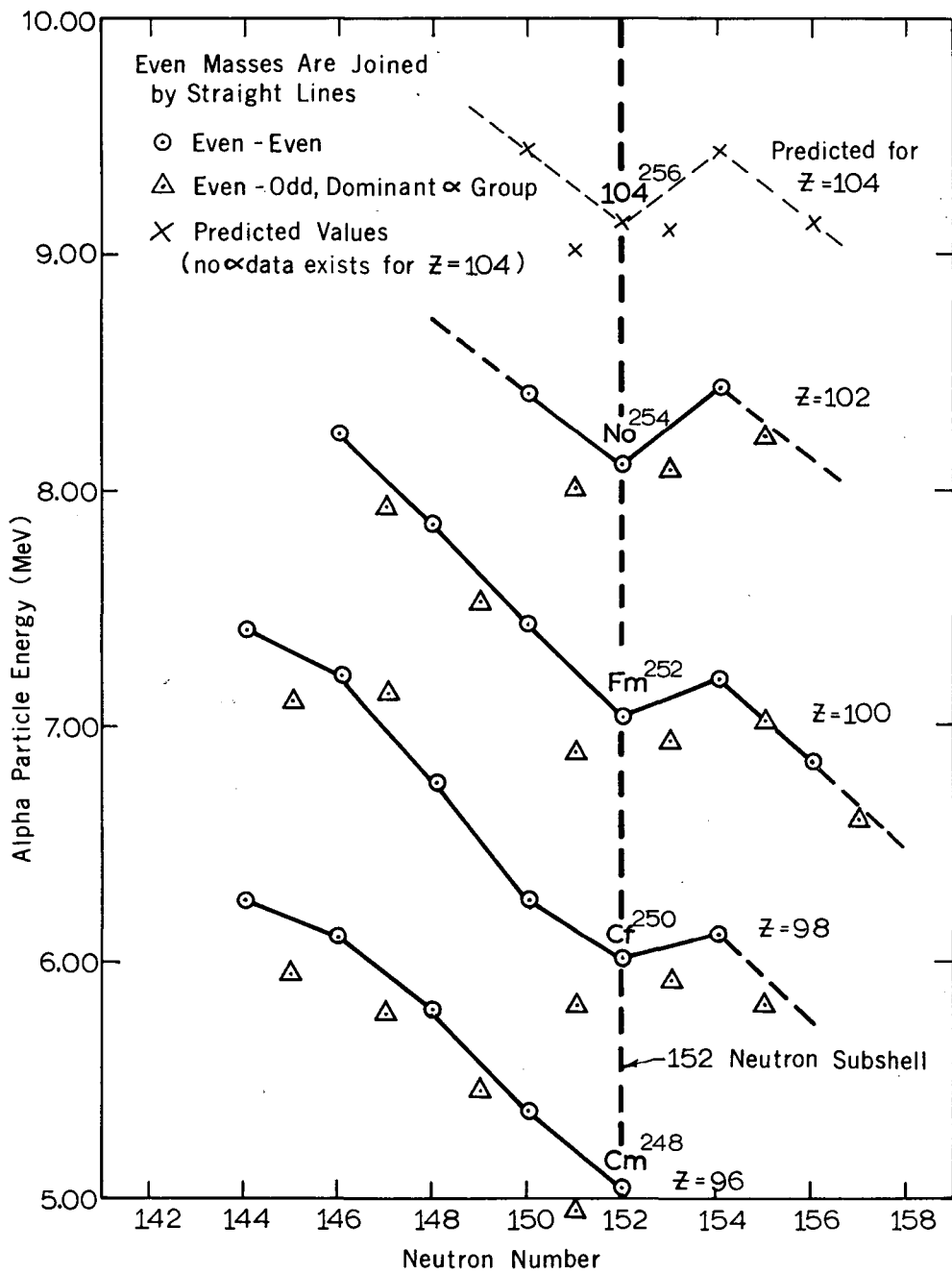
