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A HISTORY OF THE TRANSPLUTONIC ELEMENTS

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DISCLAIMER

A HISTORY OF THE TRANSPLUTONIC ELEMENTS

In the last twenty seven years it has been my good fortune to be associated with the discovery of all of the elements beyond plutonium. This time span has witnessed the development of experimental and theoretical techniques which encompass a very wide range--from the first almost intuitive gropings to find new elements to the modern sophisticated approaches where on occasion even single atoms can be identified with certainty. Now we are on the threshold of possibly opening up a vast new storehouse of knowledge if the predictions of a "magic island" of stability are borne out by experiment and this subject will be covered lightly at the concludion of this narrative.

Americium and Curium (Elements 95 and 96)

Early in 1944 GLENN T. SEABORG had pushed to near completion the fantastic development of the chemical processes to be used in the HANFORD plant that was to produce plutonium for weapons use. It was a natural step for him to undertake the task of trying to move further up the periodic table. For this purpose he assigned two of his young scientists, RALPH A. JAMES and LEON O. MORGAN, to the arduous chemical tasks that were to become necessary and asked me to concern myself with developing and using various detection devices needed in the experiments. The location of the discovery of the first two elements to be found, element 96, curium, and element 95, americium, was in the METALLURGICAL LABORATORY of the UNIVERSITY OF CHICAGO and took place in the wartime years of 1944 and 1945.

First attempts were utter failures. Small amounts of ^{239}Pu in the milligram range were bombarded by neutrons and deuterons but no new alpha particle emitters were to be found, partly because of the relatively insensitive techniques that we employed at that time and partly because the wrong chemical properties were assumed for element 95. In July, 1944, we tried a different approach and bombarded ^{239}Pu with 32 MeV helium ions accelerated by the BERKELEY 60" cyclotron. The irradiated target was flown to CHICAGO and was chemically processed in accordance with the newly-developed actinide concept. SEABORG had come to the conclusion that the elements beyond plutonium should have a characteristic stable (III) oxidation state and thus be readily separable from the plutonium target material by using a suitable chemical procedure.

On July 14, 1944 a new longer range alpha particle activity was found in the expected chemical fraction. The identification, though crude, was definite. A range curve was measured in a 2π air-filled pulse ionization chamber by placing weighed mica sheets over the sample and energetic alpha particles with a range of 5.86 mg/cm^2 were found. At the time we could not know whether the activity was due to element 95 or 96 since the reactions that were possible could produce isotopes of both elements.

(1) $${}^{239}_{94}\text{Pu} + {}^{4}_{2}\text{He} \longrightarrow {}^{242}95 + {}^{1}_{1}\text{p}$$

(2) $${}^{239}_{94}\text{Pu} + {}^{4}_{2}\text{He} \longrightarrow {}^{241}95 + {}^{1}_{1}\text{p} + {}^{1}_{0}\text{n}$$

(3) $${}^{239}_{94}\text{Pu} + {}^{4}_{2}\text{He} \longrightarrow {}^{242}96 + {}^{1}_{0}\text{n}$$

(4) $${}^{239}_{94}\text{Pu} + {}^{4}_{2}\text{He} \longrightarrow {}^{241}96 + 2\,{}^{1}_{0}\text{n}$$

We later found that reaction (3) is the one that was producing the new activity. The range of 6.1-MeV ^{242}Cm corresponds closely to the value measured. Later work showed that this isotope of element 96 could also be produced in neutron reactors by the series of reactions:

(1) $^{239}\text{Pu} + n \longrightarrow {}^{240}\text{Pu} + \gamma$

(2) $^{240}\text{Pu} + n \longrightarrow {}^{241}\text{Pu} + \gamma$

(3) $^{241}\text{Pu} \longrightarrow {}^{241}95 + \beta^-$

(4) $^{241}95 + n \longrightarrow {}^{242}95 + \gamma$

(5) $^{242}95 \longrightarrow {}^{242}96 + \beta^-$

During the rest of 1944 more vigorous bombardments of plutonium were made with deuterons. The targets were separated into fractions according to the newly learned chemistry and finally another alpha activity showed up. These new alpha particles had a range only about 0.5 mg/cm^2 longer than that of the ^{239}Pu alphas but the activity definitely followed the actinide chemistry. The counting rate of the new activity was very small by the standards of the time, perhaps a hundred alpha counts per minute. In retrospect we believe that we were observing the 5.5-MeV alpha particles from $^{241}95$ produced in part from the beta particle decay of the ^{241}Pu which was already present in the target material as a minor constituent. The characterization of this new activity was made in October, 1944. (See Fig. 2).

By January of the following year we had established the fact that both of these new activities were produced in neutron-bombarded plutonium. The picture portrayed above by the series of reactions thus gradually began to evolve. Subsequent experiments proved that this was indeed the correct interpretation when the isotope ^{238}Pu was identified as the alpha decay daughter of $^{242}96$ and when $^{242}96$ was produced by bombarding $^{241}95$ with neutrons. A further proof was furnished by the LOS ALAMOS mass spectrometer group when they discovered that the mass 241 fraction in a sample of plutonium could be fractionated. This came about because americium metal was more volatile than plutonium metal, thus enabling a separation of $^{241}95$ from its parent, ^{241}Pu.

The publication of this work (SEABORG et al., 1949) came in April, 1946, and at that time it was suggested that element 95 be named americium after the AMERICAS in analogy to its rare earth homolog, europium. Element 96 was named curium after PIERRE and MARIE CURIE, in analogy to its homolog, gadolinium, named after J. GADOLIN.

Isotopes of americium are now known from mass 237 to 246. The two most important isotopes are ^{241}Am and ^{243}Am with alpha half-lives of about 500 to 8000 years, respectively, and are available in kilogram quantities.

Isotopes of curium are presently known from mass 238 to 250. The most important isotopes are represented by masses 244 to 248, ranging in alpha half-life from 20 to 10^7 years and are available in subkilogram amounts.

Berkelium and Californium (Elements 97 and 98)

By the early months of 1946 SEABORG and many of his group had returned to the UNIVERSITY OF CALIFORNIA'S RADIATION LABORATORY and had begun to establish a powerful nuclear chemistry division. Much of the period until 1949 was spent in consolidating the chemical and nuclear knowledge of the elements through atomic number 96. We learned how to produce much greater quantities of the two new elements and new detection methods were studied to give greater sensitivity in finding new atoms. The development of gridded ionization chambers and multi-channel pulse analyzers now made it possible to characterize alpha particles which were emitted at rates as low as 1 per hour.

By 1949 we had accumulated a few milligrams of ^{241}Am from reactor bombardment of plutonium and begun attempts at creating element 97. In December of that year STANLEY G. THOMPSON, SEABORG, and I tried the following reaction (S. G. THOMPSON et al., 1950a):

$$^{241}_{95}\mathrm{Am} + ^{4}_{2}\mathrm{He} \longrightarrow ^{243}97 + 2^{1}_{0}\mathrm{n}$$

using 35 MeV helium ions from the 60" cyclotron.

The isotope was expected to have a short half-life so that a relatively fast chemical procedure was needed to enable identification of its alpha particle radiation. For this purpose we used a cation-exchange column for the first time in the heavy element regions. It had been shown that ion exchange chromatography was eminently suitable for separating the rare-earth elements--elements whose chemical properties were so similar that it was difficult to use ordinary aqueous chemical methods. SEABORG'S actinide hypothesis suggested that this method should be equally applicable to the new rare-earthlike series of (III) elements. In addition, the technique lent itself nicely to the making of thin samples for alpha particle measurement, an essential requirement in their energy analysis.

Following a bombardment and a chemical separation that took a few hours, we had what we believed was a fraction which should contain transcurium elements and examined it in our alpha pulse analyzer. We were very excited to discover that we had produced several tens of counts per minute of an activity with a totally new alpha spectrum. Three distinct peaks in the range of 6.5 to 6.8 MeV were observed and found to decay with a half-life of 4.5 hours. We subsequently found that the principal mode of decay was by orbital electron-capture and that we were only detecting about 0.1% of the decaying atoms of element 97. This meant that even though we only had about a thousandth as much ^{242}Cm to bombard, we might be able to find element 98 if we could produce an isotope which decayed principally by alpha decay.

We did not waste much time in trying this new experiment. In February, 1950, THOMPSON, KENNETH STREET, JR., SEABORG, and I bombarded a few micrograms of curium which had been produced for us in the high-flux CANADIAN reactor. The reaction again used 35-MeV helium ions from the 60" cyclotron.

$$^{242}_{96}\text{Cm} + ^{4}_{2}\text{He} \longrightarrow ^{245}_{98}\text{Cf} + ^{1}_{0}\text{n}$$

The very first experiment was successful. An alpha-particle activity with an energy of 7.1 MeV and a 44-min half-life was found to elute early from the ion exchange column in the predicted position for element 98. Several thousand counts were clearly observed and a very few experiments were sufficient to prove that element 98 had been discovered (THOMPSON et al., 1950b).

Element 97 was named berkelium and element 98 was christened californium to honor the city and university where the discoveries were made.

Isotopes of berkelium have been identified from mass 243 to 250. ^{249}Bk is a very low-energy beta emitter with a half-life of about 300 days, decaying to the 500 year alpha emitter, ^{249}Cf. It is produced now in subgram amounts in the United States reactor actinide production program. More suitable long-lived alpha-emitting isotopes with half-lives in the thousands of years are ^{247}Bk and ^{248}Bk but these can only be produced by accelerated positive ions. It is hoped that a national program can be started to produce these isotopes in milligram quantities by using the surplus beam from one of the 200-MeV proton injectors which are a part of the high-energy accelerators at Brookhaven and Batavia. Bombardment of gram quantities of ^{248}Cm should be capable of making the transmutations in a period of a year or two.

Isotopes of californium are now known from mass 242 to 255. Probably the most important of these is ^{252}Cf. It is a 2.6-year alpha emitter but decays by spontaneous fission 3% of the time. It is the neutrons from this decay which make it so important as a possible therapeutic agent in needles-implanted in patients. This nuclide was discovered by the Berkeley group in the "MIKE" debris in connection with the discovery of elements 99 and 100 which is described in the next section.

Einsteinium and Fermium (Elements 99 and 100)

On November, 1952, the first test thermonuclear explosion, code-named "MIKE," was set off by the U. S. in the PACIFIC OCEAN. This awesome event was the source of multi-neutron captures in the ^{238}U material which was included in the device and the new isotope ^{244}Pu was soon found in the mass spectrometer at the ARGONNE NATIONAL LABORATORY. This knowledge became known in our laboratory about a month after the explosion and we determined to see if even higher masses had been formed which would then beta-decay to elements of higher atomic number. Within a couple of days after receiving one of the filter papers which had been flown through the radioactive cloud we had indeed found a new alpha-emitter. This activity had an alpha energy of 6.6 MeV and a half-life of about 20 days and turned out to be ^{253}99. Again ion-exchange chromatography was used to identify a new element. It turned out to have a 17-day beta-emitting precursor ^{253}Cf and this was probably formed in the explosion in the following manner:.

$$^{238}\mathrm{U}(n,\gamma)^{239}\mathrm{U}(n,\gamma)^{240}\mathrm{U}(n,\gamma)^{241}\mathrm{U}(n,\gamma)^{242}\mathrm{U}\text{- - - - - - - - - - - - - - -}$$

$$\text{- -}\ ^{251}\mathrm{U}(n,\gamma)^{252}\mathrm{U}(n,\gamma)^{253}\mathrm{U}$$

$$^{253}\mathrm{U}\xrightarrow{\beta^-}{}^{253}\mathrm{Np}\xrightarrow{\beta^-}{}^{253}\mathrm{Pu}\xrightarrow{\beta^-}{}^{253}\mathrm{Am}\xrightarrow{\beta^-}{}^{253}\mathrm{Cm}\xrightarrow{\beta^-}{}^{253}\mathrm{Bk}\xrightarrow{\beta^-}{}^{253}\mathrm{Cf}$$

The neutron captures would have taken place in a fraction of a microsecond before the explosive separation of the material in the device and the subsequent beta decays from ^{253}U to ^{253}Cf would have taken place in seconds as the atoms were dispersed afterwards.

