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TWO NEW ELEMENTS--ATOMIC NUMBERS 104 AND 105

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TWO NEW ELEMENTS--ATOMIC NUMBERS 104 AND 105*

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In the last five years new materials and some very sophisticated techniques have become available for the preparation of new elements that go beyond the so-called actinide series, a Rare-Earth-like chemical series of elements ending with number 103, lawrencium. The latter element was discovered in 1961 at Berkeley and was the culmination of research which spanned a considerable period of time. Seven more years were to elapse before the next step was to be completed but now there is good reason to believe that within the next year elements with atomic number as high as 107 may be observed. This article will attempt to show what has happened to make these new extensions of the periodic table possible. Associated with the author in the research to be described are Matti Nurmia and James Harris of the Lawrence Radiation Laboratory and Kari and Pirkko Eskola, a husband and wife team on leave from the University of Helsinki in Finland.

The elements beyond uranium have not survived on Earth because the half-lives of their isotopic constituents are all too short. They can however be made by successive neutron-capture reactions in high-flux reactors and by charged-particle reactions with heavy-ion accelerators. The neutron reactor method is very efficient and is able to supply the transuranium elements in quantities ranging from thousands of tons of an element like plutonium, element 94, to 10^8 atoms of ^{257}Fm (the heaviest-known isotope of fermium, element 100). At atomic number 100, however, there is an almost impassable barrier in the

*Work performed under the auspices of the U. S. Atomic Energy Commission.

form of a very short-lived isotope, ^{258}Fm, which decays by spontaneous fission before it can capture neutrons, so that there the neutron-buildup process comes to an end. Fortunately the heavy-ion accelerator technique is not hampered by such a blockage. Though it is not possible to produce large quantities of the heavier elements in this manner they can be formed, but with a rapidly decreasing probability as the atomic number is increased. It is not intended in this article to discuss the very real possibility of reaching a hypothetical "island-of-stability" in the region of atomic number 114, but the techniques to be described will also be applicable to such a study.

The difference between the neutron-buildup and the heavy-ion approaches in forming new elements can be indicated in the following way. In the first case in going from a target element such as plutonium to a final product such as fermium a considerable amount of time must elapse. This is because the target material undergoes a long series of neutron-captures and beta-decays until a small portion of the original material has been transmuted into a neutron-rich isotope of the desired element.. In the accelerator approach, the desired number of additional protons can be added at once within certain narrow restrictions. The yield in this case is extremely small and the product is usually a neutron-deficient isotope. To produce elements with atomic number greater than 100, we have found it advantageous to combine the two methods. The U.S. has a large, efficient reactor program for the production of materials up to einsteinium in useful quantities. When these heavier elements are then bombarded with heavy ions of carbon, nitrogen, or oxygen, it has been possible to identify elements up to element 105. Completed in 1957, the Berkeley machine used for this work is the Heavy Ion Linear Accelerator (HILAC).

Since the new elements are short-lived, it is not feasible to separate the transmutation products from the target material by ordinary chemical techniques. Two decades ago we found it expedient to take advantage of the inherent large recoil energy which is imparted to the product when it is formed by the amalgamation of a target atom with an impinging heavy ion traveling at a speed roughly 10% that of light. This energy is large enough to enable the newly-born atom to be knocked completely out of a target and thus make it available for measurement of its nuclear or chemical properties free from the interference of the properties of the target material.

In 1964 the first attempts to detect atoms of element 104 were made at Dubna, an international nuclear science center near Moscow in the Soviet Union. G. N. Flerov and his co-workers bombarded a target of ^{242}Pu with ^{22}Ne ions, using the large 310 cm cyclotron. The transmutation products were caught in an adjacent moving metal ribbon as they recoiled out of the target. The continuous "conveyor belt" passed next to sheets of glass which were there to act as detectors of spontaneous fission disintegration of element 104. The invisible tracks from the passage of fission fragments can be made optically visible by preferential etching with sodium hydroxide. They found an activity which seemed to have a half-life of 0.3 second. Since the number of tracks that they could produce was small (1 per 5 hours) and the interfering background was not negligible, they were not certain of the meaning of their results but thought that the activity could be due to $^{260}104$.

In order to check whether it was indeed due to such a new element, they devised a chemical experiment which made use of the prediction that element 104 should behave like hafnium and thus form a very volatile chloride. The

recoil atoms were caught in a gaseous chloride environment, passed through a filter at an elevated temperature, and then detected with glass sheets as before. With an even smaller yield they observed tracks which they felt could be due to element 104 and published their work as a claim to the discovery of this element. They suggested the name kurchatovium to honor the memory of Igor Kurchatov, a pioneering Soviet nuclear physicist.

From the beginning our group felt that there was some doubt that these results demonstrated the existence of element 104. Our feeling was based on the systematic behavior in half-life of spontaneous fission decay that we had observed for the elements of lower atomic number. We thought that this half-life should be orders of magnitude shorter in the case of $^{260}104$ but that possibly the isotope in question could be of mass 261.