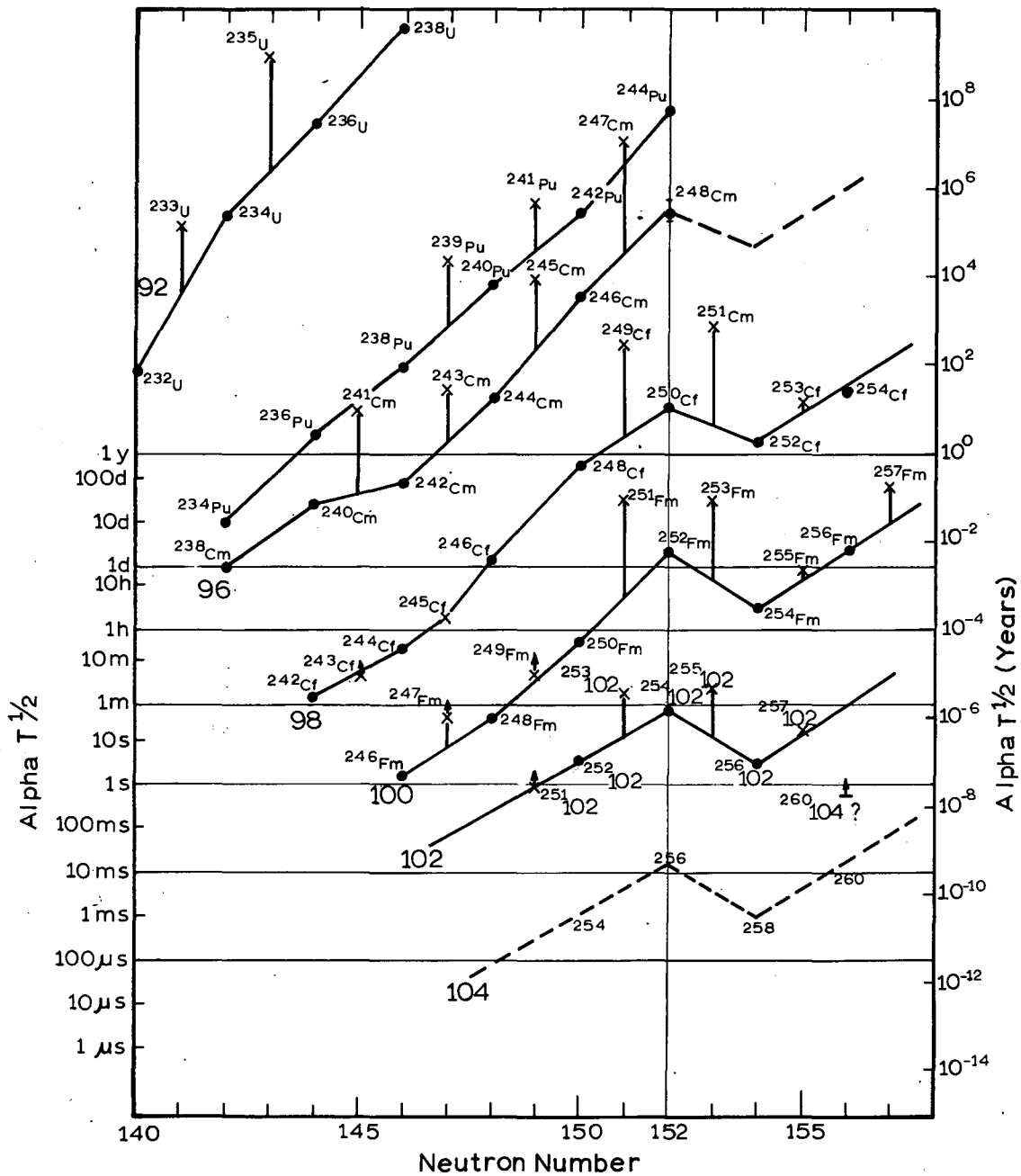