A few months later we were able to identify element 100 by working up large quantities of coral containing nuclear debris which had fallen out onto a nearby island. The isotope found was the 1-day 7.1-MeV $^{255}100$ which had been kept alive by a longer-lived isotope of element 99, the 40-day $^{255}99$. This isotope had been formed as the result of the addition of seventeen neutrons to the ^{238}U target atoms. Figure 1 is an elution curve for one of the experiments in which element 100 was discovered.

The work on these elements was finally published (GHIORSO et al., 1955a) by a large team of scientists from Berkeley, Argonne and Los Alamos who had participated in various ways in the exciting work. They suggested the names einsteinium and fermium for elements 99 and 100 and the symbols Es and Fm were eventually adopted.

One of the very important discoveries that emerged from this work was the disclosure of a nuclear subshell at 152 neutrons (GHIORSO, et al., 1954). This first became apparent when we found that the alpha energy of ^{252}Cf was almost a hundred kilovolts higher than that of ^{250}Cf. If the normal trend had continued its energy would have been lower by several hundred kilovolts. This is a very sensitive indication by even-even isotopes and eventually this shell structure was found to extend all the way to element 105. How much further it will persist is still a matter of speculation.

Isotopes of einsteinium have been identified from mass 243 to 256. The 20-day isotope with mass 253 is the one normally encountered at the moment but within a few years it is expected that the 6.44-MeV 276-day ^{254}Es will be available in multi-microgram quantities. Isotopes of fermium are known from mass 244 to 258. The longest-lived isotope is 6.53-MeV 80-day ^{257}Fm, but unfortunately it will probably never be possible to manufacture it in more than submicrogram quantities by means of neutron reactors. It is possible to make it in milligram quantities in an underground nuclear explosion by the rapid-neutron-capture method but the costs of the explosion and the chemical recovery of the material from the debris are very high.

Mendelevium, Md (Element 101)

The discovery of mendelevium was probably the most dramatic in the entire sequence of the evolution of the transuranium elements. It marked the first time in which a new element was produced and identified one atom at a time.

Within a year after the discovery of einsteinium and fermium in the "MIKE" explosion we found that we could produce the same nuclides by means of high flux neutron reactors. By 1955 we had transmuted a substantial percentage of our initial plutonium starting material into the heaviest isotopes of curium so that the equilibrium amount of ^{253}Es that was available for experiments amounted to about 10^9 atoms. It occurred to us in a very optimistic moment that it might be possible to form element 101 by the reaction

$$^{253}_{99}Es + ^{4}_{2}He \dashrightarrow ^{256}101 + ^{1}_{0}n$$

The obvious problem that we had to overcome was that the target material had only a 20-day half-life. In order to perform many experiments quickly we needed a nondestructive technique of using the target material over and over again. We decided to rely on the recoil energy that was imparted to a transmuted atom in the collision of a high speed projectile with a target atom, a technique which had never been used before. Since the recoil energy was very small in the reaction that we had selected, many new problems had to be overcome before we were successful, but finally we were able to devise a procedure which enabled us to make many bombardments before the target became useless.

Again we used the old reliable 60" cyclotron, this time with 40-MeV helium ions. The recoiling product atoms coming from the very thin einsteinium target were caught in a gold catcher foil which was then dissolved and subjected to the usual cation-exchange chromatographic separation. At first we concentrated on experiments which were aimed at finding an alpha emitter since we expected that to be the most likely mode of decay. We sped up our procedures fast enough to be able to see a 5-minute activity but without a sign of high energy alpha particles and were beginning to despair of finding anything. Finally, for we had nothing to lose, one of the irradiations was prolonged in the hope that maybe something longer-lived in the range of an hour or two might show up. We had been concentrating in the range of 5-10 minutes. Still no new alpha counts showed up in the alpha pulse analyzer but after a very long interval a single event occurred that showed up on a paper chart recorder. The recorder had been set up to measure the height of very large ionization pulses that would occur in the ion chamber whenever a spontaneous fission took place in the sample. Somewhat later, after another long bombardment, another such "big kick" was detected!

At first we were puzzled by these events but then we suddenly realized that they must be the key to the whole problem of why we could not see any new alpha activities. On the basis of these two spontaneous fission counts we hypothesized that $^{256}101$ had a half-life in the hour range and decayed by electron-capture to ^{256}Fm. This daughter isotope would then decay by spontaneous fission with a half-life also in the hour range. These were far-reaching conclusions to draw on the basis of just two events but they turned out to be quite correct! The proof of our conjecture was provided by a classic experiment performed on February 18, 1955 in which we performed three successive experiments and combined the products in the chemical separation step. The chromatographic elution gave a fraction in the predicted position for element 101 which exhibited five spontaneous fissions with a half-life of roughly three hours while the element 100 fraction which eluted somewhat later than element 101 showed eight spontaneous fission events with about the same half-life. Figure 4 shows the original elution data.

Further experiments conducted before our target had decayed too much yielded further corroboration and by the time that we published our work (GHIORSO et al., 1955b) we had successfully separated a total of seventeen atoms of element 101 with an average of about one per experiment! In this paper the team of BERNARD G. HARVEY, GREGGORY R. CHOPPIN, THOMPSON, SEABORG and I suggested that the new element should be called mendelevium after the great RUSSIAN chemist, DIMITRI MENDELEEV, who had made such great contributions to chemistry. His periodic law accurately predicted the chemical properties of known and unknown elements and was the basis for the discovery of many of them.

Isotopes of mendelevium are known from mass 251 to 258. ^{258}Md has an 80-day alpha half-life and is made by bombardment of ^{255}Es with helium ions.

Nobelium, No (Element 102)

The First of The "Controversial" Elements

Temporarily we had come to the end of the road as far as making new elements was concerned. The heaviest target material that we had was element 99 so that we had to have a projectile with at least three protons to make element 102. We had been experimenting with the acceleration of heavier particles such as carbon ions in the 60" cyclotron but the beams were very small and did not have a well-defined energy. We had been able to make very small amounts of isotopes of Cf, Es, and Fm so that it did appear feasible to go further up the atomic scale, but we needed a better accelerator for the purpose. Following a suggestion by LUIS ALVAREZ a study was set up in collaboration with YALE UNIVERSITY with the object of designing twin heavy ion linear accelerators (HILAC) to be built at the two research centers. These machines turned out to be the first in the world to take advantage of magnetic strong-focussing. Their characteristics were set by the requirements of the experiments envisioned at that time and by the availability of funds. The maximum ion energy was limited to 10 MeV per nucleon. The maximum Z that could be accelerated was around argon. The maximum average beam current was a few microamperes. Construction of the BERKELEY HILAC was completed early in 1957 and after a long de-bugging stage experiments were envisioned looking toward the production of element 102. Before these could be carried out we were surprised by an announcement from SWEDEN in the summer of 1957.

A group of workers from the ARGONNE NATIONAL LABORATORY, the ATOMIC ENERGY ESTABLISHMENT at HARWELL in ENGLAND, and the NOBEL INSTITUTE for PHYSICS in STOCKHOLM claimed (FIELDS et al., 1957) the discovery of an isotope of element 102 as a result of research done at the Nobel Institute. The group reported that in irradiations of ^{244}Cm with ^{13}C ions accelerated in the NOBEL INSTITUTE'S cyclotron they had found an 8.5-MeV alpha emitter with a half-life of about 10 minutes. They claimed that it had been identified as being an isotope of element 102 by the chromatographic ion exchange method and proposed that the new element be given the name nobelium to honor the NOBEL INSTITUTE. The name was accepted within a matter of months by the COMMISSION on ATOMIC WEIGHTS of the INTERNATIONAL UNION OF PURE AND APPLIED CHEMISTRY which had become the international arbiter of chemical nomenclature.

This acceptance of the claim to discovery of element 102 turned out to be premature. All attempts to duplicate the STOCKHOLM experiment failed to corroborate their findings. At BERKELEY we devoted many months of time with the new HILAC using research techniques orders of magnitude more sensitive than those used in SWEDEN and yet failed to find the reported activity (GHIORSO et al., 1958a). Later we were were to find out that experiments performed in DUBNA in the USSR at about the same time also did not verify the discovery but these could not rule it out because of the use of a different target and projectile combination. Although we used the same kind of target and projectile as used in STOCKHOLM without success the authors of the "discovery" paper refused to withdraw their claim feeling that there was some possibility, even though small, that their work was correct. Even this remote possibility was completely discounted in 1968 when we found that element 102 was found only in the (II) state in aqueous solution. This meant that it would not appear in the expected elution position in a cation exchange column and thus their chemical proof as well as their physical proof for element 102 vanished.

After our unsuccessful attempts to find the STOCKHOLM alpha particles we finally began to look in various ways for element 102 alpha activities with other characteristics. We felt that the half lives of the isotopes that we could make at that time would be in the seconds region. Since the technology was not yet available for the efficient direct detection of the short-lived alpha activities themselves, we resorted to a new method. In a paper (GHIORSO et al., loc. cit.) published in the summer of 1958 by TORBJORN SIKKELAND, JOHN R. WALTON, SEABORG, and the author a novel "double recoil" technique was described which permitted the identification by both physical and chemical means of an isotope of element 102. A mixture of curium isotopes was bombarded with 68-MeV ^{12}C ions provided by the new Berkeley HILAC. The following reactions were assumed to take place:

$$^{246}Cm + ^{12}_{6}C \longrightarrow ^{254}102 + 4\,^{1}_{0}n$$

$$^{254}102 \xrightarrow[\text{secs}]{\alpha} {}^{250}_{100}Fm \xrightarrow[\text{30 min}]{\alpha}$$

As shown in Fig. 5, a thin curium target with 95% ^{244}Cm and 4.5% ^{246}Cm was set up in such a manner that the recoiling element-102 atoms produced in the bombardment were stopped in helium gas and then attracted electrically as positive ions to a moving metallic belt mounted nearby. In a second recoil action approximately half of the 30-min ^{250}Fm atoms from the alpha decay of $^{254}102$ were captured by a negatively-charged foil adjacent to the belt. The discovery of element 102 was based on showing that the doubly-recoiled atoms had the atomic number of fermium. For this crucial test it was necessary to use all of the transferred atoms in each experiment to make the chemical identification and thus no half-life information was possible.

The half-life of the element 102 parent was deduced by a completely physical method. From the speed of the belt and the variation of the ^{250}Fm activity along the catcher foil, we obtained information that was consistent with a half-life of approximately 3 seconds for the 102 parent of this alpha emitter. About a year later in experiments in which element-102 atoms were attracted electrically to a stationary metallized tape which was then periodically moved into counting position within a gridded alpha chamber we found an alpha activity of 8.3 MeV and about 3 seconds half-life. We also observed spontaneous fissions with the same half-life in 30% abundance relative to the alpha emissions. Since the half-life was the same as that observed in the earlier experiment we quite naturally assumed that these characteristics corresponded to ^{254}102 which we had established by the chemical milking of ^{250}Fm.

Before this work could be brought to a satisfactory conclusion it was abruptly halted in 1959 by a disastrous radiation incident. A helium-cooled window which was interposed in the beam line between the HILAC and the curium target was ruptured by an accidental momentary overpressure. Unfortunately, the foil on the target side failed and an explosively propelled jet of helium impinged upon the fragile ^{244}Cm target. The target contained about 10^{12} alpha disintegrations per minute and this was carried into the experimental room in micron-sized particles. Unfortunately, at that time there was no secondary containment in the room and the activity was carried out into the high-bay area. The very dangerous ratioactive dust was soon deposited uniformly throughout the structure! Fortunately, I detected the spill almost immediately and gave the alarm to evacuate everyone in the building. Miraculously no one ingested a significant amount of curium and it did teach us an extremely important lesson. It was estimated that I inhaled possibly 10^5 α dis/min and I was the closest to the explosion. By an heroic effort of Health Chemistry personnel the building was ready for occupancy again in about three weeks but for many years curium continued to be found in small quantities in obscure places in the building.

The incident not only disposed of all of our precious curium that was available at the time but understandably caused us to be quite gun-shy in the use of highly active alpha-emitting targets. We might have pursued the element-102 work by using plutonium targets and oxygen ions but we knew that the smaller reaction cross sections would make the problem even more difficult so we decided to push on to try to find an isotope of element 103.