Although we felt that there was a possibility that the Dubna work was correct we decided to reserve judgment and continue in our efforts to find an alpha-particle emitter which could be assigned without ambiguity to element 104. 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 energy and half-life can be uniquely identified, whereas one that disintegrates by undergoing spontaneous fission provides little in the way of a fingerprint. This is because fission does not differ greatly in characteristics from one nuclide to another. Only the half-life can be used to separate one spontaneous fission activity from another when chemical means are not readily available.

Early in 1968 enough ^{249}Cf became available to allow us to make an attempt to find an isotope of element 104. ^{249}Cf is a 360-year alpha emitter, the daughter by beta-particle decay of 314-day ^{249}Bk. This berkelium isotope is

an important product in the long chain of neutron-captures and beta-decays mentioned earlier. By chemically isolating berkelium, a pure sample of californium was obtained after a suitable decay period and we now had available about 100 micrograms of the material. This was just the beginning, however, for there are many problems posed by the target-making process. The highly radioactive target has to be several hundred micrograms per cm^2 in thickness to give an optimum recoil yield when element 104 is made with carbon ions and has to be free of such impurities as lead and bismuth since these will produce interfering activities. It has to withstand an intense beam of ions (typically 2×10^{13} ions per second per cm^2) for many weeks and be mechanically sturdy. We had a very bad experience in 1959 when a curium target was mechanically ruptured and contaminated the entire HILAC building and we did not want to take any chances.

The final method, developed over a period of many months, was so successful that we have used the same ^{249}Cf target to make both elements 104 and 105. It has survived many thousands of microamperehours of concentrated bombardment over a period which spans some 18 months and has not deteriorated so far as we can tell. The target was electrodeposited from an isopropyl alcohol solution onto a thin palladium coating which had been sputtered upon a 2.5 mg/cm^2 beryllium foil. The beryllium provided very good thermal conductivity and excellent strength while the palladium surface provided an inert base for the electroplating. After the californium was baked in an oven at several hundred degrees centigrade a thin coating of aluminum was evaporated upon it to keep the heavy ion beam from ejecting small quantities of the target itself and thus gradually contaminating the target chamber. Unfortunately, when this particular target was made, we had not yet mastered the techniques necessary to prevent all lead contamination from appearing in the final product and consequently

there was present about 10 nanograms of lead with the 60 micrograms of californium. There were also a few nanograms of mercury which were added when the palladium was sputtered onto the beryllium.

With a suitable target and an adequate heavy-ion beam current, new atoms can be produced and detected. It must be noted, however, that the detection of a new alpha-particle activity is considerably more difficult than the detection of a new spontaneous-fission activity because heavy-ion transmuted recoil atoms, which are stopped in a solid, are usually buried so deeply that alpha-particle energy analysis is rendered impossible, whereas it is only necessary to detect fission fragments. In our laboratory some years earlier it was discovered that it was possible to transfer these atoms to a collecting surface efficiently and quickly so as to form a weightless sample for analysis. This was done by absorbing the recoil energy in a helium atmosphere and then carrying the atoms along in the stream of gas which flowed through a small orifice into a rough vacuum. The jet of helium impinged upon a collecting surface a few millimeters away and a very high fraction of the recoil atoms were found to adsorb upon the surface. This technique had been used successfully by both United States and Soviet scientists in studies of the isotopes of nobelium and lawrencium, elements 102 and 103. The usual apparatus involved the use of a flanged wheel to act as a conveyor to carry the transmutation products to alpha particle detectors for analysis.

Since the nuclides beyond lawrencium were predicted to have half-lives in the millisecond range, we developed a new technique that was not limited by the speed with which we could rotate a wheel. We found that the helium (or hydrogen) gas stream could carry the atoms very efficiently through small diameter tubing for distances of many centimeters. Since the velocities reached

were in the sonic range unusual effects were observed. We found that by flaring out the exit end of the tubing as in a Venturi orifice, a small ring a few millimeters in diameter was formed on a flat collecting surface held a centimeter or two away from the exit. By this I mean that the entrained heavy atoms in the gas stream could be collected in a ring pattern as demonstrated by autoradiography. In theory this meant that we could place a solid-state detector in this position and receive the recoil products directly on the face of a crystal to be analyzed for their alpha radioactivity. We did actually succeed in accomplishing this feat but the basic technical difficulties were too great to allow the method to be very stable.

While the system was working, we examined the alpha particle spectrum produced by ^{12}C ion bombardment of our new ^{249}Cf target (Fig.). We were surprised to find new alpha particle peaks at 8.7 and 9.1 MeV which decayed with a half-life in the range of seconds rather than milliseconds. By carefully comparing the spectrum produced by a lead target, we decided that the new alpha peaks were produced by a $^{12}C,4n$ reaction and thus must be due to an isotope of element 104 with mass 257. Since now we knew that we did not have to use a high speed system to detect the new activity, we decided to build an elaborate multi-detector system in which a large wheel would be the atom transporter. This would automatically solve the problem which was basic to the Venturi method--the buildup on the detecting crystal of long-lived activities--to say nothing of the continual problems of detector damage.

