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Publication Date
1959-03-01

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OF THE TRANSPLUTONIUM ELEMENTS

Albert Ghiorso

Talk given at Mendeleev Chemistry Conference
Moscow, March 1959

TECHNIQUES USED FOR THE PRODUCTION AND IDENTIFICATION
OF THE TRANSPLUTONIUM ELEMENTS

by

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The last fifteen years have seen the discovery of ten transuranic elements. The techniques used in this body of research have undergone great change: and, in general, have reflected and affected other scientific work. In this paper I will deal only with the transplutonium elements, i.e., those elements with atomic number greater than 94. I would like to concern myself mostly with a detailed discussion of the techniques that have been devised for producing and identifying elements 101 and 102 and the possible applications of these methods to elements beyond. However, I think it would be of some interest to review briefly the discovery of all of the transplutonium elements to show the gradual change in experimental methods from the early days when chemistry was much the most important tool to the present day when it is only barely possible to perform any chemistry at all.

It will be seen that usually the production of a new heavy element has depended upon the more massive production of elements of smaller atomic number; thus the discovery of new elements has proceeded in jumps and the process of building up the sequence of man-made elements has consequently been like a military campaign. As each objective was obtained, there had to be a pause to bring up reinforcements in the way of new knowledge and improvement in methods before pushing on toward the next goal. The information gained by new and radically different techniques has been the key to the discovery of elements 101 and 102 and possibly even more radical methods will be needed to produce and identify elements beyond 102.

After the discovery of element 94, plutonium, by Seaborg et al. in 1940[1] there was a period of a few years during which there were no further attempts to produce heavier elements. This time, of course, was spent in developing the plutonium technology and in performing the necessary basic

research in the development of actinide chemistry. However, some time was also spent in learning how to bombard the new element plutonium. This involved entirely new hazards that had not been encountered before, and deadly amounts of plutonium had to be handled in these bombardments. In these early years I must confess that probably adequate precautions were not always taken in the handling of this material.

Element 96, curium, was the first to be discovered. Many fruitless attempts had been made before Seaborg et al. in 1944[2] finally produced the 160 day isotope, Cm^{242}, by helium ion bombardment of Pu^{239} in the famous Crocker Laboratory 60-inch cyclotron. Careful chemistry was needed to distinguish this new element. Much confidence was gained from this discovery and with the new element many tracer chemistry experiments were performed. Thus it was only a short time afterward in 1945 that Seaborg et al.[2a] discovered element 95, americium. The 500 year isotope, Am^{241}, was found in plutonium which had been irradiated in a neutron reactor. The americium was shown to come from a beta-particle emitting isotope, Pu^{241}. The chemical properties of americium and curium are so much alike that it was not until quite a few years later that methods for the complete separation of these elements were worked out. The key to the discovery of these two elements was Seaborg's thesis that these two elements are part of a family of elements with chemical properties analogous to those of the rare earth elements. He called this family the actinide elements.

The instrumental methods used in the discovery of americium and curium were quite crude. At that time ion chamber pulse analysis techniques had not yet been well developed, and it was necessary to use gross counting methods to detect the alpha particle activities. For energy analysis we had to use mica absorption curves which required a relatively large amount of activity to make a good determination. The methods used, though slow and inaccurate, were adequate for the time.

A long period then elapsed for the development of new techniques and for the build-up of a sufficient amount of Am^{241} (a few milligrams) for bombardment with helium ions in the Berkeley cyclotron. Element 97, berkelium, was made late in 1949 by Thompson et al.[3] by means of an $(\alpha, 2n)$ reaction. The 4.3 hour electron capturing isotope, Bk^{243}, was produced in a sufficient quantity to

identify it as a new element. At that time scintillation counters had not yet
been developed and consequently it was very difficult to measure the character-
istic x-radiation. It was the alpha branching decay, consequently, that we
actually detected. Since the alpha branching is only one part in 1,000, it
meant that the amount of alpha activity was very weak. By this time the
development of ionization chambers had proceeded to a point where it was pos-
sible to detect with certainty small amounts of a given alpha energy. The
ion exchange method of chemical separation which had been applied to the rare
earth elements was used to great advantage for the chemical isolation and
identification of element 97. This technique made it possible to separate
the new element quickly from the Am^{241} target material. Elaborate precautions
had to be taken because of the high level of alpha radioactivity in the target.
Bolstered by the experience gained by this discovery, Thompson et al.,[4] a few
months later in 1950, produced element 98 by the bombardment of a few micro-
grams of Cm^{242}. The 45 minute isotope Cf^{245} was identified by its distinctive
alpha radioactivity. Only a tiny amount was made (approximately 5,000
atoms), but the chemical and electronic discrimination against unwanted
curium made it possible to be absolutely certain of the chemical nature of
the new element. It was shown to elute in its predicted position from a
cation exchange column.

