

**PROCEEDINGS OF THE
SYMPOSIUM
COMMEMORATING THE 25TH
ANNIVERSARY OF THE DISCOVERY
OF MENDELEVIUM**

**Lawrence Berkeley Laboratory
Berkeley, California**

**Scientific Editor
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CONTENTS

Introductory Remarks	
<i>Glenn T. Seaborg, Lawrence Berkeley Laboratory</i>	1
Reminiscences	
<i>Albert Ghiorso, Lawrence Berkeley Laboratory</i>	13
<i>Bernard Harvey, Lawrence Berkeley Laboratory</i>	21
<i>Gregory Choppin, Florida State University</i>	25
Chemical Properties of Mendeleevium	
<i>E. Kenneth Hulet, Lawrence Livermore Laboratory</i>	32
Nuclear Properties of Mendeleevium	
<i>Darleane C. Hoffman, Los Alamos Scientific Laboratory</i>	45
Radioactive Decay of Md Isotopes	
<i>Arnold M. Friedman, Argonne National Laboratory</i>	56
Concluding Remarks	
<i>Glenn T. Seaborg, Lawrence Berkeley Laboratory</i>	65



Introductory Remarks

Dr. Glenn T. Seaborg

I think we might get started. Welcome to the Symposium honoring the 25th Anniversary of the discovery of mendelevium. I think you are going to find it to be an interesting occasion. I might say to the speakers that we are going to record what you say and later issue, as we have on the occasion of the other 25th anniversaries, a kind of commemorative report. However, perhaps you should regard this as just a sort of hint that we would rather have a manuscript later to cover your remarks put together in a more considered fashion.

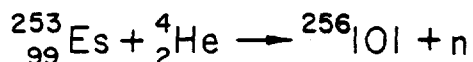
I think it is appropriate to observe an occasion like the 25th Anniversary of the discovery of a chemical element. I certainly think from a standpoint of history this is worth doing, but also, as has been the case for the other 25th Anniversaries, it has resulted in a rather useful summarization of the information on the element as of the dates of the symposia. Mendelevium, I believe, is an interesting element as you will learn from what you are going to hear today.

We have more information concerning the discoverers in this case; more on the record than we have had with any of the other elements. And I think you are going to be a little surprised in a few moments as to what we have available to show you. It is very pleasant to have this reunion of the discoverers and their friends. A sad note, of course, is that Stan Thompson is not here. We certainly miss him and he was one of the key contributors to the discovery of this element. As you know, as I have stated on a number of occasions, Stan

was a life-long friend of mine; we knew each other when we were 13 years old, Freshmen starting high school in the Los Angeles area and, as fate would have it, we spent a good deal of our life collaborating in research in a number of areas.

Now, I am going to lay the ground work with some descriptive remarks concerning the background for the discovery of this element but illustrated by a number of slides. This element was really the turning point in the synthesis of new elements. This is the first element whose discovery was based on the one atom at a time approach, and the first element that used some of the techniques that are necessary for detecting isotopes or elements on such a small scale; all of the elements that have been synthesized and identified since mendelevium have used basically the techniques that were worked out for this occasion.

Now, with that, let me give you the first slide that shows the reaction that was used:



Slide 1

I am going to be rather brief because I am sure that the reminiscers who are following me are going to fill in some of the details. In this case, the reaction that was used is shown here. It was remarkable in that this was really the first time that such a small amount of target material was used; an invisible amount, and I mean a really invisible amount, something of the order of 10^9 or 10^{10} atoms. And the einsteinium-253 had been synthesized over a number of years by the bombardment of lighter isotopes beginning with

plutonium in the Materials Testing Reactor at the Arco Reactor Station. This was then put on a gold backing foil and bombarded in the 60-inch cyclotron with helium ions (40 MeV) with the expectation that this reaction might occur, based on previous experience with reactions of this kind. Now this was a kind of calculation that was made before the experiment was attempted to indicate whether it might be feasible to produce and identify such an element. This is a rough calculation:

$$N \cong N^i \sigma I t$$

$$(10^9)(10^{-27})(10^{14})(10^4) \cong$$

One atom per experiment

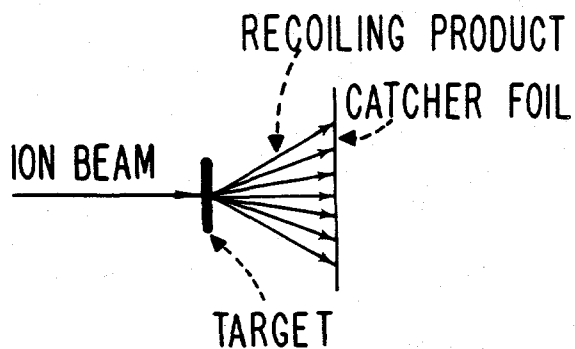
Slide 2

calculating the number of atoms that might be produced which would be equal to approximately the number of atoms of target material times its cross section times the ion beam intensity times the time of bombardment, which of course, would be related to the half-life of the product when bombarding for a time of the order of its half-life. And, on that basis, using for example 10^9 atoms and an estimated cross section of 10^{-27} cm² (about a millibarn), based on measurements of similar reactions in this region, and a beam of 10^{14} helium ions per second, which could only be obtained by literally rebuilding the 60-inch cyclotron because we weren't getting beams of this order of magnitude, and something of the order 10^4 seconds, we came out with the result that one would expect perhaps one atom per experiment. And it was on this basis that it was decided that it was feasible to go ahead.

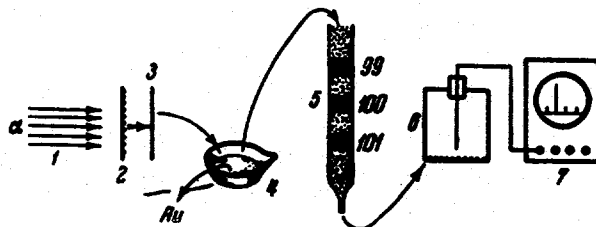
Now one of the key changes or improvements in technique that was introduced here for the first time by Albert Ghiorso, was the recoil technique,

placing the target element on the opposite side of the target from the beam and catching the recoiling atoms on a catcher foil (Slide 3). Some co-workers had already done this in a few previous experiments, I believe, at the 184-inch cyclotron. This was then applied for the first time to the identification of a new element in this manner. And the identification was made as shown here in a schematic manner (Slide 4) in a Russian publication, called "Priroda" (I believe that means "Nature"), that appeared shortly after the announcement in The Physical Review of the discovery of element 101. In this case, the helium ions, labeled 1, struck the target, gold and einsteinium-253 on the backside of the gold foil, labeled 2, and then No. 3, the gold catcher foil, was symbolically dissolved in the crucible shown as No. 4. The transmutation products were then put through a Dowex-50 ion exchange column, actually at high temperature (87°C), and eluted with an alpha-hydroxyisobutyrate solution, and then symbolically detected, as shown here in an ionization chamber to measure, in this case, spontaneous fission.

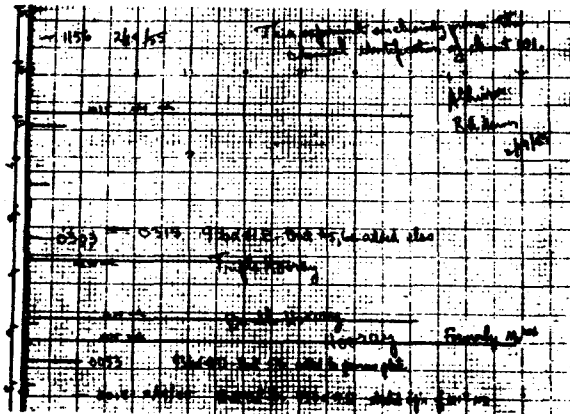
Next we see the famous picture of the recording of their data, with the handwriting of Ghiorso and Harvey made during the all night experiment when the discovery was made (Slide 5). As work went on it appeared that there was a spontaneous fission activity that was involved in a kind of complicated double decay, the mendelevium-256 actually decaying by electron capture with the half-life indicated at that time to be a little under an hour, now known to be 76 minutes, to a spontaneous fission activity, fermium-256, decaying with a half-life of 2.6 hours. The mendelevium was isolated, in the manner that I indicated on the previous slide, by the ion exchange adsorption elution technique. Then, in the definitive experiment, the spontaneous fissions were recorded in this manner during the night of February 19, 1955. The first one