Although the "conventional" wheel system is not plagued by the problems that beset us with our Venturi device, it does have complexities which have to be handled carefully. The method lends itself nicely to the use of many detectors for the purpose of taking much data simultaneously (our latest system uses 28

crystals and a new one that we are designing will use 3 x 28 !). Since the number of atoms per hour that can be made is limited, it is imperative that a large number of characteristics be measured simultaneously. The net result is that a very large amount of electronic wizardry must be fabricated and kept operating and, because of the many parameters involved, a fair-sized computer must be programmed to keep track of all of the information.

It took many months to locate and eradicate all of the inevitable "bugs" that developed in the new vertical wheel system, but finally this was accomplished and we were ready to perform meaningful experiments. We were delighted to find that the 20-detector system performed its functions with great precision and stability. The energy resolution attained was considerably better than with our old wheel system and the spurious backgrounds observed were usually negligible. The reproducibility of a given experimental measurement was now excellent and it was possible to run the system for days at a time with no more than minimal attention. Now it was possible to detect new activities at counting rates as low as a few per day if necessary.

In the case of element 104 this was not necessary. With the new system we again observed the unknown alpha particle peaks along with several other new groups at a counting rate of 30 counts per hour! This was no longer a marginal experiment--we were able to do alpha spectroscopy. The increased sensitivity and discrimination of the vertical wheel (VW) system was needed badly for it turned out that the spectral lines from $^{257}104$ alpha particle decay were very complex (see Fig.). Since the decay was spread among many peaks of comparable height, it meant that the height of any one of them was correspondingly reduced. However, we were very gratified with the results and

we found that when the computer had normalized the amplifier gains (which varied a little between amplifiers and from one run to another), the composite spectrum was very clear. The half-life for $^{257}104$ turned out to be 4.5 ± 1 seconds.

With the confidence gained from this experiment we proceeded to look for $^{259}104$ by using ^{13}C ions, hoping that we would be able to detect its production via a $^{13}C, 3n$ reaction. We were rewarded to find a different activity with a somewhat shorter half-life--groups of alpha particles at 8.77 and 8.86 MeV decaying with a half-life of about 3 seconds. The yield from this reaction was substantially less but it was confirmed somewhat later when we bombarded a ^{248}Cm target with ^{16}O ions.

The important parameter to be measured in these cases was the atomic number to show that we had identified a new element. This we proceeded to do by developing a very sensitive technique for identifying the element-102 daughters of these alpha emitters. When an atom emits an alpha particle with a given momentum, there is an equal momentum in the opposite direction given to the residual nucleus which has been left behind. This energy is much more than sufficient to release the atom from the surface on which it rests if the alpha particle is emitted into the surface. Since the alpha particles are emitted with an equal probability in all directions, this means that half of the atoms on the surface of the wheel will be released and half will be embedded; thus, we should expect that there will be as many nobelium-daughter atoms recoiling into the detectors as there are element-104 alpha particles detected. To make use of this genetic relationship, we arranged to periodically shuttle these detectors away from the wheel a short distance so that we could measure the properties of the daughter atoms. To increase the geometry for this measurement to almost 100%, the detectors were moved to positions opposite other identical

detectors. Finally, to avoid the loss of detection of element-104 atoms while measuring the recoil daughters we duplicated the system so that we could measure the mothers and daughters concurrently. A drawing of the geometry of each detecting station is shown in Fig. .

With this recoil-milking system we were able to show that a known isotope of element 102, ^{253}No, is recoiled off the wheel by the alpha decay of its parent, the 4.5-second ^{257}104. Both the alpha energy and the half-life of the nobelium daughter were measured in the daughter-detecting mode. Similarly, in other experiments we showed the presence of ^{255}No resulting from ^{259}104 decay.

By this time we realized that the half-lives of the element-104 isotopes that we could produce with the HILAC were substantially longer than expected. We now predicted that ^{261}104 should have an alpha half-life of about a minute if it did not decay much more quickly by spontaneous fission as suggested by the Russian experiments. Our calculations showed that the best way to produce it was by bombardment of our ^{248}Cm target with ^{18}O ions, via a compound-nucleus reaction wherein five neutrons are emitted.

At first when we initiated this experiment, we were puzzled and disappointed because we did not observe a new alpha peak. The only heavy element peak that we saw clearly was that of 8.3-MeV ^{257}No, which is known to have a half-life of 26 seconds. After we had accumulated enough statistics, though, we saw that the half-life of this familiar broad peak was longer than 26 seconds, in fact, it appeared to be about a minute. Then we knew that ^{261}104 emitted alpha particles with energy very close to those of its daughter--a somewhat surprising finding. Mother-daughter measurements proved that this was the case and showed that the half-life was 65 seconds. This was the third new alpha emitter of element 104 that we had found and, of course, it also showed that the Russian

0.3-second activity could not be $^{261}104$.