With the attainment of element 98 it was thought by us that it would
be many years before elements 99 or beyond could be produced. The next two
years were spent in producing other isotopes of the heavy elements and in
general increasing our knowledge of the nuclear characteristics of these ele-
ments. Plutonium, however, was also put in a high flux reactor so that ulti-
mately we hoped to produce elements beyond 98. The discovery of elements 99
and 100, however, represents an outstanding example of the unexpected in
science. These two elements were discovered in debris from the test thermo-
nuclear explosion of November, 1952 (Operation Mike). Material was collected
on filter papers by drone airplanes flying through the explosion clouds and
this was brought to a number of United States laboratories. Chemical inves-
tigations at the Argonne National Laboratory and the Los Alamos Scientific
Laboratory showed that the material contained some new heavy isotopes of

plutonium. This suggested that new elements might have been built up from uranium by many successive captures of neutrons in the explosion and after hearing of this we undertook to look for elements beyond 98 in the material. Ion exchange experiments immediately brought a new element to light! To identify it, more material was called for. Many hundreds of pounds of coral were collected from an atoll near the explosion area and worked over, and this led to the positive identification of elements 99 and 100 in the form of 99^{253} and 100^{255}. All of the work was done in 1953 but it was not until 1955 that the work was published by Ghiorso et al.[5] Element 99 was given the name einsteinium, symbol E, in honor of Albert Einstein. Element 100 was named fermium, symbol Fm, in honor of Enrico Fermi. It is interesting to note that element 100 was discovered with only about 200 atoms. No basically new techniques were used in these experiments. The discoveries were made possible principally by careful work based on long experience.

Early in 1954, the plutonium which had by then been in the high flux reactor for several years was removed and carefully processed in a search for heavy elements. Some of the same isotopes of elements 97, 98, 99, and 100 that had been found in the debris from the thermonuclear explosion in 1952 were found. Since the quantity of heavy elements made in the reactor was potentially greater than that which was available from the explosion it meant that ultimately we might be able to bombard the 20 day E^{253} isotope with helium ions in the cyclotron to produce element 101. At that time the only heavy ion beams available to us were produced in the Berkeley 60-inch cyclotron with relatively small currents and with a continuous energy distribution. It had been found that the yield of reactions in which only neutrons were emitted was very, very small. Thus it was not feasible at that time to bombard plutonium with nitrogen ions to produce element 101.

By 1955 we had produced enough E^{253} to make possible an attempt at manufacturing another new element. With 10^9 atoms of the isotope E^{253} available we calculated that if we could obtain a helium ion beam of about 200 microamperes per square centimeter we would make something like 1 atom per experiment if the cross section for production of a predicted 101^{255} were about 1 millibarn. This was based on the assumption that the half life of

the 101 isotope would be a few minutes. It was necessary for us to work very fast to separate the very tiny amount of expected element 101 from the target. Conventional methods would thus not have sufficed. Even if it were possible to do a chemical separation fast enough it would have been necessary to make a new target every time a bombardment was to be made. Looking for an isotope with half life of only a few minutes would have required many bombardments and the very small amount of the target material available would quickly have been used up by attrition. Fortunately, a new method was devised and was tried for the first time.

The nuclear recoil produced by the transmutation was used to advantage. It was postulated that if the target were to be made thin enough by careful purification, the new atoms would be ejected from the target and could be collected in a vacuum in a separate catcher foil. It was found by test bombardments with Cm^{244} to produce the isotope Cf^{246} that high yields could indeed be obtained if the target was carefully prepared by electroplating. Since the range of the recoil atoms is very small (approximately 50 micrograms per cm^2) the yield can easily vary from one target to another; for this reason a small amount of Cm^{244} was incorporated with the target E^{253} so that there would be an internal monitor to show that the yields were proper. The requirement that we had to have a very high helium ion intensity per square centimeter of target was fulfilled by modifying the 60-inch cyclotron at Berkeley so that the internal beam was even more highly focused than usual. A suitable probe was then designed and constructed so that the catcher material could be removed very quickly. After many trials, the apparatus was found to work successfully and experiments were begun.