Slide 3



Slide 4

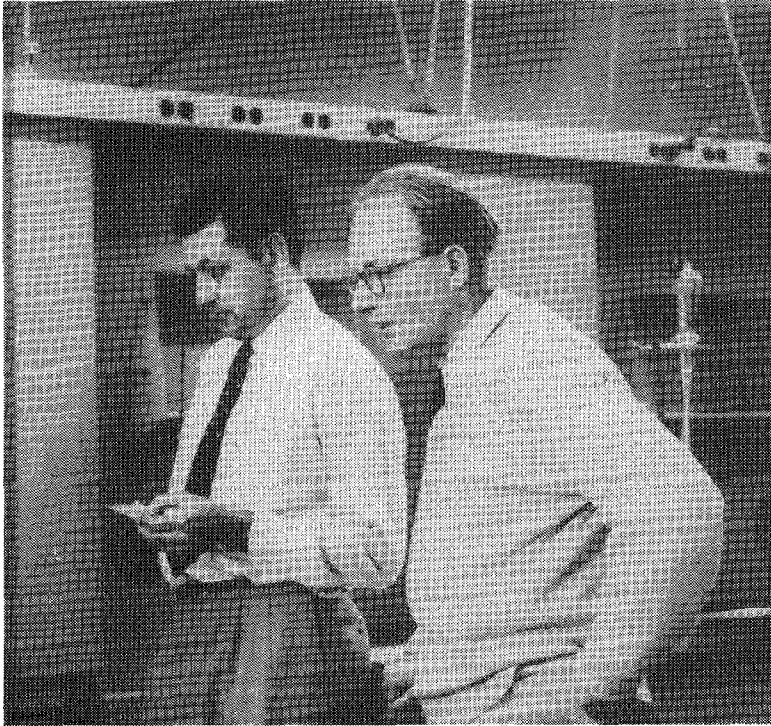


Slide 5

here is identified with a "Hooray, formerly Mv^{256} ," then a "Double Hooray" and a "Triple Hooray"; then there was a fourth one, and the inscription "This experiment conclusively proves the chemical identification of element 101" signed by A. Ghiorso and B. G. Harvey, and dated February 19, 1955.

We have an advantage over the other 25th Anniversaries in that we have pictures taken at approximately this time, actually a little later in 1955. A movie of a reenactment of the discovery was filmed as part of a T.V. program of ten shows on the chemical elements. This series included this episode in one of the later programs which was devoted to the heavier transuranium elements. Here we see, from this film, a younger Albert Ghiorso and Bernard Harvey (Slide 6). Here we see Stan Thompson (Slide 7) at the other end carrying on the chemical identification.

As was often the case in the discovery of these elements, a party was held, in this case at Larry Blake's restaurant, celebrating the event (Slide 8). Here we see Nels Garden in the buffet line along with Albert Ghiorso, and Bart Jones, who was one of the operators of the 60-inch cyclotron. And here at the head table, we see Bernie Harvey, Albert Ghiorso, Bernie Rossi, me and Nels Garden (Slide 9). And here's another view, including Alfred Chetham-Strode, Bart Jones, Tom Parsons, and perhaps you can identify some others (Slide 10). And here the head table again, with Bernie Harvey (speaking), Al Ghiorso, Bernie Rossi, me, Nels Garden (Slide 11). You did not see Stan in these pictures because he was away in Sweden on a sabbatical leave by the time we got around to this party. But here he is in effigy (Slide 12). And here is another picture of the same kind (Slide 13). Now, if I could have the lights.