The finding of the 65-sec $^{261}104$ brought the possibility of doing certain kinds of aqueous chemistry experiments which would be important to demonstrate that element 104 was indeed quite different from the actinide elements. We decided to undertake the task even though we knew that on the average at best we would be dealing with only one atom in every five experiments. In this work we were joined by Robert Silva, a former colleague of ours now at the Oak Ridge National Laboratory. He had worked with us in earlier fast experiments on the chemistry of lawrencium but now we had an order of magnitude less activity for this purpose.

Element 104 long ago was predicted by Glenn Seaborg to have the chemical behavior of hafnium. This meant that it would have four valence electrons and a similar ionic radius and thus should lend itself readily to a fast simple separation from the normally-trivalent actinide elements. The method that we chose was that of the cation-exchange column. It was predicted that element 104 would not be adsorbed readily by the resin and that, using an alpha-hydroxy-isobutyrate eluant, we could wash the eka-hafnium atoms straight through the column. Under the conditions to be used, the first actinide element that would be eluted from the column would be lawrencium, and it would not appear until perhaps a hundred column volumes of wash had gone through.

The target chamber system used the same principle as the VW apparatus except that the atoms of element 104 were caught on the bottom of a pneumatic rabbit. The procedure was to make a bombardment of a couple of minutes duration with ^{18}O ions on ^{248}Cm, shoot the rabbit quickly to the chemistry area, dissolve the atoms from the surface of the rabbit, force this solution through a short cation column, quickly evaporate the first few drops that

passed through, and, finally, analyze these drops for their alpha activity by means of solid-state detectors. This process had to be finished quickly because of the short half-life and had to be repeated hundreds of times to have enough atoms separated to be statistically meaningful. On the average because of the time taken for the chemistry (approximately 50 sec) and the inevitable mechanical losses that the fast process entailed, we observed only one atom in every twenty experiments. But this was enough! The total number of events observed in the early elution position was seventeen and conclusively demonstrated that element 104 did not belong to the actinide family.

While the search for alpha emitters was being pursued, we also were doing different experiments which were sensitive only to spontaneous-fission decay with the hope that we could either confirm or deny the Dubna claim to discovery of a 0.3-second element-104 emitter. Instead of using an endless conveyor belt to carry the embedded atoms past the detecting material, we used a drum. In order to reduce the background effect of longer-lived fission-emitters, such as 2.7-hr ^{256}Fm, which could be produced at the same time as element 104, the drum was moved slowly along its axis as well as rotated swiftly. The detecting material used in our case was usually mica. With this apparatus (Fig.) we achieved a very high sensitivity and could detect spontaneous-fission half-lives as short as 100 microseconds. It was tried out with the ^{249}Cf target and we were pleased to find a new activity with a half-life of 11 milliseconds which we thought was probably due to ^{258}104 made by means of a ^{12}C,3n reaction. When we used ^{13}C ions we again saw the same spontaneous fission period, but this time it was produced at a somewhat higher bombarding energy and with a higher yield as would be expected if the

activity were made by a $^{13}C,4n$ reaction. Finding an 11 millisecond half-life for $^{258}104$ corresponded very well with our predictions for that nuclide and thus reinforced our scepticism that $^{260}104$ could have a 0.3-second period.

All our attempts to find the Dubna fission activity have so far been unsuccessful. We have made bombardments with ^{16}O and ^{18}O ions of ^{248}Cm and ^{246}Cm targets which should have produced amounts of $^{260}104$ that we could readily detect if it had this half-life. We have also bombarded ^{253}Es with ^{11}B ions without observing such an activity but here our sensitivity was somewhat marginal because of the small amount of target material available. After we published our work on the element-104 alpha emitters, the Dubna group repeated their measurements on the short-lived fission emitter and came to the conclusion that its half-life might be closer to 0.1 than to 0.3 second. Since it was possible that we might have missed it in our first experiments which were designed for the longer half-life, we repeated the measurements and again we were unsuccessful in finding the activity. We now believe that $^{260}104$ will be found to have a spontaneous fission half-life in the microsecond range and plan experiments to find this missing nuclide.

In view of the preceding observations we decided to assert the traditional right granted to the discoverers of a new element to propose a suitable name. This was done in November 1969 at the Welch Foundation Conference on Transuranium Elements--The Mendeleev Centennial held in Houston, Texas. In honor of Ernest Rutherford, the great pioneer of nuclear physics, we suggested that element 104 be called rutherfordium with the symbol Rf.

With four isotopes of rutherfordium identified we felt great confidence that we should be able to detect an isotope of element 105. From the

alpha energy information that we had obtained for element 104 we could predict with fair accuracy what might be expected for the next higher element. Using the same ^{249}Cf target it seemed as if we should be able to observe ^{260}105 by using ^{15}N ions--it should have an alpha energy somewhat higher than that of ^{257}104 and a half-life in the range of seconds with a production cross section only a little smaller than that for making ^{257}Rf.