The initial experiments were confined to looking for short-lived alpha radioactivity. Many tries yielded nothing and we were almost discouraged to the point of discontinuing the experiments. During one of the experiments, however, a single event was noticed. One single large pulse was observed which was thought to be caused by a spontaneous fission! When this occurred some time later in another experiment enough confidence in the instrumentation had been accumulated so that it was possible to hypothesize that we must be producing a new isotope which underwent spontaneous fission at a very much higher

rate than had ever been seen before. The isotope that seemed most likely to
have this high spontaneous fission rate was fermium-256. This isotope could
be produced directly via a (He,p) reaction as well as from the electron-capture
decay of 101^{256}. Since we expected that the half life of 101^{256} would be
longer than that of 101^{255}, it meant that by increasing the bombardment time
we could increase the yield of the reaction actually observed in the laboratory
and so overcome the time consumed by the chemical operations. A chemical sepa-
ration was needed because there was enough beta-particle activity induced in
the gold catcher foil by passage of the beam through it to overwhelm the ion
chamber detecting apparatus. The catcher foil was usually dissolved and a
gross actinide element separation was made with an anion exchange column. This
usually took on the order of 5 to 10 minutes.

New bombardments were then started and this time they were increased
in length. We were rewarded almost immediately by finding that we could pro-
duce on the average one spontaneous fission per experiment. The chemical
separation used showed that spontaneous fissions were of actinide origin. To
distinguish between elements 101 and 100 an additional chemical procedure was
performed to separate the individual elements. The same cation resin column
procedure as used before was used, and the initial experiments were confined
to fission counting of the trans-fermium and the fermium fractions. Several
experiments were consistent in their demonstration of spontaneous fissions
occurring in both fractions. To determine more precisely the elution position
of the element responsible for the trans-100 spontaneous fission activity a
more elaborate experiment was then undertaken.

For this purpose we increased the amount of useful data by combining
adjacent experiments. Three successive three hour bombardments were made and
in turn their transmutation products were separated quickly on a cation resin
column. The isotope E^{253} was added in each case so that it together with the
Cf^{246} produced during the bombardment would help to calibrate each column run.
Five spontaneous fission counters were then used to count simultaneously the
corresponding drops of eluant from the three runs. This elution curve showed
that the spontaneous fissions were present in both the 101 and the 100 fractions.
The explanation for this was proved by later milking experiments. It was shown
that we had produced the isotope 101^{256} with an electron-capturing half life of

roughly 1/2 hour decaying to the isotope Fm^{256} which had a spontaneous fission half life of about 3 hours. In a total of eight separate experiments, we isolated in that time only 17 atoms of element 101 but because of the unique nature of the experiment we felt complete confidence in publishing our results. The discovery of element 101 was published by Ghiorso et al in 1955.[6]

We suggested for the name of element 101, mendelevium, with the symbol Mv, in recognition of the pioneering role of the great Russian chemist Dmitri Mendeleev whose name this conference honors. He was, of course, the first to use the periodic system of the elements to predict the chemical properties of undiscovered elements.

The discovery of mendelevium required the ultimate in sensitivity for its detection for it was first identified on the basis of only a couple of atoms and produced by transmutation in a target which itself was so small that it was unweighable. In the case of the other members of this strange family of elements we had worked with minute samples but with nothing as small as this. The production of new elements, however, was quite well understood in our laboratory by this time. It was based largely on our ability to detect with great efficiency and assurance the alpha particle radiations of the heavy elements. Such research depends largely on identification by alpha energy analysis. This is possible with great precision and with low background. Alpha backgrounds, however, are not entirely zero but in this case since we were detecting spontaneous fissions we were able to obtain an essentially zero background. Spontaneous fission was first discovered in U^{238} by Flerov and Petrzhak in 1940[7] but was found to be a phenomenon which was quite rare. As new elements with higher atomic numbers were added to the Periodic Table, the rate of spontaneous fission was found to increase very rapidly with atomic number. For example the isotope Cf^{252} was found which undergoes aponeaneous fission in 3% of its decay. This still meant a half life of 66 years, however. The shortest spontaneous fission half life observed up to this time was Cf^{254} which decayed by spontaneous fission with a half life of 60 days. Fermium-254 was known to have a half life of about half a year. It was thus a great surprise to find that Fm^{256} had a half life as short as 3 hours. This did, however, make it possible to discover element 101 at that time.