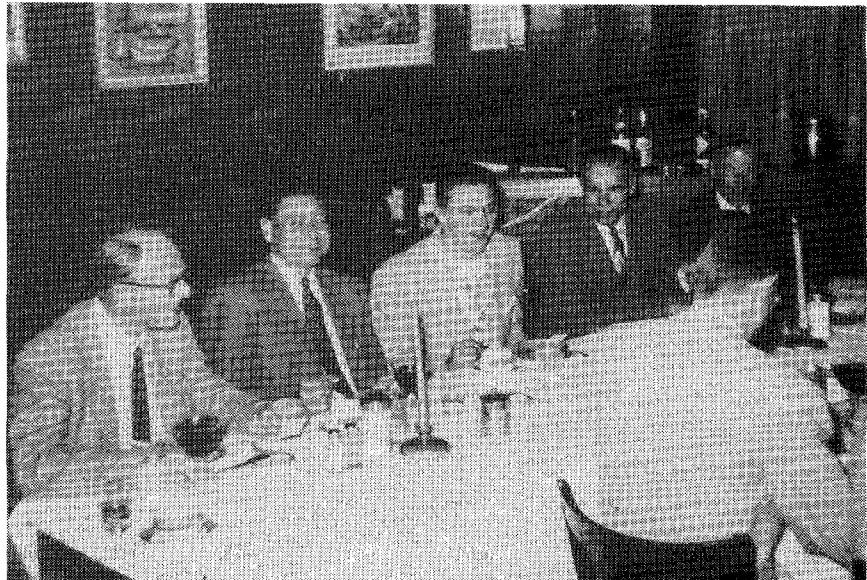
Slide 6



Slide 7



Slide 8



Slide 9



Slide 10



Slide 11



Slide 12



Slide 13

At a little later time, the Daily Cal ran a story that I happened to find in my files, and I am going to read some excerpts of this that I think you might find amusing.

The story goes as follows:

"The University of California Nuclear Metaphysical Laboratories have announced a startling new finding in the world of atomics. The new discovery is an entirely novel element named 'Percentium' by its discoverer, the 15 1/2 year old Leonardo da Vinci. The element, number 101 in the atomic series, follows element number 100, 'Centium.' The youthful da Vinci said that this was the reason for the new element's name, Percentium. 'The interesting fact about Percentium,' said Moosbrugger, 'is that it has a negative half-life. That is,' he went on, 'its radioactivity and total mass increase 1 percent every 100 years. Probably it is the first of a series of elements that spontaneously integrates the successive members of the series.'

"'Percentium is characterized,' said M. Morris, a lab assistant on the project, 'by a satellite proton revolving around the nucleus inside the electron sheath. Preliminary investigations indicate that all the higher elements beginning with number 101 have similar binary nuclei. The satellite proton of number 101, it is believed by the department, picks up neutrons and protons from surrounding matter.'"

"The process is presumed to continue up to element number 202; at this point the twin nuclei are of equal mass. When number 202 adds another proton to the satellite, the latter becomes more massive than the primary nucleus, and the resulting short-lived atom of element number 203 undergoes fission with emission of a proton and a neutron, and becomes two atoms of number 101."

"Apparently natural processes are incapable of producing any of the elements beyond number 100. If they were so capable, the universe would be overflowing with Percentium. It would appear that the artificial production of Percentium has initiated a slow cosmic revolution in the atomic population. It is estimated that after 101 billion years all the matter in the continuum will consist of the elements from 101 to 203 inclusive."

"But this is nothing to worry about," said Kidder with a grin; "we'll all be dead by then."

And by the way, I would also like to call your attention to the fact that in 1972, in a journal that came out periodically, Adventures in Experimental

Physics, edited by Bogdan Maglich, we have a rather good description by Al Ghiorso of the discovery of element 101.