In 1961 during work which led to the successful identification of element 103 we reported finding another isotope of element 102. This activity, produced by the HILAC bombardment of a mixture of californium isotopes by boron projectiles, decayed by the emission of 8.2-MeV alpha particles with a half-life of approximately 15 secs. We assigned this activity to $^{255}102$ on the basis of comparative yields of this and other product activities and the variation of its yield with energy of the bombarding particles. This mass assignment was not definite since the excitation function was very broad, both because it was the product of a (^{11}B,pxn) reaction and because the target was not monoisotopic.

Let us now return to 1957 to look at the parallel line of research being undertaken by large scientific teams in the USSR. The director of this work was GIORGI N. FLEROV, a pioneering nuclear physicist who had discovered spontaneous fission in ^{238}U, following OTTO HAHN'S famous report on the discovery of neutron-induced fission. FLEROV and his men did not have curium available to them at this time for the production of element 102. Even if they had it is doubtful that they would have used it since they had been similarly "burned" by a radioactive incident with the 500-year ^{241}Am. They chose instead to bombard the relatively inactive isotopes of plutonium with ^{16}O and ^{18}O ions. At the KURCHATOV INSTITUTE in MOSCOW the cyclotron had been converted to accelerate heavy ions for the purpose of making new transuranium elements and they were hard on the heels of the BERKELEY group.

Their first interesting result (FLEROV, 1958) was obtained in the autumn of 1957 in bombardments of ^{239}Pu with ^{16}O ions in which they found an alpha activity with an energy of 8.8 ± 0.5 MeV having a half-life in the range of 2 to 40 seconds. The SOVIET scientists bombarded targets containing 100-200 micrograms/cm^2 of ^{241}Pu with intense currents of ^{16}O ions. The product atoms were caught in vacuum in an aluminum catcher foil that was cyclically shuttled a distance of 200 cm in 2 seconds and placed next to a nuclear emulsion. (See Fig. 6 Optical scanning of the emulsion and a measure of track lengths gave them a rough capability for detection of alpha particles. (Fig. 6).

Much or all of the 8.8-MeV activity turns out to be caused by an isomer of polonium, the 25-sec 8.87-MeV ^{211m}Po, produced by heavy ion interaction with a tiny lead impurity in the target material. This same activity plagues all experiments in this field even today since it simulates some isotopes of 103, 104, 105 and probably 106 and 107! It is likely that some of their alpha counts were indeed due to 2.3-sec 8.43-MeV ^{252}102 but the very crude resolution afforded by their use of nuclear emulsions for detectors makes it impossible to discriminate against the background activities (another is 8.43-MeV ^{214}Fr kept alive by 3-sec ^{214}Ra produced from lead impurity). In Fig. 7 is shown their gross alpha-particle spectrum obtained from ten 3-hour bombardments. Although it certainly is possible that element 102 was first produced in these experiments since historically they preceded the Berkeley efforts, I do not believe that they can be considered as a discovery of the element since they do not persuade any knowledgeable person that such an identification was made.

SOVIET experimenters soon moved to the new DUBNA LABORATORY, an international research center set up cooperatively by the SOCIALIST countries. There they began using a very large 3.1 meter cyclotron which had been constructed specifically for heavy ion acceleration. They started a new series of experiments which culminated in 1963 with the successful identification of a new element-102 isotope (DONETS et al., 1964; DRUIN et al., 1964). In bombardments of ^{238}U with ^{22}Ne ions they produced ^{256}102. The method was based on the BERKELEY double-recoil technique with the chemical identification of the alpha emitting daughter, ^{252}Fm. The half-life of ^{256}102 was deduced to be about 8 seconds from the relative amounts of daughter activity transferred by alpha recoil to the catcher foil. (A better value of 3.2 seconds was later determined by our group).

In December, 1965, FLEROV and his group identified (DONETS et al., 1966) ^{254}102 by the double-recoil method and arrived at the surprising conclusion that its half-life was 50 ± 10 secs, not 3 seconds as indicated in our 1958 work, and this result was soon verified by another method of production. At about this time they also began to use a new method, excellent for alpha spectroscopy, which had been pioneered at BERKELEY by MACFARLANE and the author a few years before.

The method was the so-called "gas-jet" technique to transport recoil atoms, which were stopped in a carrier gas such as helium, to a place of collection where they could be examined by sensitive semi-conductor alpha particle detectors. In the SOVIET version of this method the jet of helium gas impinged upon one arm of a collector shaped like a Maltese cross. After a suitable bombardment, the cross was periodically rotated 90° to put the collected heavy atoms in front of a detector while another arm was simultaneously placed to collect a new batch of jet-borne atoms from the target chamber. With a two-parameter analyzer the decay and energy of the various alpha activities could be followed by noting the time of arrival and pulse size of each event after each rotation of the cross.

With the aid of this modern technique and the employment of a second heavy-ion accelerator, a new 1.5 meter sector-focussed heavy-ion cyclotron at DUBNA, a SOVIET group (ZAGER et al., 1966) identified the alpha particles from $^{254}102$ and found that they had an energy of 8.10 MeV and a half-life of 30 to 40 seconds, again much longer than the old BERKELEY value. The mass assignment was proved when they detected the 30-min ^{250}Fm daughter nuclei which recoiled into the detector face after alpha decay of $^{254}102$. They looked for spontaneous fissions and concluded that this isotope did not exhibit a prominent branch decay by this mode as indicated by the 1959 BERKELEY work. Later another group identified $^{252}102$ as emitting 8.41 MeV alpha particles with a half-life of 4.5 ± 1.5 secs. Spontaneous fissions which were observed in this case with about the same half-life were attributed to a lower-Z element. The isotope $^{255}102$ was found to have an 8.08 MeV energy and a two minute half-life and thus this nuclide also did not correspond to our assignment. Two other activities were found so that by October, 1966, at an INTERNATIONAL HEAVY ION CONFERENCE held at DUBNA FLEROV's groups were able to announce the seemingly complete identification of isotopes of element 102 from mass 252 to 256.

Responding to this challenge by the SOVIET scientists, whose results seemed to be at variance with all of the earlier BERKELEY research on element 102, we set out in November 1966 to repeat the old experiments with the objective of clearing up the discrepancies. This time we had the advantages of greatly improved HILAC beams and superior detection capabilities. In a relatively short time we were able to produce and identify all of the isotopes of element 102 from 251 to 257 (GHIORSO et al., 1967a) and a few years later we identified masses 258 and 259. For most of these experiments we used a gas-jet system to transport the transmutation products to the periphery of a wheel. The wheel acted as a conveyor to successively place the collected atoms in front of a series of four semi-conductor alpha-particle detectors for energy analysis. The relative numbers of atoms observed at each detecting station gave a measurement of the half-life of each species detected.

To span the range of the element-102 isotopes, we bombarded at various energies monoisotopic targets of ^{244}Cm and ^{248}Cm with concentrated beams of ^{12}C and ^{13}C ions. Compound nucleus reactions with these target-ion combinations had the distinct advantage, as compared with the combinations used by the DUBNA groups, of much larger cross sections to produce the same nuclide. This enabled us to make substantially more accurate measurements of alpha half-lives and energies and more definitive observation of spontaneous fission branching decay.

The first observation that we made was that ^{254}102 did not indeed have a 3-second half-life as claimed in our 1958, 1959 papers; a value of 55 ± 5 sec was found in agreement with Dubna. On the other hand its formation cross section was about a microbarn, consistent with the value found in 1958 in our milking experiments when we chemically isolated its daughter, ^{250}Fm. Clearly there was some error in the milking experiments which implied a 3-second half-life by the physical identifications of "^{250}Fm."

The next step in unraveling the puzzle was finding that ^{252}102 had a half-life of 2.3 ± 0.3 secs 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, a branching ratio for spontaneous fission of about 30% was found and this important fact was in excellent agreement with the work of 1959 when we used a grid chamber as our detector. The old 8.3-MeV energy measurement can be corrected to 8.4 MeV by using the modern value for the alpha energy of ^{248}Fm as an internal standard and the old 3-sec half-life is within the standard deviation errors in the two measurements.

It is impossible to be certain as to the reason why our original experiments gave the <u>wrong half-life</u> for ^{254}102 but it is likely that the confusion was caused by ^{252}102. Since there was 20 times as much ^{244}Cm in the old target as ^{246}Cm, it is possible that the 25-min 7.22-MeV ^{244}Cf granddaughter could have been mistaken for 30-min 7.43-MeV ^{250}Fm.

$$^{252}102 \xrightarrow[\alpha]{2.3\ \text{sec}} {}^{248}\text{Fm} \xrightarrow[\alpha]{36\ \text{sec}} {}^{244}\text{Cf}$$

In those early experiments we were plagued by relatively poor energy resolution and drift problems in the multi-chamber system used to analyze the small amounts of activity available at that time. In the chemical milking experiments we were not bothered by this problem since ^{244}Cf was separated from the ^{250}Fm by the ion-exchange column.

The isotope, ^{257}102, was identified in later experiments and found to have prominent groups at 8.22, 8.27, and 8.32 MeV with a half-life of 23 ± 2 secs. These data correspond reasonably well with the 15-sec 8.2-MeV alpha activity observed in 1961 in connection with the experiments on element 103.

We believe that it is fair to draw the following chronological conclusions regarding the discovery of element 102:

a) The 1957 work at the NOBEL INSTITUTE was completely in error.

b) The 1957 work at DUBNA was so vague and inconclusive as to be virtually meaningless although, of course, it is possible that the radiations from an isotope of element 102 were included as a small part of their range spectrum.

c) In 1958 at BERKELEY the isotope, ^{250}Fm, was <u>chemically</u> shown to exist in a recoil-milked fraction thus proving the atomic number of the ^{254}102 parent.

d) In 1959 at BERKELEY the isotope, ^{252}102, was discovered as shown by its alpha energy, half-life, and spontaneous fission branch decay.

e) In 1961 at BERKELEY the isotope, ^{257}102, was discovered as shown by its alpha energy and half-life.

f) In 1963 at DUBNA for the first time in <u>their</u> laboratory the isotope, ^{256}102, was positively identified by the chemical milking of the recoil daughter, ^{252}Fm.

At the time this definitive work was completed ten years had passed since the original STOCKHOLM experiments and the name nobelium had come into common usage. As a consequence of this situation, we decided not to contest the name. In a review paper (GHIORSO et al., 1967b) on the element 102 research we suggested that it be retained, but the symbol No for the element will always be for us a sharp reminder of its somewhat painful history.

Quite recently a relatively long-lived isotope of nobelium has been produced. This is ^{259}No, an alpha emitter with a half-life of about an hour, which should make possible more extensive studies of the chemistry of element 102. The important discovery of the divalent characteristics of nobelium in aqueous solutions was made using the 3-min ^{255}No isotope.

Lawrencium (Lr) (Element 103)

In 1961 SIKKELAND, ALMON E. LARSH, ROBERT M. LATIMER, and the author reported the discovery (GHIORSO et al., 1961) of element 103. In this case it was not possible to identify chemically a known daughter isotope and thus prove the atomic number of the new activity, so instead we had to rely on purely physical arguments. The recoiling atoms from the bombardment of a mixture of californium isotopes by either ^{11}B or ^{10}B ions accelerated by the HILAC were electrically attracted to a metallized mylar tape which was pulled successively in front of four semi-conductor alpha detectors. The art of making and using solid state detectors was in its infancy so there were many problems to be overcome. The great value of these sensitive detectors was proved in these experiments since the amount of activity that we could make was very small. Only about 100 total alpha events were observed in all of the many experiments so that there were many possibilities for error. We reported an 8.6-MeV alpha activity with a half-life of about 8 seconds. By cross bombardment and substitution experiments we came to the conclusion that the new activity was due to element 103 and that its mass number was probably 257 although 258 and possibly even 259 were also possibilities. We suggested the name lawrencium for the new element in honor of E. O. LAWRENCE, the pioneering inventor of the cyclotron.

In 1965 the DUBNA groups under FLEROV again challenged our claim to the discovery of a new element. They found an alpha emitter, 45-sec ^{256}Lr, which they identified chemically through its granddaughter, ^{252}Fm, using the same double-recoil system that had been successful in the identification of ^{254}No and ^{256}No (DONETS et al., 1965). During the next several years they looked for ^{257}Lr and finally concluded (FLEROV et al., 1967) that it had the same alpha energy and half-life as ^{256}Lr. Since they did not find our 8.6 MeV alpha activity, they assumed that it must be due to some error on our part.