This pioneering work in the discovery of element 101 has been corroborated since that time in our laboratory several times by using much larger targets and thus it has been possible to determine with greater accuracy the half lives of the isotopes involved. Phillips et al.[8] have reported that the Mv^{256} decays with a half life of 1.5 hours and that a better half life of Fm^{256} is 2.7 hours. In addition to this they have found an alpha particle activity which is probably due to Mv^{255} and has a half life of 45 minutes.

This brought us up to element 101, and two years were to pass before any further work in the new element domain was to be published. Since element 99 was the heaviest element to be available in quantities that could be bombarded, it meant that the only way one could go higher than atomic number 101 was to use a bombarding ion with more than 2 protons. Thus, one might try bombarding element 96 with carbon ions to make an isotope of 102.

Such an experiment was tried at Stockholm in 1957. In a series of very difficult experiments an international group of scientists from the United States, England, and Sweden bombarded curium with C^{13} ions accelerated in the Stockholm cyclotron and saw a very tiny amount of alpha activity which they attributed to an isotope of element 102. The characteristics of this alpha particle activity seemed to be that it had a half life of about 10 to 15 minutes and seemed to elute in the chemical position which would be proper for that for element 102. The experiment was performed by bombarding a sample of Cm^{244} of about 1 milligram per cm^2 thickness. It was covered with a thin aluminum foil of 50 to 100 micrograms per cm^2 and mounted in vacuum such that the nuclear recoils could go through the aluminum stopping foil and lodge in a thin plastic catcher foil. Many experiments were done and with many targets and though the data were quite erratic they seemed to indicate that there was an activity which had the above mentioned characteristics. The amount of other activities made was miniscule. The published experiment by Fields et al.[9] showing 5 events of the supposed element 102 activity indicated that only about 1/10 of a count per minute of Cf^{246} and a very tiny amount of Fm^{250} were also produced in these bombardments. The number of events observed which were attributed to element 102 were always few in number; consequently it was very difficult to make definitive tests of the activity. The C^{13} ion currents used in these experiments were only .03 to 0.1 microamperes. The energies of the ions was approximately 90 to 100 Mev.

At about this time the heavy ion linear accelerator (HILAC) which had been under construction at Berkeley for about a year and a half was completed and experiments were initiated to try and confirm the results of the Stockholm group. However, the beams that were first available from the HILAC, were too small to be able to duplicate the Swedish experiments and it was not until later in the year that it was possible to check their findings.

It was soon found[10] that we could not confirm their conclusions. We spent many months in 1957 performing different kinds of experiments in a strenuous effort to try to duplicate what had been found in Sweden. The first method used was to bombard several targets simultaneously with catchers behind each target. Since the beam currents available to us were larger than those used at Stockholm, thermal heating by the beam became a problem, and it was soon found that it was not possible to mount the targets in vacuum. Instead, we placed the targets in a helium atmosphere and mounted the catcher foils immediately adjacent to the targets. At first to insure that we lost none of the nuclear recoils in a stopping foil, we used none. Later aluminum foils as thick as 100 micrograms per cm^2 were used to reduce the amount of curium knocked out of the target. In the initial experiments chemical methods were used to separate the small amount of curium that was knocked out of the target by the bombarding beam. Many experiments were performed at various energies with C^{12}, C^{13}, and O^{16} ions. Our attempts to find the elusive 10-minute 8.5-Mev alpha particle activity that had been attributed to element 102 were never successful, even though we produced as many as 100 counts per minute of Cf^{246} and similar amounts of Cf^{245}, and Fm^{250}. These amounts were hundreds of times larger than that produced by the Stockholm experiments. We never saw more 8.5-Mev events than could be accounted for by very tiny amounts of background in our apparatus.