Fig. 3



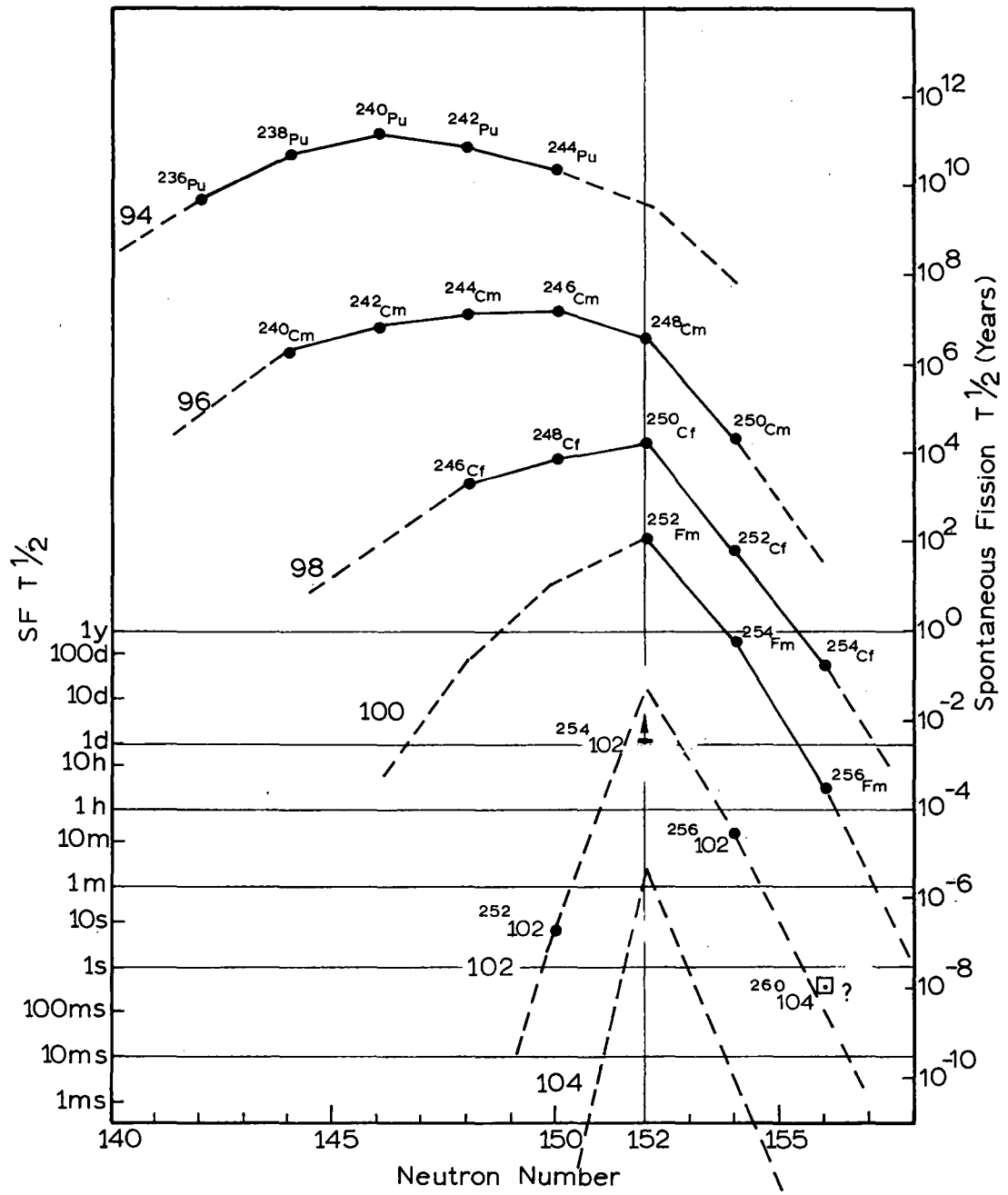
XBL6732028

Fig. 4



XBL672-2015.

Fig. 5



XBL672-2016

Fig. 6

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.