On March 20, 1970 we set up the experiment and within a few hours we knew that again we had succeeded 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 we had already seen this alpha peak more than a year before. At that time the same experiment had been tried after finding the ^{257}Rf activity. We had observed a new 8.87-MeV alpha peak with a half-life of 0.7 second together with a very tiny group at about 9.1 MeV and it seemed possible that both peaks could be due to element 105. However, within a few days we had dismissed this possibility for we found that the 8.87-MeV activity could also be made by ^{13}C ion bombardment of ^{249}Cf and thus could not possibly be due to an element with atomic number greater than 104. Quite naturally we also threw out the at-that-time marginal 9.1-MeV peak. Now, a year later, with a vastly improved technology and a much better understanding of the problem we could readily identify this persistent peak as coming from the isotope ^{260}105.

In rather short order we were able to establish beyond a reasonable doubt the atomic number of the new nuclide. The mother-daughter recoil-milking experiment in this case showed that a known isotope of lawrencium, 30-second ^{256}Lr, was transferred by alpha-particle recoil to the mother crystals and that 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 proof we tried a new technique which promises to be important in detecting elements 106 and 107.

When an atom of element 105 emits an alpha particle which is detected by one of the mother crystals, it is obvious that the newborn atom of its element-103 daughter must be kicked into the surface of the wheel. If we now wait a suitable period of time, this daughter atom will also emit an alpha particle and if it is emitted out of the wheel we can detect it too. This method of looking for delayed alpha-alpha coincidences was successfully applied to the $^{260}105 \longrightarrow {}^{256}Lr$ case thus giving a further proof of the genetic relationship.

As in the case of element 104, rutherfordium, there were also prior claims to the discovery of element 105. The Dubna experimenters under G. N. Flerov had bombarded ^{243}Am with ^{22}Ne and found a miniscule number of alpha counts which they attributed to the isotopes $^{260}105$ and $^{261}105$. In a similar time correlation experiment to that described above they looked for delayed coincidences of alpha particles 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.

The Dubna conclusions depended for their validity on the assumption that the 8.35 to 8.6 MeV window in the alpha spectrum included the emissions from both ^{256}Lr and ^{257}Lr and that their half-lives were also both the same, about 30 seconds. Now we know that ^{257}Lr has quite different characteristics and thus could not have been detected in their experiment as the daughter of

$^{261}105$. It has turned out that the 8.87-MeV 0.7-second activity which we thought in 1968 might be due to element 105 is actually ^{257}Lr. Since our work with $^{260}105$ shows it to have a completely different energy than their 9.4 or 9.7 MeV alphas, this, too, can be excluded as being detected in their experiments. They have more recently found a spontaneous-fission activity with a half-life of about 2 seconds which they believe might be due to element 105. It might equally well be due to a small electron-capture branch of ^{258}Lr to ^{258}No which is known to have a millisecond fission half-life.

Since we felt that we had established the existence of element 105 beyond question, we again decided to assert our right as discoverers. In honor of the late Otto Hahn, the famous pioneer and discoverer of fission, we suggested that the new element be called hahnium and be given the symbol Ha.

We hope soon to find a longer-lived isotope of hahnium by bombarding ^{253}Es with ^{13}C ions to make ^{262}Ha. We would predict that this nuclide might have a period in the range of a minute just as in the case of ^{261}Rf. If this proves to be true, we should be able to do an aqueous chemistry experiment to see if, as expected, element 105 has an eka-tantalum bahavior.

Further jumps in atomic number are possible but depend entirely on whether or not spontaneous-fission decay takes precedence over alpha decay. If the fission half-lives do not become too short, we calculate that with great effort we should be able to produce and detect the nuclides $^{261}106$ and $^{263}106$. Since the daughters and granddaughters in both cases are now well-known, we should be able to identify them unambiguously. Element 107 is also within reach since it should be hindered even more against fission decay, but the problem in this case is that the production cross section is substantially

smaller. We are not likely to detect anything beyond element 107 unless the predicted doubly-magic region of 114 protons and 184 neutrons exists. Next year we will make an attempt to reach this region by bombardments with exotic ions all the way up to uranium. For that purpose we are in the process of modifying our accelerator into the SuperHilac--but that is another story.