Nonetheless, we continued the experiments in different ways to make sure that we were not being misled. The catcher foils that we used initially were of 0.9 milligrams per cm^2 palladium leaf. Immediately after the bombardments, which were usually about half an hour long, we would dissolve the palladium and separate the actinide fraction from it. Approximately five minutes after the end of the bombardment we were able to alpha pulse analyze

this gross actinide fraction. The possibility was raised that since califor-
nium can be easily vaporized from uranium metal perhaps element 102 was even
more volatile and that since it was being caught in a metallic foil it might
be vaporized by the beam heating of the foil. To test this hypothesis we
tried catching the recoils in Mylar plastic. To prevent the catcher foils
from being burned by the beam it was necessary to pass helium gas through the
apparatus to cool them. It was assumed that the recoils would become oxidized
as soon as they had lodged in the Mylar foil and thus be prevented from vola-
tilizing. The Mylar plastic was burned away by ignition on a platinum plate.
With this method we were able to look immediately with no chemistry, and to
examine the bombardment products by pulse analysis within 1 minute of the end
of bombardment. Again, although we saw large amounts of Cf^{246}, Cf^{245}, and Fm^{250},
we saw no activity that we could ascribe to element 102.

A third method was used which we feel would certainly have detected
a 10-minute 8.5-Mev alpha emitting isotope of element 102. This involved the
use of electrostatic collection of target recoils. This new method was first
used in the identification of a very short-lived isotope of element 102 and
will be described later in this paper. After we had successfully produced and
identified this short-lived isotope of element 102 we felt that surely the
basic method should also be capable of demonstrating the existence of a much
longer-lived isotope. The nuclear recoils of element 102 were collected on
a negatively charged aluminum plate mounted near the targets in the helium
atmosphere. It was possible after a short bombardment to look quickly at the
aluminum plate directly in an alpha pulse analyzer. Although large quantities
of Cf^{245}, Cf^{246}, and Fm^{250} were observed, we did not observe any long-lived
isotope of element 102. The experiments were conducted over a wide range of
energies and with beam currents as high as a quarter of a microampere. A
fourth method has been used more recently in which the target recoils were
collected on a continuous tape and examined almost immediately in an alpha
grid chamber mounted near the target system. This will be discussed in the
last section. Such experiments we feel would easily have demonstrated the
existence of the postulated long-lived isotope of 102.

Our challenge to the validity of the Stockholm experiments on which
their claim to the discovery of element 102 was based, may be summarized as

follows: (a)   We have used curium targets with substantially the same iso-
topic composition and bombarded them with C^{13}, C^{12}, and O^{16} ions over a wide
energy range with monoenergetic beam currents as much as 10 times greater
than those of wider energy spread of the cyclotron experiments.   (b)   We
have used four different methods of handling the transmutation recoils from
the targets.   All of them were successful in that we detected large amounts
of actinide element products but nevertheless failed to produce 10-minute
8.5-Mev activity.   (c)   The experiments were conducted under geometrical con-
ditions which were very similar to the Stockholm experiments.   In most of
the later experiments it was necessary to use a thin aluminum stopping foil
to keep curium activity from leaving the targets.   Thus it is valid to com-
pare the Cf^{246}, Cf^{245}, and Fm^{250} activities with that produced at Stockholm.
We feel that this is a more accurate comparison of the bombardment conditions
than a beam current reading since it represents a total yield from target to
energy analysis.   (d)   That our experiments would have detected an unknown
activity of 102 is made certain by the fact that one of these methods (the
electrostatic collection of target recoils) was used in the identification
of a very short-lived isotope of element 102.

Our failure to confirm the Stockholm results may possibly be ex-
plained by the following circumstances.   The cyclotron experiment was an
extremely difficult one to perform since it had to make use of the internal
beam of the machine.   The amount of activity produced varied very erratically
and was always extremely tiny, and not more than a few events in any one ex-
periment were ever seen;   in most of them none were detected.   Only 12 out
of 50 bombardments were successful so that under these very difficult condi-
tions it was not possible to be sure that the unknown activity could not be
ascribed to a light element impurity or to other artifact.   It should be
noted that the 90 Mev C^{13} energy used most of the time in the cyclotron ex-
periments would produce predominantly those neutron evaporation reactions
where six or more neutrons would be emitted.   This would tend to optimize
the mass number of any isotope of element 102 that could be produced to 253
or less.   It is quite possible that some isotope of element 102 such as 102^{257}
or 102^{259} would have a half life of 10 minutes, but these isotopes can only
be produced by bombardment of heavier curium isotopes even when using lower

energy bombarding ions. It would also be expected that a 10 minute alpha half life for an isotope of element 102 would imply an alpha particle energy for the most abundant group closer to 8.0 than to 8.5 Mev. The only conclusion that we can draw from these observations is that the activity found by these experimenters cannot be assigned to element 102.