The SOVIET experiments were not too surprising to us and we deduced from them that the mass number of our lawrencium isotope was most likely 258. Faith in our work was not misplaced for by 1968 we had proven quite conclusively that this was indeed the correct assignment (GHIORSO et al., 1969) and that its half-life was closer to 4 seconds than to 8 (ESKOLA et al., 1971). Since it is the atomic number that is essential in the determination of a new element, we take the position that a wrong assignment of mass number is of no consequence--in fact, is almost to be expected in the first exploratory measurements. In the new experiments, contrary to the DUBNA work, we found that ^{257}Lr did not have the same characteristics as ^{256}Lr, but instead had a half-life of 0.6 sec and an energy of 8.87 MeV (ESKOLA et al., loc. cit.). This proved to be a crucial factor in the identification of isotopes of element 105.

In 1970 our group (SILVA et al.,1970a), in a tour-de-force of chemistry, succeeded in showing that lawrencium reverted to the III state in aqueous solution as expected by the actinide hypothesis. This extremely difficult experiment employed the 35-sec ^{256}Lr isotope so that speed was vitally necessary. A solvent extraction method was used to distinguish between 2+, 3+, and 4+ oxidation states. On the average one atom was detected in each manually-performed experiment and several hundred experiments were eventually conducted.

By 1971 our group was able to greatly extend the list of known isotopes of lawrencium from mass 253 to mass 260. ^{260}Lr is an alpha emitter with a half-life of 3 minutes and thus the longest-lived isotope known.

Element 104--Rutherfordium (Rf) or Kurchatovium (Ku)?

There is certainly no question that G. N. FLEROV and his DUBNA colleagues made the first claim (FLEROV et al., 1964) to the discovery of element 104. In 1964 the isotope ^{242}Pu was bombarded with ^{22}Ne in the 3.1-meter heavy ion cyclotron with the objective of making a spontaneously-fissioning isotope of element 104. They found a new 0.3-second activity which they concluded was probably due to ^{260}104 but the number of fission events observed was so small that it was extremely difficult to be certain as to the identity of the atomic number involved. At best, even with a lot of activity, it is difficult to define the atomic number of a spontaneous-fission emitter since in the heavy element region the energy and mass distributions of the fragments do not differ greatly from one element to another.

Starting in 1966, efforts were made at DUBNA to support their claim by chemical experiments designed to show that the spontaneous-fission activity came from a non-actinide element. It was expected that element 103 would be the last of the 3+ actinide elements; element 104 should thus have chemical properties similar to hafnium and zirconium and form a relatively more volatile chloride compared to the actinides. I. ZVARA et al. (1966) at DUBNA set up a very complicated system in which the nuclear reaction products from the cyclotron bombardment were chlorinated by reaction with $NbCl_5$ and $ZrCl_4$ vapors at 200° to 300° C. A small fraction of the total spontaneous-fission activity was able to pass through the hot filter and it was deduced that only the more volatile 104-tetrachloride would be able to do so in accordance with predictions. The apparent half-life of the activity which survived the chemical treatment seemed to agree with the 0.3-sec value obtained in the purely physical experiments but the number of detected events was small and the variation very large.

Since the chemical experiments seemed to confirm the physical experiments, the DUBNA groups decided that element 104 had indeed been discovered and suggested that it be given the name kurchatovium (Ku) in honor of IGOR KURCHATOV, a pioneering SOVIET nuclear physicist. The international nomenclature committee has not accepted this name since the DUBNA work was soon questioned by the BERKELEY group. Suddenly, for a change, we were the challengers rather than the challenged!

At first we were inclined to accept the SOVIET work since it sounded convincing and we were deeply immersed in the problems associated with the nobelium isotopes. Our doubts were aroused, however, when we discovered that ^{258}No had a spontaneous-fission half-life of only a millisecond. Over a period of time we had formulated an empirical spontaneous-fission systematics (GHIORSO et al.,1958b) which indicated that there was a maximum half-life for this mode of decay for those nuclides with 152 neutrons and that as the atomic number was increased the half-lives became drastically shortened (see Fig. 8). Following this view we would expect that ^{260}104 with the same number of neutrons (156) as ^{258}No should have an even shorter half-life, perhaps in the microsecond region.* This argument would not rule out the possibility that the DUBNA activity was due to ^{261}104 or ^{259}104 since nuclides with an odd number of neutrons tend to be highly hindered as to spontaneous-fission decay.

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*Considerable strength to this argument was added in 1970 by the discovery that 158-neutron ^{258}Fm had an SF half-life of only 300 microseconds (HULET et al.,1971).

We set to work to see if we could produce the 0.3-sec activity by bombarding einsteinium, californium, and curium isotopes with ions from ^{11}B to ^{18}O. In experiments which should have been successful we did not see any such short-lived spontaneous fissions but our efforts were hindered by a considerable background from ^{256}Fm, the 2.7-hr SF emitter. After expending much effort in this direction without success we decided to reserve judgment and continue in our efforts to find an alpha emitter which could be assigned without ambiguity to element 104.

An atom that disintegrates by undergoing spontaneous fission provides little in the way of a "fingerprint" since one fission differs little from another and only the half-life can be used to distinguish them. On the other hand an atom that decays by the emission of an alpha particle with a distinctive energy and half-life to form a daughter atom which in turn decays with a different alpha energy and half-life can be uniquely identified. With this technique as our ultimate goal we proceeded with our search for element 104 and succeeded in developing over a period of time a marvelously sensitive apparatus that was vital to the discovery of the alpha-emitting isotopes of elements 104 and 105.

Early in 1968 enough ^{249}Cf became available to allow us to bombard it effectively with carbon ions and hope to see short-lived alpha activities. After some initial experiments which disclosed that there was 3-5 sec alpha emission in the region of 8.8 MeV, we began the development of a system which used a vertically-mounted large wheel as a conveyor for the jet-deposited new atoms. (See Fig. 9) This new system was mechanically and electrically superior to our old apparatus used for the investigation of the nobelium isotopes. In addition we equipped it with auxiliary detectors which would allow us to identify the element-102 daughter products simultaneously with the identification of the element-104 mother atoms. A small computer was installed to make it possible to handle the wealth of information that was to ensue.

These mother-daughter measurements were accomplished in the following way. Just as in the double-recoil experiments which were successful in the case of element 102, we took advantage of the separation of daughter from mother afforded by the recoil energy after alpha decay. By periodically moving the detecting crystals which were "looking at" the alpha particles emitted by the mother atoms on the conveyor wheel to positions off the wheel, we were able to measure the activities from those atoms which had been embedded in the crystals by 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 mother and daughters simultaneously. This meant the use of four detectors at each station, each with its own preamplifier-amplifier system. The 28-detector system, though highly complex, performs its functions with great precision and stability. The reproducibility of a given experimental measurement is excellent and it is possible to run the system for days at a time with little more than minimal attention. As the work on element 104 progressed, the system, which started with only four crystals, was gradually improved until, with the conclusion of the element-105 work, we had built it up to the use of twenty-eight detectors.

With the advent of the new equipment we were able to identify (GHIORSO et al., 1969) unambiguously the isotope, $^{257}104$, in ^{12}C bombardments of ^{249}Cf. It was found to have a complex alpha spectrum (See Fig. 10) with lines ranging from

8.70 to 9.00 MeV and a half-life of 4.5 ± 1.0 secs. Its known daughter, ^{253}No, was found to be transferred by alpha recoil as expected. The same target when bombarded by ^{13}C ions produced another isotope of element 104, this one with mass 259 having two prominent groups at 8.77 and 8.86 MeV and a somewhat shorter half-life of 3 seconds. Again the recoil daughter was identified, this time the isotope, ^{255}No. By using a different method we discovered an 11-msec SF activity which we tentatively assigned to ^{258}104. This assignment agreed well with our predictions for this nuclide and further cast doubt on the origin of the 0.3-sec SF's. The members of the research team publishing these results, MATTI J. NURMIA, JAMES A. HARRIS, KARI ESKOLA, PIRKKO ESKOLA, and the author suggested that element 104 be named rutherfordium with the symbol Rf in honor of LORD ERNEST RUTHERFORD.

About a year later in bombardments of ^{248}Cm with ^{18}O ions, a much longer-lived isotope of element 104 was found, ^{261}104, with a half-life of 65 ± 10 seconds and an alpha energy of 8.3 MeV. Once more the recoil daughter, this time ^{257}No, was shown to be transferred into the detecting crystals as they viewed the wheel. These findings seemed to reinforce our empirical criticisms of the assignment of the 0.3-sec SF activity to element 104 since now its mass could not be 259 or 261.

After the publication of our research, the DUBNA groups endeavored to repeat their work on element 104. Now they found that the half-life of the SF activity was about 0.1 sec and that these recoil products tended to be ejected in a cone with a sharply-peaked forward distribution. The angular distribution experiment would rule out the possibility that the 0.1-sec activity could be due to an isomer of a nuclide near the target nucleus in atomic or mass number but could not rule out one in the region of nobelium or lawrencium. The smaller value for the half-life also meant that the old chemical experiments could not have worked since there was a minimum time of about a second necessary for the atoms to transit the hot filter. Very recently they have measured a 4.5 ± 1.5-sec component in their ^{242}Pu + ^{22}Ne bombardments which they attribute to ^{259}104 and claim that this must be the SF activity which they observed in their chemical experiments. It is certainly possible that this is a correct interpretation since our old experiments were not set up to rule out such a possibility. It would be very surprising, however, if this turns out to be the case since ^{261}104 certainly does not have a prominent SF-branching ratio. One possibility is that the new SF activity is actually due to ^{252}No produced by the reaction ^{238}Pu(^{22}Ne,α4n) since ^{238}Pu is present in their ^{242}Pu target to the extent of about 1%.

In 1970 the first aqueous chemistry (SILVA et al., 1970b) was performed with rutherfordium. Although the half-life of the isotope used, the 65-sec ^{261}Rf, was longer than that available for the lawrencium chemical experiment, the number of events was an order of magnitude less. As a consequence of this difficulty we were not able to make the tests as complete as one would like. After several hundred individual experiments aimed at the separation of element 104 from the actinide elements by means of a cation-exchange column, we had a total of some seventeen events which showed that rutherfordium had an aqueous chemistry similar to that of hafnium. Further chemical tests are intended when we have successfully completed an automatic system which can repeat experiments over and over again. It is just too difficult to do this sort of operation manually!

Hahnium (Ha)--Element 105

Again the SOVIET groups were the first to make a claim (FLEROV et al, 1968) on the discovery of element 105 and again we were to challenge the validity of their experiments. The system used at DUBNA consisted of catching the jet-borne atoms on the flange of a wheel which was viewed by an annular semiconductor detector through which the helium jet passed. Whenever an interesting event occurred the wheel would be stopped for an interval long enough to allow detection of an alpha particle emitted by the element-103 daughter atom. In bombardments of ^{243}Am with ^{22}Ne projectiles they found a miniscule number of "correlated" alpha events which they attributed to ^{260}105 and ^{261}105. As indicated above they looked for delayed coincidences of alphas in the range 8.8 to 10.3 MeV succeeded by those in the range 8.35 to 8.6 MeV. They tentatively concluded that there was a positive effect beyond statistical variation at 9.4 and 9.7 MeV. The extremely low rate involved, approximately one per day, made it very difficult to refine these data; they reported that the half-life for the first activity was between 0.1 and 3 seconds and that of the second was probably somewhat greater than 0.01 seconds.

The DUBNA conclusions depended completely on the assumption that the 8.35 to 8.6 MeV window in the element-103 daughter alpha spectrum included the emissions for ^{256}Lr and ^{257}Lr and that their half-lives were both about 35 seconds. Now we know that ^{257}Lr has an energy of about 8.9 MeV and a half-life of only 0.6 seconds and would not have been detected in the above experiment, thus ruling out the detection of ^{261}105. The identification of ^{260}105 in this work is invalid since, as will be shown shortly, its alpha energy is several hundred kilovolts less than 9.4 MeV.