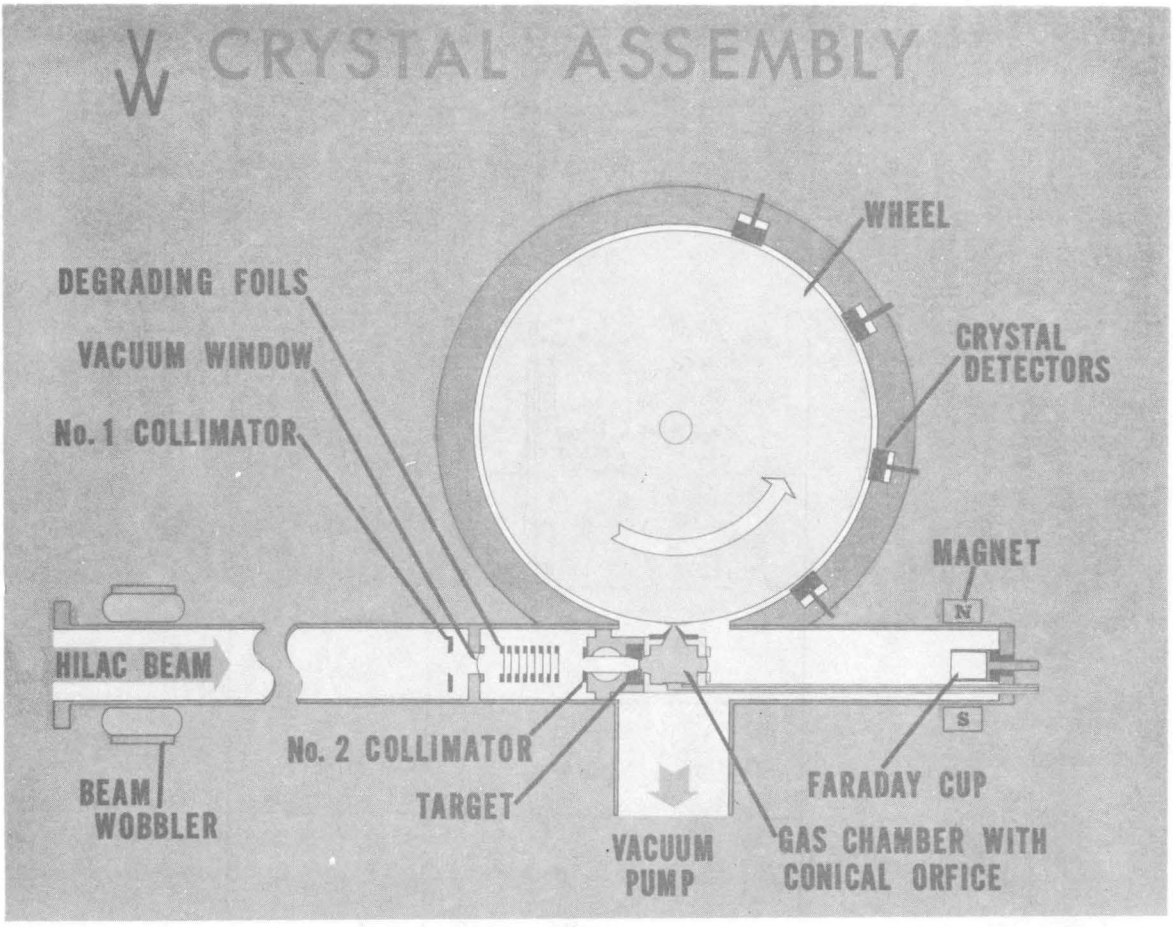
FIGURES

Fig. 1. Schematic diagram of the vertical wheel detection system used in the discovery of element 104 alpha radioactivities. At that time there were only four stations. There are now seven.

Fig. 2. A recent measurement of the ^{257}Rf alpha spectrum obtained by using our latest 7-detector station system. The activities are those detected by the crystals when they are in the "on-wheel" position.