Now I am going to show you the "piece d' resistance." Actually as I have indicated, we have here the unique situation that the discoverers were filmed just a few months after the discovery experiment in a sequence recreating the experiment. Now I happened to have a print of this in my files. Al Ghiorso reminded me that the film existed. I then looked into my files and we found this old film in the warehouse where they keep these things. It is not the greatest in the world, but I think it is good enough for you to be able to recognize the participants in action. And with that, roll the film: (Film shown at this point.)

I am not sure this film was received with the respect that it deserves. Actually, I do not think that the predictions I made at that time were so bad. You must recall that we had not begun to use heavy ions yet, and, of course, everything beyond element 101 was out in the unknown. I do hope that some of the young actinide chemists who are present in the audience this afternoon got some tips on how to carry on their work in this region. Yes, I think there were some practices there that wouldn't be allowed today, even to the extent perhaps of not being allowed to show the samples of actinide elements as I did on educational T.V. I noticed, however, that I didn't have plutonium among those that I demonstrated. I do not recall whether there was a prohibition against that even at that time.

Introduction of Albert Ghiorso

Dr. Seaborg

Now we get on to the first of those among us who are going to reminisce a bit today, and that is Albert Ghiorso. I am going to dispense with any long introduction. We all know Albert. He began his career as a nuclear chemist at the Metallurgical Laboratory of the University of Chicago in 1942. It was there, after carrying on some of the essential war-work, that he could go on to broader investigations that led to the discovery of the next two transuranium elements, elements 95 and 96, americium and curium. He returned to the University of California, the Radiation Laboratory here on the hill, in 1946 and in the intervening years has been the key person in the synthesis and identification, that is the discovery of, all of the heavier transuranium elements, up to and including element 106. Albert...

Reminiscences

Albert Ghiorso

In thinking about what I should say at this 25th anniversary of the discovery of mendelevium, I decided that it might be of interest to tell the story of how the idea originated for this unique experiment—the bombardment of a nearly weightless target to make a new element, one atom at a time.

I used to find that when I went on trips I would inevitably get new ideas; this particular one occurred on a plane trip to Idaho. Bernie and Stan

and Greg and I were going there this time to do some experiments with ^{253}Es to make new nuclides by neutron bombardment in the MTR high flux reactor. On the plane I started playing with some numbers related to the amount of einsteinium that we could obtain, and I suddenly realized that if we took the maximum amount of ^{253}Es that could be obtained at that time, about 3×10^{10} atoms, and bombarded it with a beam of 100 microamperes per cm^2 of ^4He ions, we would make one atom of element 101 every five minutes or so assuming a cross section of about a millibarn. The assumption of beam intensity was about an order of magnitude greater than had ever been obtained, but I blithely assumed that this problem could be overcome.

Very excitedly I took the calculations over to the others and we started to discuss the possibility of making the new element. I remember that Stan and Bernie were not enthusiastic. One of the most critical problems was that the einsteinium target had a half-life of only 20 days so it would be necessary to do the experiments in a very limited time. I argued that we could make it a little easier by making only one target, a recoil target. This would allow us to separate the transmutation products without dissolving the target by merely catching them in a foil placed next to it. This had never been done before with low energy light ions, but I was confident that the technique would work. We finally decided to make the try but to reach our goal took a lot more effort than I had envisioned.

In principle, the idea was perfect but it took us a while before we were able to make it work. We made something like five targets before we had a successful one. The reason for our difficulties was that we at first made the targets by vaporization (I was known as the blow-torch chemist in those days!) and yet they were too thick. We incorporated ^{244}Cm with the ^{253}Es so that we

could monitor our success by the amount of ^{246}Cf that was caught in the recoil catchers. The reason that the early vaporized targets didn't work properly was that a thin transparent deposit of tantalum oxide or tungsten oxide was also vaporized onto the target and this absorbed the fusion recoils very readily. After a number of failures (after each failure the target material had to be recovered and repurified), Bernie got disgusted and said, "Let's electroplate the target." He did so and the target worked very well--an almost weightless target in a very small area. That target survived many bombardments but by the time that we got to the crucial experiments the amount of ^{253}Es was down to about 10^9 atoms, a very marginal amount.