Returning to the BERKELEY work now I will review the sequences of our element-105 research. In November 1968 after we had found our first traces of rutherfordium we tried bombarding our ^{249}Cf target with ^{15}N ions in the hope that the production cross section might be high enough for us to also see a trace of element 105. In a couple of long runs we observed 8.9-MeV alpha particles with a short half-life and, in ten times smaller abundance, a peak at 9.1 MeV. We were discouraged from pursuing the matter to a conclusion when we discovered that the 8.9-MeV peak could also be produced by ^{13}C bombardment and thus could not possibly be due to element 105. Quite understandably we threw out the 9.1-MeV peak also since it was only marginally detected. We assumed that the 9.1-MeV activity was due to some artifact and temporarily abandoned the search for element 105 in order to find out more about the isotopes of lawrencium and rutherfordium. We had not yet eliminated all of the "bugs" from our new vertical-wheel system and thus had not yet built up a high degree of confidence in its ability to ferret out rare activities. By early 1970 we had discovered that the 8.9-MeV peak was due to 0.6-sec ^{257}Lr and with a vastly improved detection system once more we set out to find element 105.

On March 20, 1970 the experiment was begun again and within a few hours we knew that we had succeeded (GHIORSO et al., 1970) once more for we were seeing alpha counts at 9.1-MeV decaying with a half-life of a second or two. We realized somewhat sheepishly that this was the same peak that had been observed more than a year earlier! In rather short order we found alpha groups at 9.06, 9.10, and 9.14 MeV with a half-life of 1.6 ± 0.3 secs and were able to establish beyond a reasonable doubt the atomic number of the new nuclide. (See Fig. 11) The mother-daughter recoil-milking experiment in this case showed that the 35-sec ^{256}Lr was transferred to the mother crystals and the amount thus observed at each detecting station decreased with the same half-life as the 9.1-MeV alpha activity. In addition to this rather good proof, we tried a time-correlation technique which promises to be important for the detection of even heavier elements.

The seven detecting stations were arranged at 45° intervals around the wheel so that only eight collection spots were produced. This was done in order to measure a time correlation between the emission of a mother alpha particle and that of its daughter. Thus, when an atom of element 105 emits an alpha particle which is detected by one of the mother crystals, it follows that the newborn element-103 daughter atom must be kicked into the surface of the wheel. If the spot on the wheel where this emission takes place is tracked by the computer, a watch can be made for the subsequent emission from that spot of an alpha particle from the daughter. If the amount of background activity is not too large this method can be very successful, so periodically new spots on the wheel are used automatically by advancing the wheel a few steps to reduce the buildup of longer-lived interfering activities. The technique when applied to the $^{260}105$ - ^{256}Lr case worked out beautifully and gave a further confirmation of our discovery. Since we felt that we had identified element 105 in a completely unambiguous fashion, the discovery team, which was the same as for element 104, suggested the name hahnium with the symbol Ha to honor the late OTTO HAHN, the discoverer of fission.

At about this same time the DUBNA group reported finding a 2-sec SF emitter which they thought could be due to element 105. Gas chromatography experiments of the same type used in their element-104 work were cited to show that the 2-sec activity behaved like Ta but their evidence was not conclusive. Our work shows that it cannot be due to ^{260}Ha since we find less than a 25% branching ratio for fission of that isotope. There is some possibility, however, that the SF activity is due to a branch decay of ^{261}Ha. In later experiments we were able to identify this nuclide as an 8.93-MeV 1.7 ± 0.8-sec alpha emitter. The daughter, ^{257}Lr, was found both by recoil-milking and time-correlation experiments. We take the position that even if the SF activity is due to a branch decay of ^{261}Ha this does not imply discovery of the element by the DUBNA group. It seems to us that the discoverer is the group that first proves to its scientific peers that it has indeed done so.

Very recently we have produced a third isotope of hahnium, ^{262}Ha. This was found to have an 8.45-MeV alpha energy and a half-life of about 42 seconds. Again its daughter, ^{258}Lr, was identified by recoil-milking and time-correlation techniques. No chemical experiments have yet been performed by the Berkeley group, but we hope to be able to do so with our automatic chemistry system when the SuperHILAC resumes operation late in 1971.

Future Heavy Elements

We believe that it will be possible to produce and identify alpha-emitting isotopes of elements 106 and 107. Our first choices for element 106 are to look for ^{261}106 and ^{263}106 by bombarding ^{249}Cf with ^{16}O and ^{18}O ions. The energies of these two isotopes is in the range of ~9 MeV and the half-lives are expected to be roughly a few seconds. The daughters and granddaughters are now know so that it should be possible in principle to identify a mother-daughter-granddaughter sequence of alpha emission and thus be entirely certain of the results. The main stumbling block is that of their production cross sections. If they do not undergo significant branch decay by spontaneous fission, they should be observable at a rate of one or two alpha counts per day.

For element 107 we anticipated that the best choice may either be ^{19}F on ^{249}Cf to produce $^{264}107$ or ^{16}O on ^{253}Es to produce $^{266}107$. Here the cross sections will be even smaller and the work will be very difficult indeed. It is conceivable that the best approach may be to use two or more targets in tandem through which the beam passes simultaneously so that the data rate can be increased significantly.

In February 1971 the DUBNA group reported finding a ca 2-sec SF activity by bombarding ^{243}Am with ^{26}Mg ions. Conceivably the activity could be due to element 107 but the experiment suffers from the same uncertainties found in the element-104 and element-105 work.

Superheavy Elements

Since 1964 a great deal of the theoretical work has been published in which there seems to be a consensus that there could well be an island-of-stability in a region of nuclides near neutron number 184 and proton numbers 114 and possibly 124 or 126. Although the predictions of half-lives for nuclides on this island vary enormously, it seems that there is an excellent chance that elements in this region can live long enough for detection. The problem is--how does one make such superheavy nuclei?

At the present time we believe that there is a possibility of reaching the island by bombardments of the heavy elements with superheavy ions. There have been a few unsuccessful attempts to produce suitable reactions using Ar, Zn, and Kr ions at Berkeley, Orsay, and Dubna, but the tests were not of the type that could be regarded as conclusive--new accelerator capability is vitally necessary. To this end the Berkeley HILAC was shut down in February 1971 for an extensive modification which will make it possible to accelerate any atom up to energies as high as 8.5 MeV/nucleon. The new machine, which we designate by the name SuperHILAC, will be operating by the end of 1971 and we should soon know whether the "magic" island exists.

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ILLUSTRATIONS

Fig. 1 Original mica absorption data showing presence of $^{242}96$ alpha particles in helium-irradiated ^{239}Pu.

Fig. 2 Original mica absorption data showing presence of $^{241}95$ alpha particles in deuteron - irradiated plutonium.

Fig. 3 Elution curve for element 100. From second chemical identification (March 1, 1953). Except for ^{249}Bk the detected activities are due to alpha particles. Resin used was Dowex-50 at 87°C with citrate elution.

Fig. 4 Elution data showing the chemical identification of element 101 by means of its daughter, the 2.7-hr SF-emitting ^{256}Fm. The ^{253}Es and ^{246}Cf activities are due to alpha particles.

Fig. 5 Double-recoil experiment used in the detection of ^{250}Fm as the alpha-decay daughter of $^{254}102$.

Fig. 6 Experimental setup of the first Dubna attempts to identify an alpha particle emitting isotope of element 102. The beam (4) of ^{16}O ions traverses a collimator plate (5) and energy-degrading foils (8) to knock the transmutation recoils out of a ^{241}Pu target (2) onto a metal ribbon (1) which then carries the atoms in front of nuclear emulsions (6). The emulsions serve as alpha particle track detectors; the range of each track gives a rough measure of the alpha-particle energy and the position gives an indication of the half life.

Fig. 7 Alpha-particle range distribution as detected by nuclear emulsions in the early U.S.S.R. experiments aimed at producing element 102 by the bombardment of ^{239}Pu with ^{16}O ions. The broad peak at ~50 microns is mostly due to 25-sec ^{211m}Po.

Fig. 8 Empirical correlation between spontaneous fission half-life and neutron number for even-even nuclides. Notice the sudden decreases beyond the 152-neutron subshell.

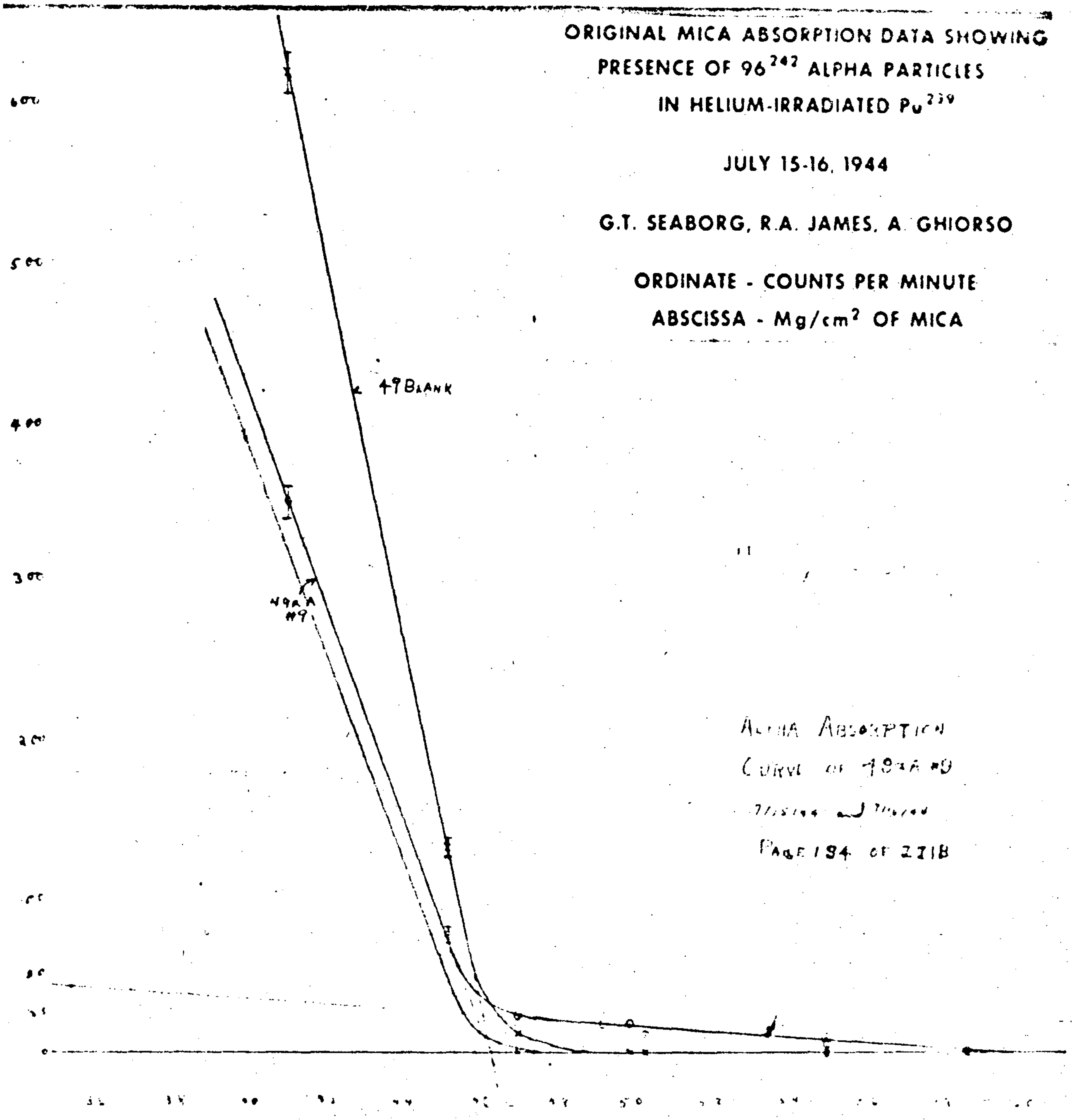
Fig. 9 Early 4-station system used for the detection of the 4.5-sec alpha-emitting isotope, $^{257}104$, as well as other heavy nuclides.

Fig. 10 Alpha spectrum of $^{257}104$. The lines at 8.70, 8.78, 8.95, and 9.00 MeV are due to this isotope. That at 8.87 MeV is due to 0.6-sec ^{257}Lr and 25-sec ^{211m}Po and that at 8.60 MeV is due to 4-sec ^{258}Lr.

ILLUSTRATIONS
(con't)

The spectra shown are from each detecting station around the wheel with their sum plotted topmost.