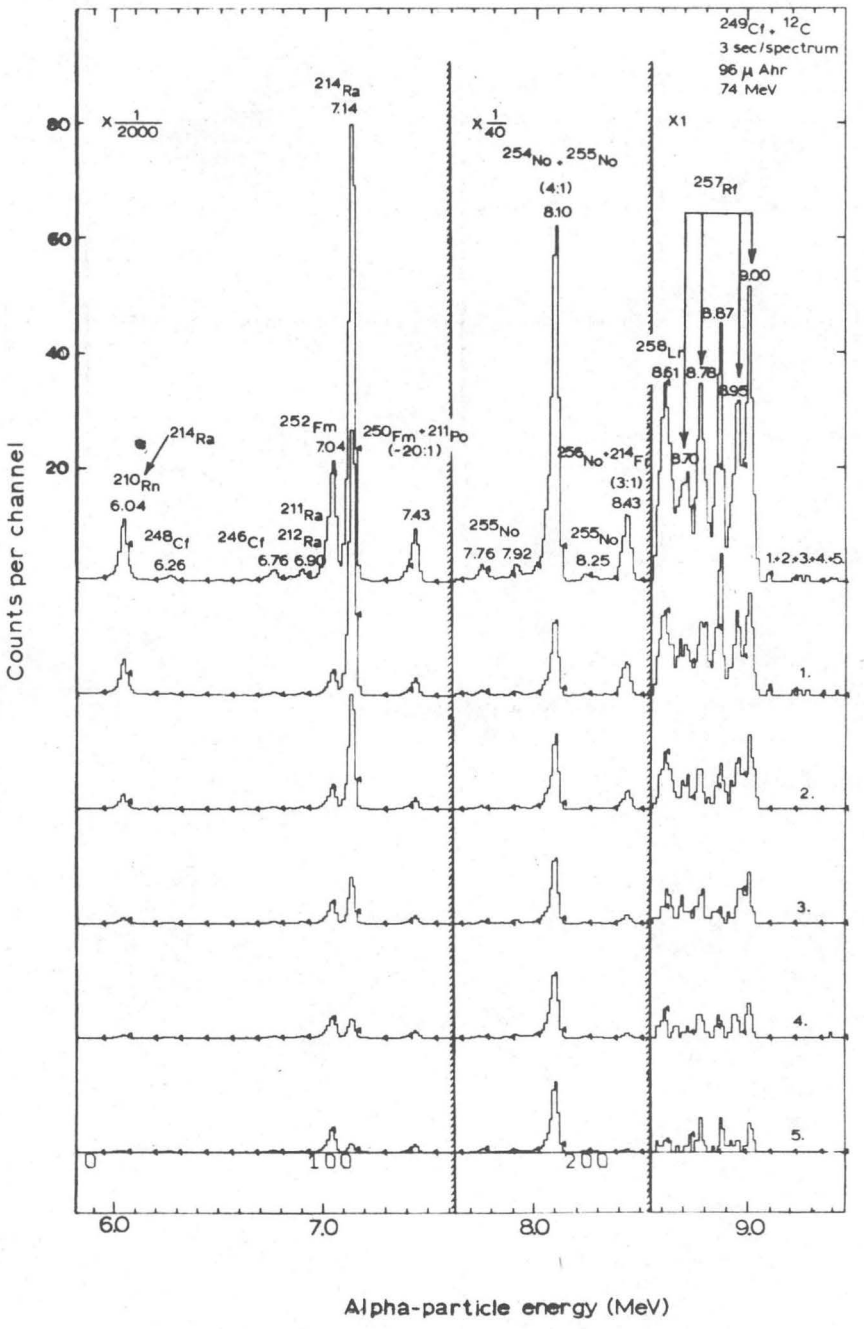
Fig. 3. Detail of detector shuttle system used for the identification of mother-daughter activities by the alpha particle recoil.

Fig. 4. Drum mica system.