Before we got to this point, however, we had to have a cyclotron that would give us a helium ion beam of at least 100 microamperes per cm^2 --this would be 5 microamperes through an area of 0.05 cm^2 . At that time (this was well before strong-focusing quadrupoles came into use) the only way we could get such an intensity was by taking advantage of the vertical focusing inherent in cyclotrons. We used the internal beam just after it was deflected. At this point the beam was only $1/32$ " high and $1/4$ " long, and this is where we placed the target. The target and recoil catcher cooling requirements as well as the intense radiation problem made the target probe equipment quite complicated. After each bombardment the catcher foil became extremely radioactive, but by judicious quick manipulation we were able to keep our personal radiation exposures down to safe levels.

It was also necessary to improve the 60-inch cyclotron to make the experiment possible at all. This was done by Bernie Rossi who is no longer with us, having died a few years after the experiments. I would like to pay tribute to his memory not only for his work in making this discovery possible but

also for his role in the development of heavy ion technology. I used to work with him one day (and one night) a week to try and accelerate various heavy ions to usable energies and intensities. One of our important findings was the discovery of harmonic acceleration by which C^{2+} and C^{6+} could be accelerated simultaneously. We even developed a beryllium beam at that time! This heavy ion work led directly to the building of the HILAC. Rossi played a very crucial role.

The target problems were considerable but they were overcome. The trouble was that we didn't see anything--no short-lived alpha particles that could be identified as originating from element 101. The reason for the fast little car seen in the film (my supercharged Volkswagen) is that we were sold on the idea that the new hypothetical alpha activity would have a half life of about five minutes so that it was necessary to cut the time from the end of each bombardment until the end of chemical separations to an absolute minimum. We used to telephone the guard at the Laboratory gate that we were coming so that there would be no delay in making entry. I remember one time, however, when someone didn't phone. As usual, as the driver, I raced through the gate going like mad. This particular guard, who was a real eager beaver, jumped out of his post and shouted, "Stop or I'll shoot!" Well, believe me, I wasn't about to stop--I thought he just might shoot! He was very much distressed; he came up to our lab afterwards and we apologized but said that we were too busy with the experiment to talk about the incident at the moment. We got away with it for a while.

As the experiments proceeded we became discouraged because we couldn't find anything after using several days of bombardments. One night after our usual failures we decided to look for something longer lived. We made a some-

what longer bombardment around midnight and the separated trans-einsteinium fraction was placed in a counter for an overnight alpha pulse analysis. I had hooked up a special circuit that would also record the pulse height of the "big kick" that would come from any spontaneous fission that might occur. This circuit was connected to a large chart recorder and we left for the night.

In the morning a look at the chart showed two very high energy events that could only come from spontaneous fissions, and they were separated by a few hours from one another and the end of the bombardment. Just these two events in all the bombardments that we had made--what did they mean? I was very bold and proposed that we had produced an electron-capturing isotope of element 101 decaying to element 100 which then underwent spontaneous fission decay with a half-life of a few hours. The mass number would be 256.

This hypothesis completely changed our whole course of action. Up to this point we had assumed that the isotopes of element 101 that we would make would be short-lived alpha emitters. There was no way of knowing that they would be highly hindered for alpha decay. And now we had the possibility of finding an electron-capturing nuclide decaying to one undergoing spontaneous fission with both nuclides having reasonable half-lives. This was a really wild lucky guess considering that our only evidence were the two "big kicks."

Our strategy now was to assume that the $^{256}_{101}$ isotope had a half-life of about an hour and the $^{256}_{\text{Fm}}$ isotope a half-life of several hours. A number of bombardments were then made and the successive element 101 fractions combined and placed in the alpha grid chamber so that we could analyze for high energy events. (In later experiments we used a group of counters that counted only fissions to obtain a complete chemical elution curve.) In the film, you saw a

picture of the chart showing the times when we observed spontaneous fission events in the element 101 chemical fraction. Our various "Hoorays!" written on the chart were deeply felt.