After a period of about four months during which we unsuccessfully attempted to find the 10-minute isotope of 102, we felt equipped to look for a short-lived isotope. For this work a radically new method was tried. It had been suggested and found by Ghiorso[11] that under the proper conditions nuclear transmutation recoils could be attracted essentially quantitatively by an electrically charged plate mounted in a gaseous atmosphere. This meant that the long range recoils (ca. 1 cm in helium at atmospheric pressure) could be stopped in the gaseous atmosphere and then collected on the surface of a conducting foil. With the thin sample thus obtained, it was possible to allow the alpha recoil daughter to escape from the foil; thus, in principle, one could perform a continuous milking experiment wherein the atoms of the daughter element 100 were separated from the parent element 102 by taking advantage of the recoil due to the element 102 alpha particle decay.

The target consisted of a mixture of isotopes of curium (about 95% Cm^{244} and 4.5 Cm^{246}) mounted on a very thin nickel foil. The target was approximately 0.5 milligram per square centimeter thick and was covered with 75 micrograms per square centimeter of aluminum to prevent curium knock-over. The curium was bombarded with monoenergetic C^{12} ions at energies from 60 to 100 Mev. The transmuted atoms were absorbed in helium gas, and it was found that with sufficient electric field strength, practically all of these positively charged atoms could be attracted to a moving negatively charged metallic belt placed directly beneath the target. The element 102 atoms were carried on the conveyer belt under a foil which was charged negatively relative to the belt. Approximately half of the atoms undergoing alpha decay caused the daughter atoms to recoil from the surface of the belt to the catcher foil. After each bombardment the catcher foil was cut transversely to the direction of the belt motion into five equal length sections. After a time of bombardment suited to the half life of the daughter atom to be examined, the five foils were then alpha pulse analyzed simultaneously in a multi-plex assembly

consisting of five Frisch grid chambers, amplifiers, a single Wilkinson type "kick sorter" and a printer. With this equipment it was easily possible to make all the desired measurements for identifying the atoms caught on the catcher foils and thus to measure the half life of the parent of the recoiling atoms. The method was first successfully used in bombardments of Pu^{240} with C^{12} ions to identify a new isotope of element 100, Fm^{248}. It was shown to have a half life of 0.6 minutes by analysis of the amount of the 20-minute Cf^{244} transferred to the catcher foils.

After we had demonstrated that the method would succeed, experiments were started which were aimed at finding a short-lived isotope of element 102. The most likely isotope that could be detected by this method was deemed to be 102^{254}. We predicted a half life of seconds for this isotope leading to the known 30-minute 7.43-Mev alpha particle emitter Fm^{250}. A careful series of experiments showed that Fm^{250} could indeed be collected on the catcher foil in accordance with a parent of half life of approximately 3 seconds produced by the reaction, $Cm^{246}(C^{12},4n)102^{254}$. The excitation function for producing Fm^{250} in this manner was found to peak sharply at 70 ± 5 Mev and corresponds to a $(C^{12},4n)$ reaction in accordance with a recently developed method for calculation of (c,xn) reaction cross sections.[12,13] That the atoms collected are ejected by alpha recoil of atoms from the belt is proved by the fact that neither Cf^{246} nor Cf^{245} which are collected in far greater amounts in the belt are found prominently on the catchers. Changing the belt speed was found to change the distribution of the Fm^{250} on the catcher foils in a manner conforming to a 3-second parent. The number of Fm^{250} counts in a single experiment was as great as 40 and corresponds to a maximum cross section of a few microbarns for the reaction with Cm^{246}.

The final chemical identification of the atoms ascribed to Fm^{250} was carried out by dissolving the activity from the catcher foil and separating it from the other actinide elements by elution with ammonium alpha-hydroxy-isobutyrate from a column packed with Dowex-50 cation resin. In one experiment two atoms of Fm^{250} were identified and in another experiment nine events were observed in the element 100 position. Thus we feel that there can be no doubt that we have identified element 102 in these experiments by this direct milking of its daughter element, fermium.