Fig. 11 Alpha spectrum of $^{260}105$. The lines at 9.06, 9.10, and 9.14 MeV are due to this isotope. The five-station system was used in these experiments and the individual spectra are shown with their sum plotted topmost.



ORIGINAL MICA ABSORPTION DATA SHOWING
PRESENCE OF 96^{242} ALPHA PARTICLES
IN HELIUM-IRRADIATED Pu^{239}

JULY 15-16, 1944

G.T. SEABORG, R.A. JAMES, A. GHIORSO

ORDINATE - COUNTS PER MINUTE
ABSCISSA - Mg/cm^2 OF MICA

ALPHA ABSORPTION
CURVE OF 49αA #9
7/15/44 and 7/16/44
PAGE 194 OF 221B

Figure 1

ORIGINAL MICA ABSORPTION DATA SHOWING PRESENCE OF 95^{241} ALPHA PARTICLES IN DEUTERON - IRRADIATED PLUTONIUM

OCTOBER 17–18, 1944

G.T. SEABORG, L.O. MORGAN, R.A. JAMES, AND A. GHIORSO

49 DD #31 - RANGE CURVE FOR 95^{241}
49 DD #32 - COMPARISON RANGE CURVE FOR Pu^{239} FRACTION

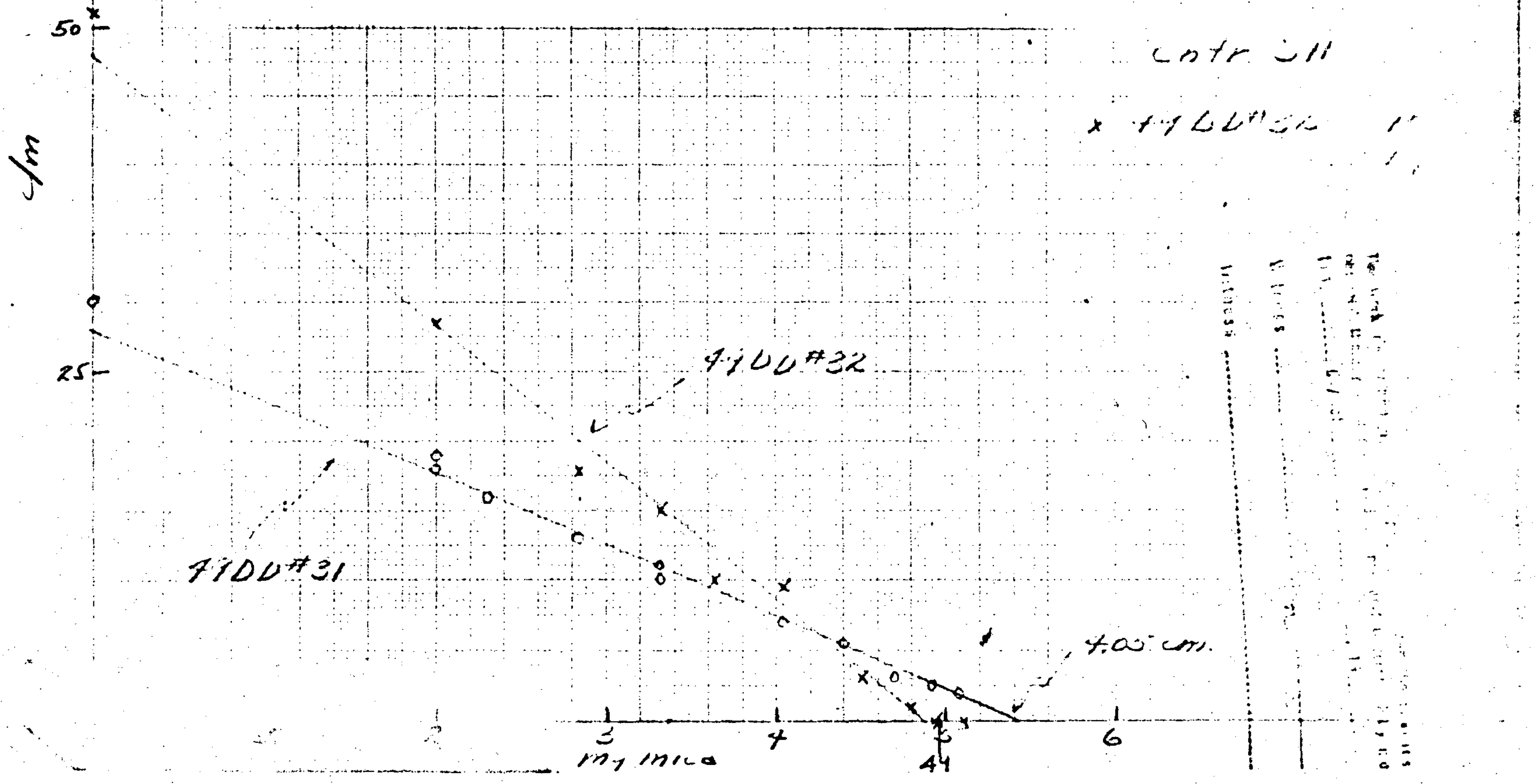


Figure 2

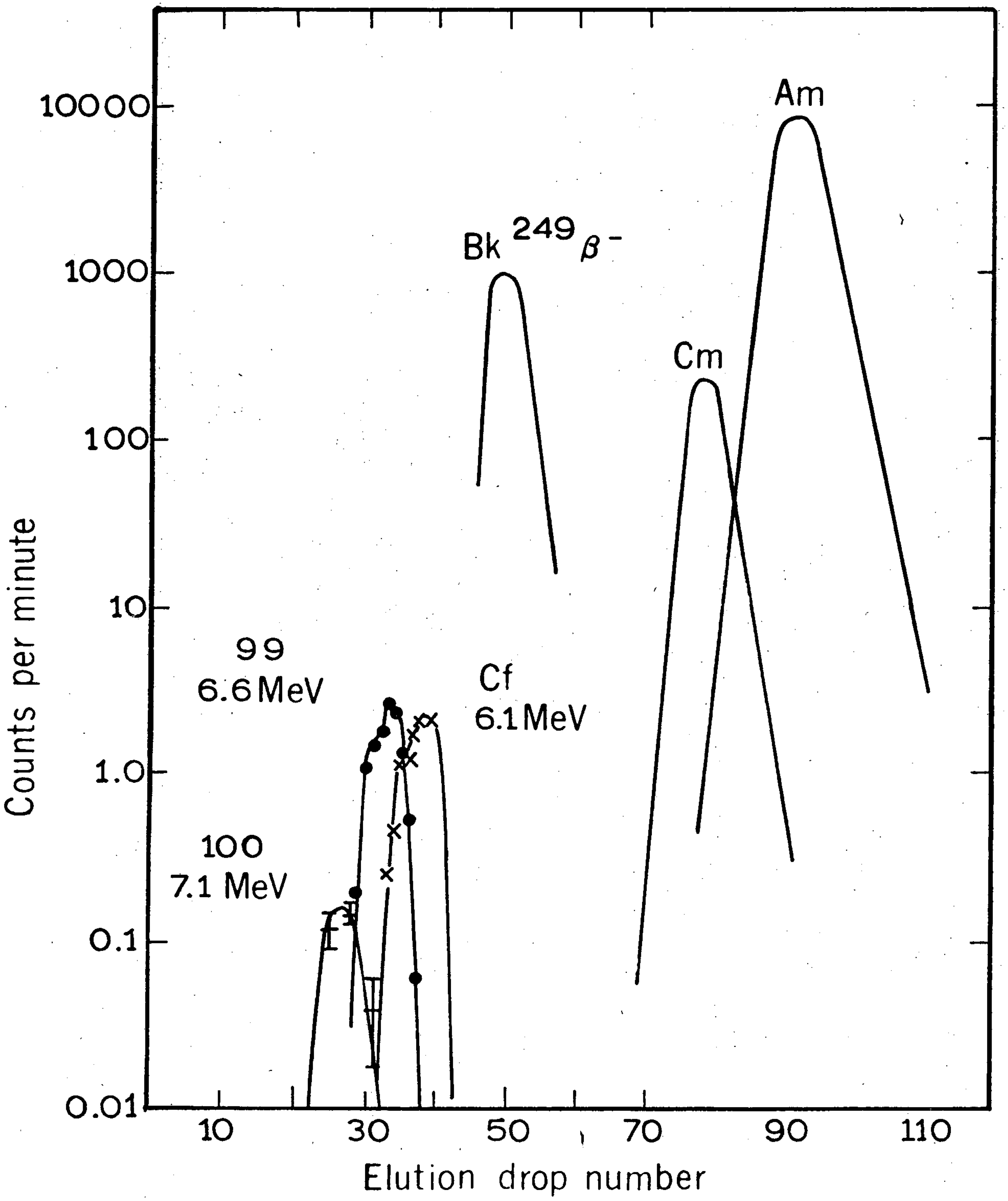


Figure 3

XBL 716 6360

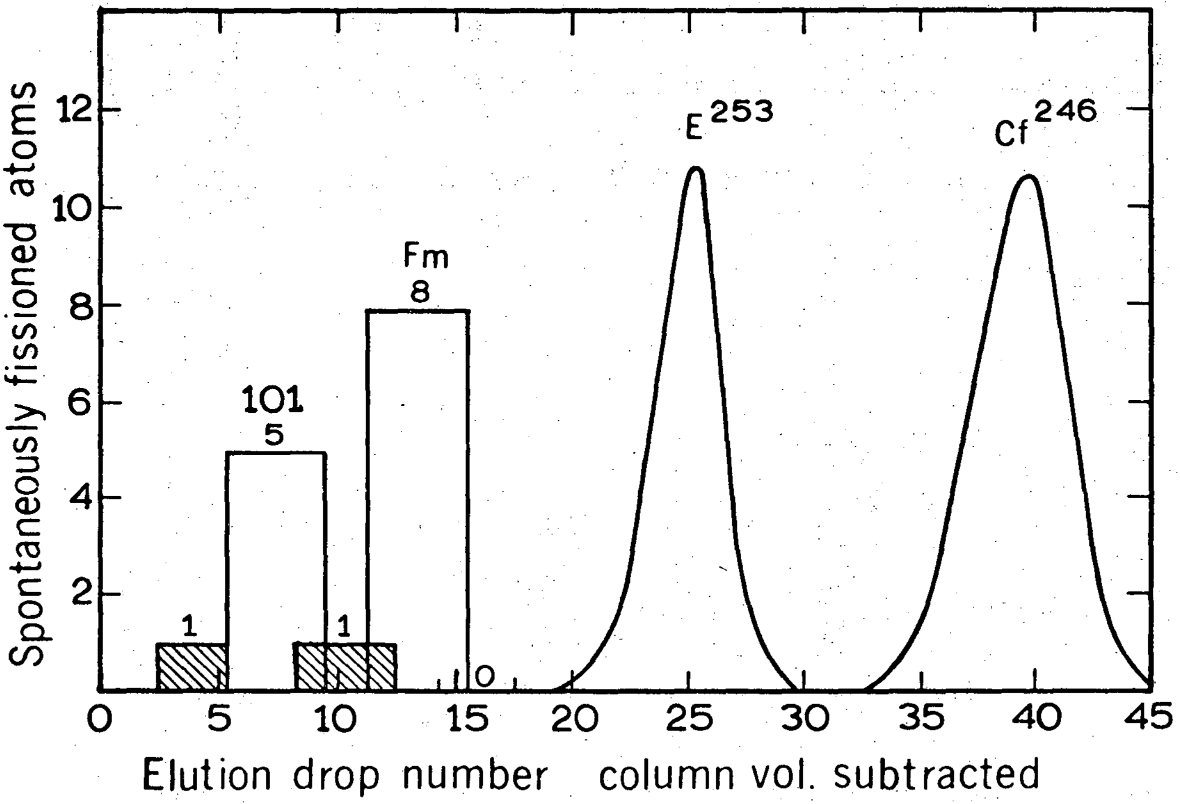


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Figure 4

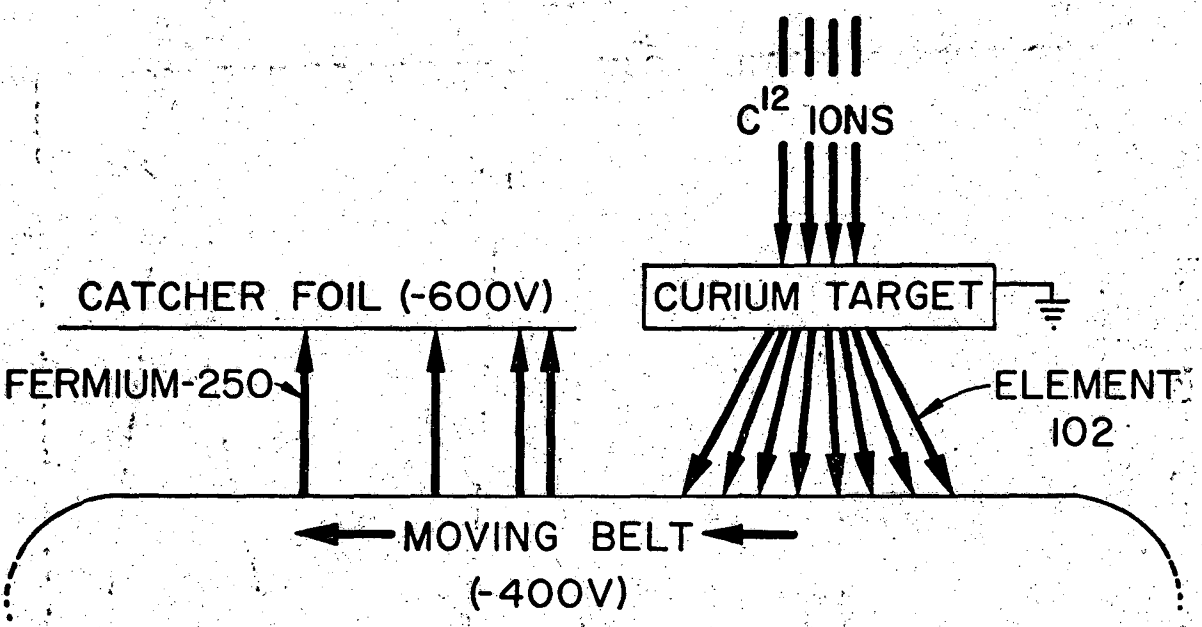
C^{12} IONS
CATCHER FOIL (-600V)
CURIUM TARGET
FERMIUM-250
ELEMENT 102
MOVING BELT
(-400V)