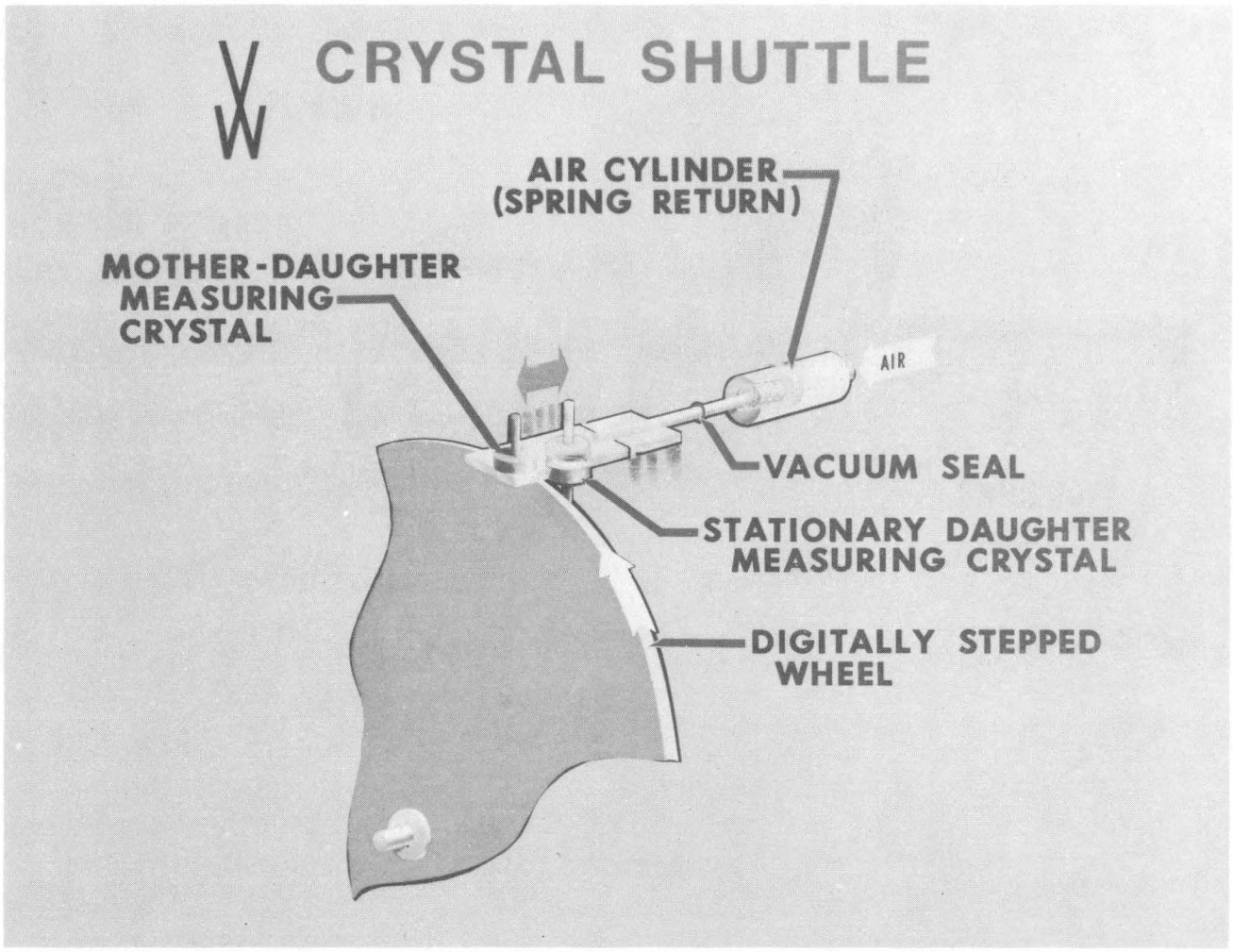
CBB 689-5712

Fig. 1



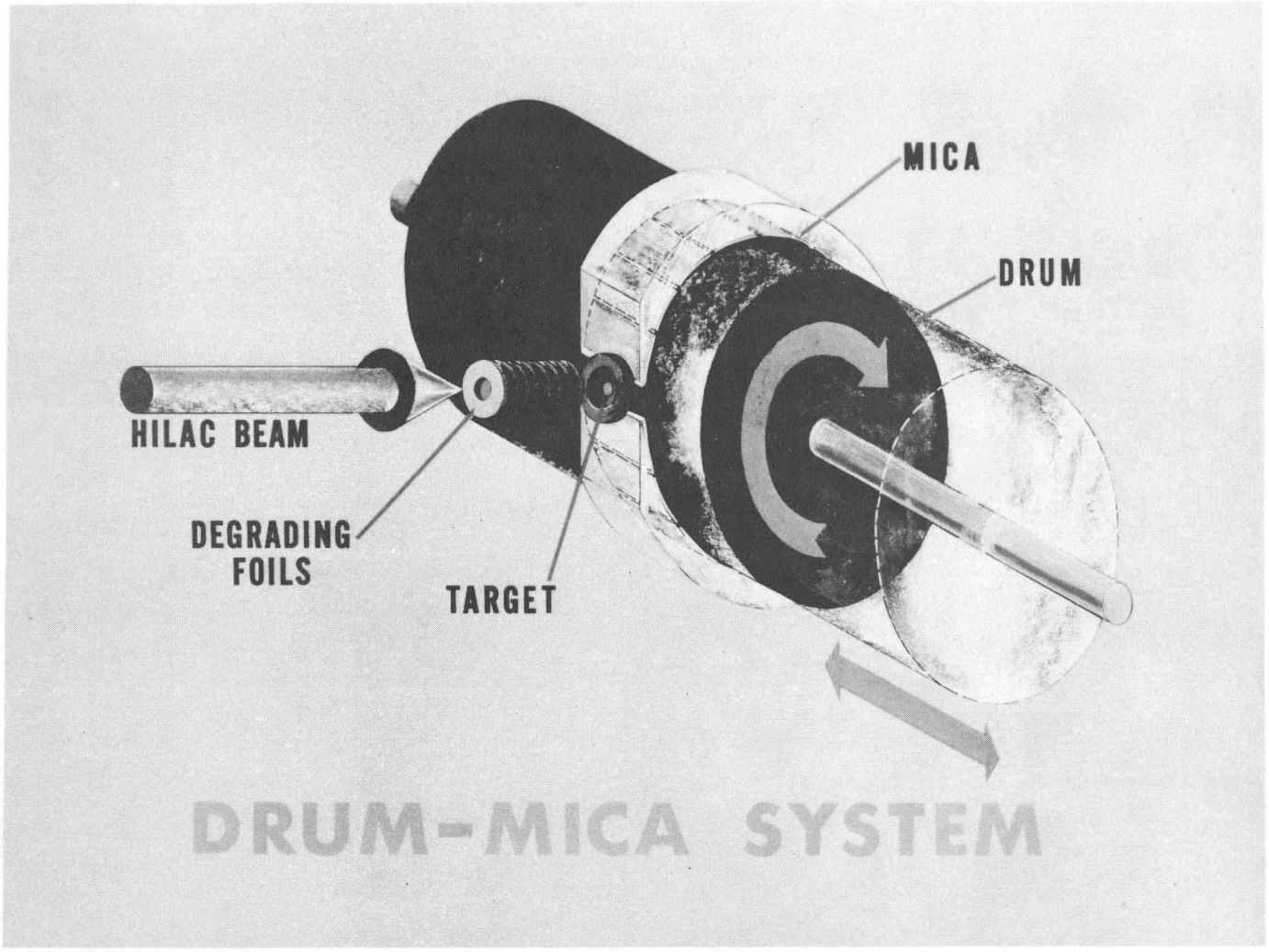
XBL 702 6141

Fig. 2



CBB 693-1899

Fig. 3



CBB 689-5714

Fig. 4

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