This work was published by Ghiorso et al.[14] in July of 1958. A short time later, the very fine work of Professor Flerov and his group[15,16] in Moscow was published. The method that these experimenters used, though quite different from that used in the Berkeley experiments, should easily have demonstrated the existence of a long-lived isotope of 102 of mass less than 254. They reported that they did not see any long-lived 8.5-Mev alpha activity which could be ascribed to element 102. However, they did see a short-lived activity with a half life in the region of seconds with an alpha energy of 8.8 Mev. These experiments were extremely difficult and were hampered by background produced by small amounts of lead impurity in the targets. The target in their case was Pu^{241} and the bombarding particles were O^{16} ions produced in the Moscow cyclotron. The method though very sensitive was not capable of uniquely determining the atomic number of the new alpha activity.

After our experiments in which we proved the existence of an isotope of element 102, we set about to modify our apparatus so that we could determine the alpha-particle energy of this isotope or any others that might be produced. For these experiments the same basic method was used but with some important variations. Instead of a continuously moving metallic belt, we made use of a conducting tape of .001 inch thick Mylar made conducting on both sides by a very thin film of aluminum. The nuclear recoils were stopped in helium as before and attracted to the tape. This tape was then pulled into an alpha grid chamber mounted near the targets and the decay products analyzed by a 100 channel pulse analyzer apparatus.

This apparatus could be used in two ways: (1) the tape could be allowed to move continuously from the target to the ion chamber or (2) the tape could be quickly moved at the end of a short bombardment cycle into the counter and analyzed while the next sample was being collected. The advantage of the second method is that the grid chamber could be located farther from the targets and thus more shielding used to attenuate the very extensive beta-gamma activity induced by the heavy ion beam. Another advantage of this method was that the half-lives of many isotopes could be measured simultaneously by additional counters connected to the alpha pulse analyzer and a time sequencing device.

After almost eight months of continuous development we now feel
that we have an apparatus which will measure short-lived alpha-emitting
isotopes directly at the target with great certainty and fair resolution.
Though the method is very difficult to use, it is proving to be extremely
valuable. One of the many reasons why the method is very difficult is that
it is somewhat critical as to the purity of the helium gas used in the
target chamber. This, coupled with the fact that we have to use an argon-
methane gas mixture in the grid chamber for alpha-particle pulse analysis
and the fact that the tape moves quickly from the target chamber to the
grid chamber, implies a very difficult mechanical arrangement. However,
after many modifications we now have an apparatus which is quite successful.
With it we have been able to measure 7.8-Mev alpha particles from Fm^{248}, the
30-second alpha emitter found early last year by the continuous milking of
californium-244 in the conveyor belt experiments.

Very recently we have endeavored to bombard curium targets to
look for alpha-emitting isotopes of element 102. We immediately encountered
great difficulties with the fact that very tiny amounts of lead impurity in
the target produced alpha particle energies of 8.3 and 8.8 Mev with half-
lives from 5 seconds to 22 seconds. By carefully purifying new target
material and by careful electroplating of the targets onto clean nickel
foils, we were able to reduce this background to a negligible value. Un-
fortunately, our apparatus has not been working long enough to be able to as
yet demonstrate any conclusive results. We do see what appears to be a 3-
second 8.3-Mev isotope of element 102 and what may also be spontaneous fis-
sions from the same isotope, probably the 3-second 102^{254}. The branching
ratio for spontaneous fission, approximately 30%, is surprisingly high--many
orders of magnitude higher than predicted! The amount of activity produced
in these experiments was not more than 10 to 20 counts per hour. Thus,
under the difficult and complex conditions of counting at the target we can-
not be sure until more extensive experiments are undertaken that the activity
is actually caused by element 102. The same experiments did not show any
large amount of 8.8-Mev alpha activity which we would expect to see if 102^{253}
decayed with this alpha particle energy. One of the most likely reactions
that would have been observed in the Moscow experiments was $Pu^{241}(O^{16},4n)102^{253}$.

In our case the same isotope should have been produced by the reaction
$Cm^{246}(C^{12},5n)102^{253}$. To increase the yield of this isotope we tried bom-
barding the curium targets with C^{13}. In these experiments 102^{253} would
be produced by a C^{13},4n reaction on Cm^{244}. The greater abundance of
Cm^{244} in our targets would enhance this reaction by a factor of as much
as 15.