Figure 5

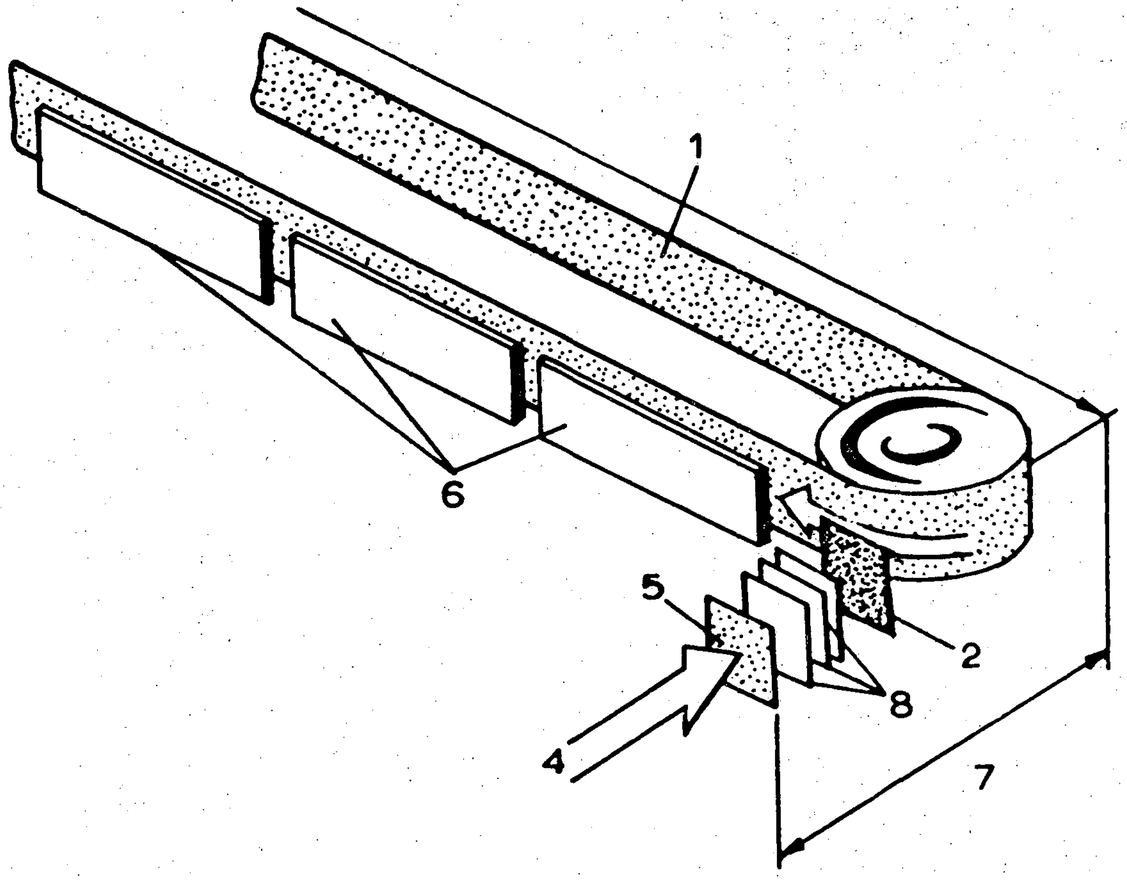

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Figure 6

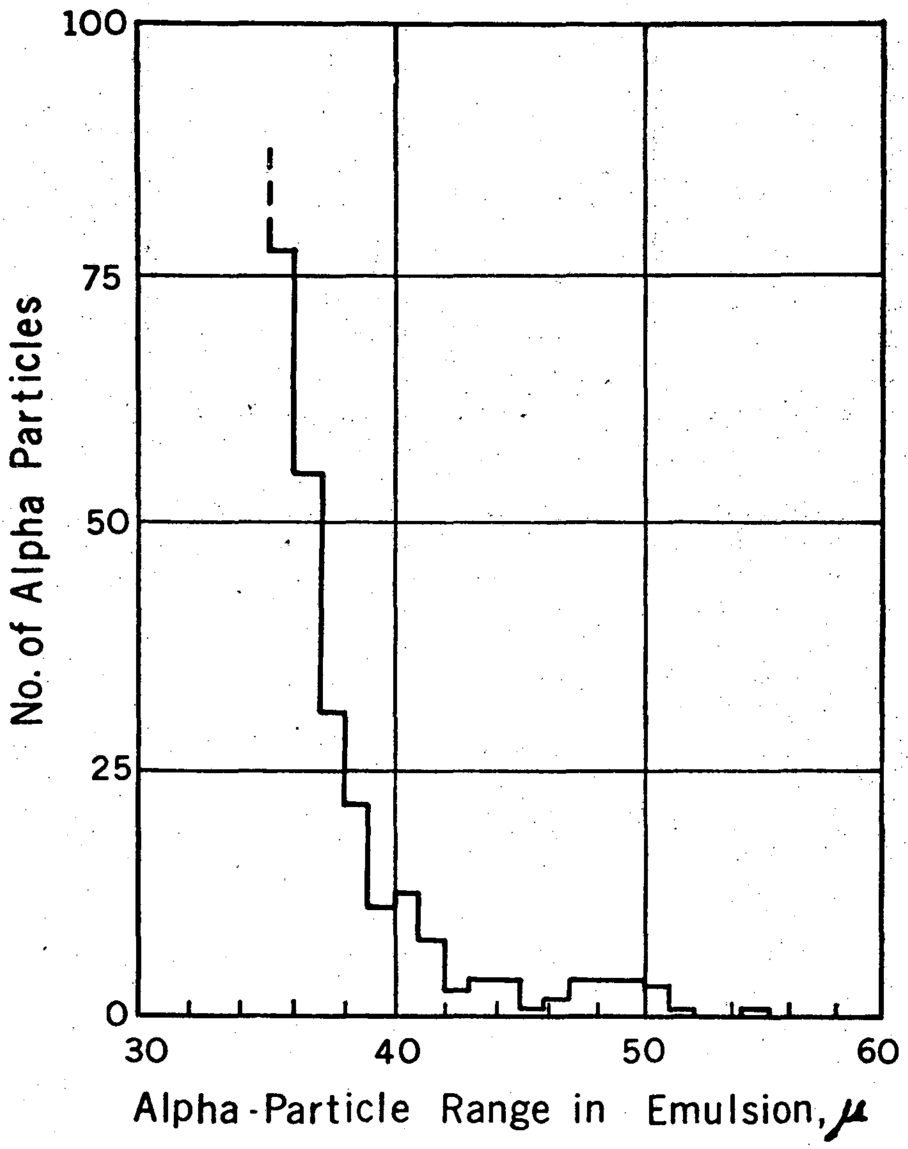

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Figure 7

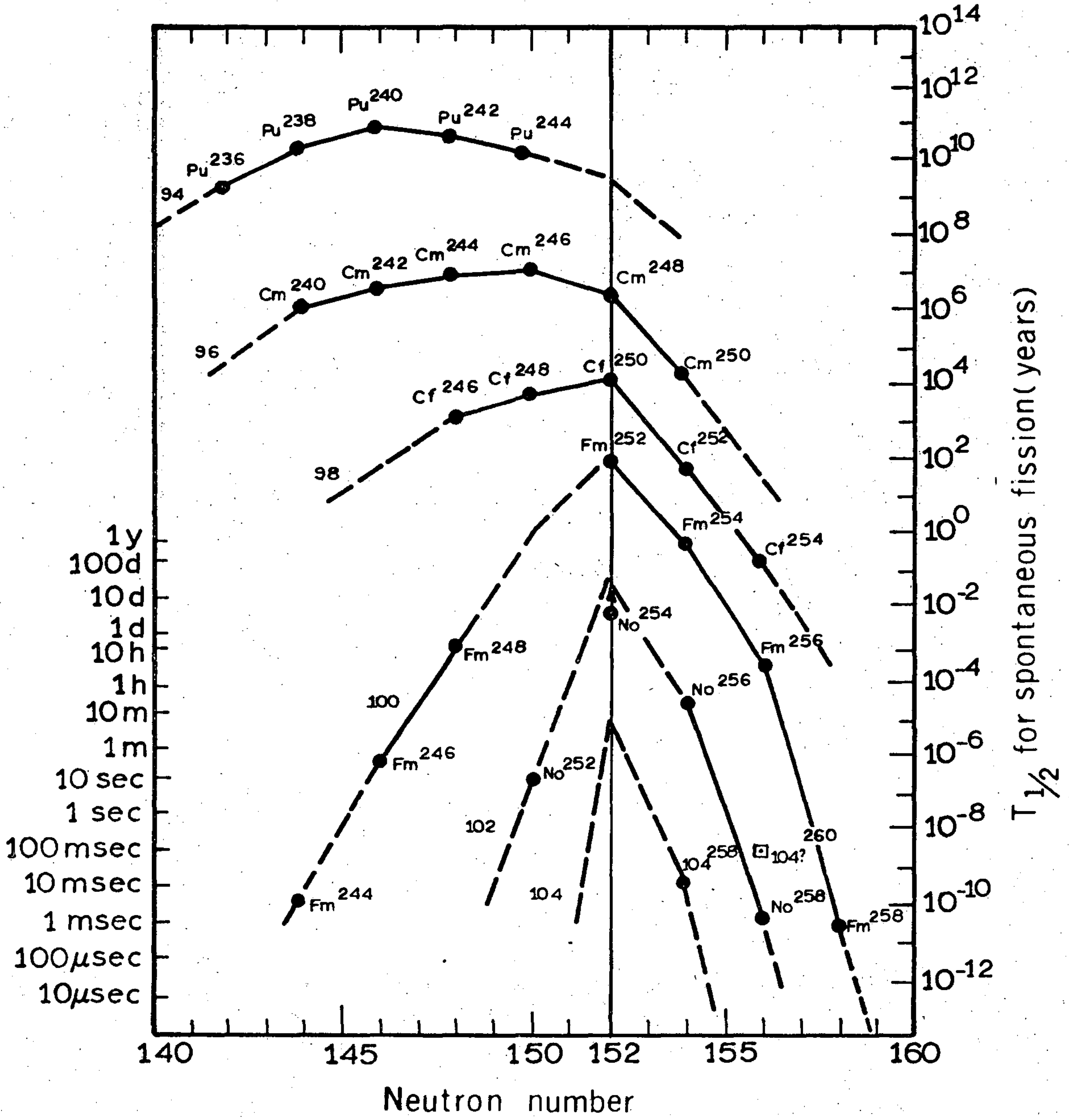

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Figure 8

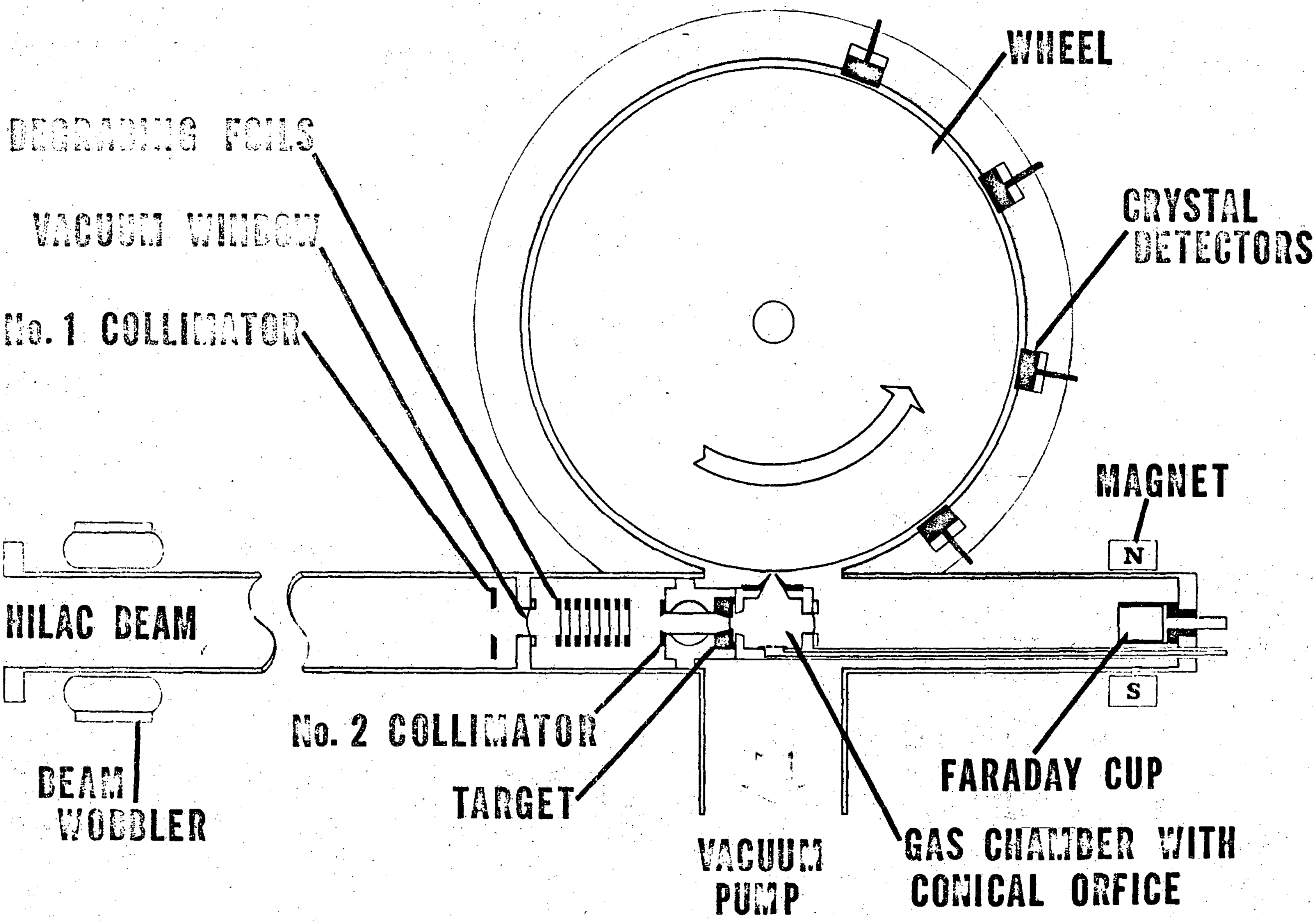
WHEEL
DEGRADING FOILS
VACUUM WINDOW
CRYSTAL DETECTORS
No. 1 COLLIMATOR
MAGNET
N
HILAC BEAM
S
No. 2 COLLIMATOR
FARADAY CUP
BEAM WOBBLER
TARGET
VACUUM PUMP
GAS CHAMBER WITH CONICAL ORFICE

Figure 9

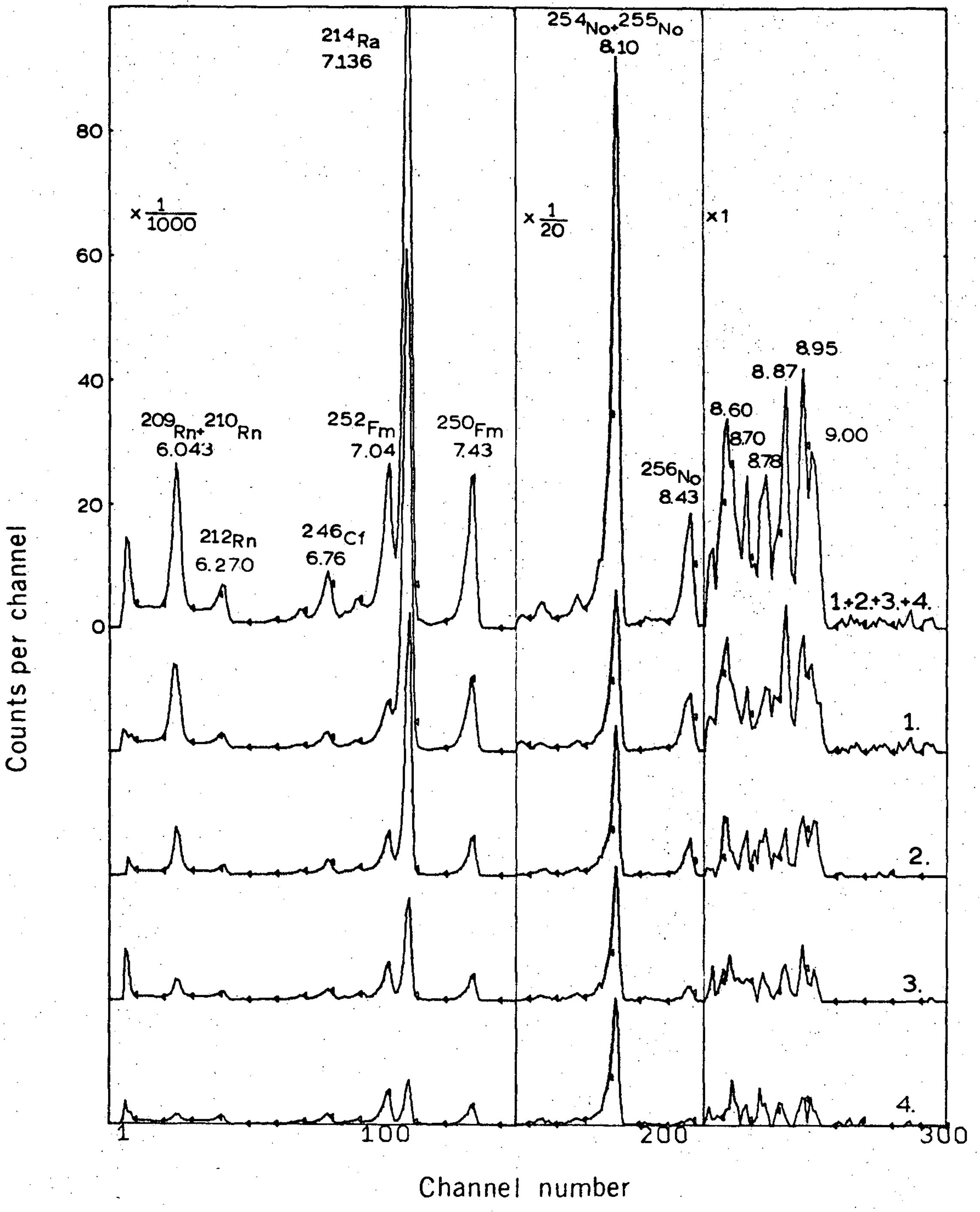

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Figure 10

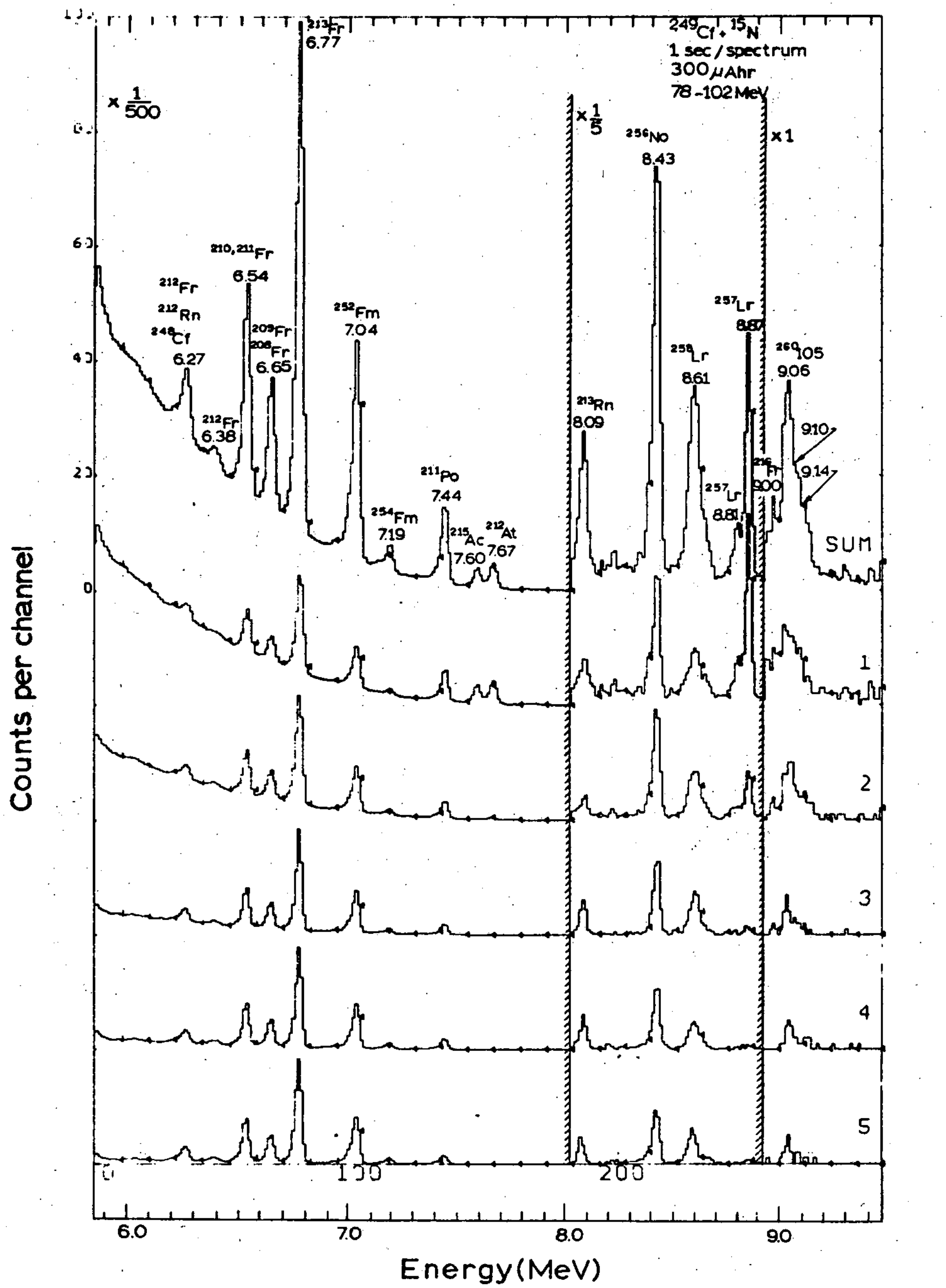

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Figure 11

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