In these experiments which were undertaken just a few days ago,
we have seen short-lived 8.8 Mev alpha particles which may be due to 102^{253}.
If this is borne out by future experiments then this will be an independent
confirmation of the Moscow work. These data are still preliminary and it
will be some time before we can be certain of our results. I am very opti-
mistic that we will be able to shortly examine many isotopes of element 102
in this fashion.

Other target materials which will soon be used are Cf^{250} and
Cf^{252}. These will produce isotopes of element 102 with possibly greater
abundance since they will make use of carbon ion reactions with much higher
cross sections. They will also have the advantage in that they should
produce much heavier isotopes of element 102 so that before long we hope
to measure more nuclear characteristics of this newest of the elements.
It is even possible that an isotope, such as 102^{259}, can be produced which
will live long enough to allow chemical experimentation.

The application of these new methods to the production and identi-
fication of elements beyond 102 is clear but the task should prove to be a
very difficult one. We feel now that the cross sections to produce isotopes
of the very heavy elements will be extremely small. Since only relatively
small amounts of target material can be used and relatively small quantities
of ion beams are available it will probably be a while before any laboratory
successfully accomplishes this objective. If in addition to these difficulties
there is the added complexity of short spontaneous fission half-lives, which
might be indicated by our very tentative results in finding spontaneous fis-
sion at the very high rate from 102^{254}, then it may be extremely difficult to
prove that one has found a new element. Spontaneous fissions are not charac-
teristic enough from one element to another to differentiate in any way where-
as alpha particle radiations are. However, it is probable that even in this

case there can be produced an odd neutron isotope which will be hindered
enough for spontaneous fission decay so that its alpha particle energy can
be identified; it thus seems quite likely that elements 103 and 104 will
be found sometime in the future.

One of the interesting basic experiments which should be performed
when element 104 has been discovered is to show in some fashion that it is
different from element 103 and the other actinide elements. Element 103 will
mark the end of the actinide series and it will be thus quite important to
show that 104 is a homologue of hafnium. I should like to suggest a possible
method by which this can be done even with half-lives of the order of milli-
seconds. The method I would suggest is essentially a chemical one and uses
the transit time measurement of an ion drifting in an electric field through
a gas. If the ion after being slowed by the gas to essentially thermal
velocities is attracted by a linear electric field then one would expect
that the transit time of an isotope of element 104 would be shorter than that of
an isotope of 103 of approximately the same mass number. This follows from
the fact that one would expect the ionic radius (which should affect the
drift velocity) of element 104 to be about 13% less than that of 103, whereas
the other actinide elements change only slightly from this. Thus, if one can
devise a method of separating the ions with adequate resolution by using this
transit time difference it should be possible to show the essentially chemical
difference between elements 104 and 103. We have thought of several possible
ways of performing this experiment but have not yet found time to test
them.

Another method which I think has a reasonable chance of being suc-
cessful would enable identification of the mass number of any non-gaseous
reaction product. This method would make use of the fact that the nuclear
recoil could be stopped in a gas and attracted electrically to a small hole
by a properly shaped electric field. By differentially pumping through the
small hole it is quite likely that one could carry the atom from the volume
of relatively high pressure in which it is necessary to stop the recoil into
a high vacuum through successively differentially pumped irises. After the
singly charged ion has been conducted into a vacuum, a very high electric
field could then accelerate it through a high potential so that it could be

analyzed by a magnetic field in the conventional manner. The ion could be either caught directly in a counter and measured for its nuclear radioactivity or detected by an electron multiplier and counted as an ion of a given mass number. Both of these methods, of course, present great difficulties, but I feel that the great potentialities of such systems warrent the effort to overcome the obstacles.

There is another method which is of possible use in creating elements of very high atomic number. This might require the international cooperation of nations such as the USA and the USSR. I refer to the bombardment of very large quantities of the heaviest elements by an instantaneous high flux of neutrons produced by a nuclear detonation in a suitable "container". If one were to bombard perhaps 10 grams of Cf^{252} in this way one might hope to produce very heavy isotopes which might decay to regions of new stability and thus produce super heavy elements. Such a dream is not entirely outside the realm of possibility. In any case it is certain that the region of the transplutonium elements will remain a very interesting and fruitful field for investigation for the next decade or so.

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