But it wasn't 0300 yet. We had decided that if we found the element we would call Glenn at that time to let him know the good news! The last one came in at 0315 so I called him around 0330. Although quite sleepy he, of course, was very pleased. I went one step further and told him that we had not only found the element we had also decided on a name for it - mendelevium. The name suggestion came about a year before our discovery when Jack Hollander proposed it at lunch one day. He said, "Why don't you guys name an element after the famous Russian chemist, Mendeleev?" We all thought it was a good suggestion but difficult to implement because of the cold war but filed it away in our minds for future consideration.

After we had finally found the recalcitrant element, we discussed among ourselves what its name should be and found that we were in complete agreement about using the name mendelevium. However, we felt that if we used the cautious approach and asked the opinions of Lawrence and Seaborg they might agree for scientific reasons but demur out of political considerations. Thus we felt that an aggressive approach might be in order--that if we just called it mendelevium maybe it would be all right. And it was. I think Glenn must have gulped over the phone but he didn't object. Neither did anyone else! Everyone seemed to be pleased as a matter of fact. At the 1955 Atoms for Peace Conference in Geneva, the French chemist Haissinsky told me that our naming of element 101 in honor of a Russian scientist had probably done more good for international relations than anything that John Foster Dulles had ever done!

In subsequent trips to Russia, Glenn and I both found that the gesture was warmly remembered.

Comments Following A. Ghiorso's Talk

Dr. Seaborg

Thank you very much.

I think it is true that the naming of this element, mendelevium, had a dramatic impact in furthering, in a positive way, American/Soviet relations at that time. I think it really is rather difficult in today's atmosphere to realize what a step that was at that time. It was quite a step, and there were a number of people who criticized us for it.

I recall that at the time of the visit of Vice President Richard Nixon to the Soviet Union in 1959--the time of his famous kitchen debate with Premier Khrushchev--it occurred to me that it might be useful if he knew the story of mendelevium. He happened to be a long-time friend. I therefore wrote him a letter and told him that we had recently named this element, mendelevium, after the Russian chemist hero, and that at some point or other that might be of some advantage to him in his discussions. I learned later that he used this information; that is, he told some people about this and got quite a good reception. And one of the aftermaths of that was that a little later I got a copy of Mendeleev's chemistry book, autographed by Mendeleev, from a second-hand book dealer in the Soviet Union, who told me that he had learned about this from Vice President Nixon. This was a book that the book dealer had picked up somewhere; actually it was the book that was sent to a physician in

England who played an important role in treating Mendeleev's son for some sickness at the time of a visit that Mendeleev made to England. So, that is an interesting aftermath.

Introduction of Bernard Harvey

Dr. Seaborg

The next co-discoverer we are going to hear from today is Bernard Harvey. I first learned of Bernie's existence when he was working at the Chalk River Laboratory. He was working on the chemistry of plutonium. And he had written a report. He, with a co-worker or two, covered much of the work that the whole American team had done on the chemistry of plutonium. So I thought this is certainly a remarkable chemist; I want to meet him. After I met him on one of my visits to Chalk River, I thought this would be a great fellow to bring to the Radiation Laboratory at Berkeley, and I communicated that to him. For a while, we had to overcome the prohibition on any foreigner--in those days even a Canadian was considered to be a foreigner--and it took some time, a number of years, before we got permission to offer him a position. And then, as Bernie has pointed out many times, I wrote him a carefully worded letter offering him a temporary position in the chemistry group here at the Radiation Laboratory and that is the only thing he has to show for his present position. He is still here on a temporary status. This has never been rectified or amplified, and it is still the basis on which he is here.

Soon after he arrived, within a year to two, he got involved in this experiment on mendeleevium that we are commemorating today. When the 88-inch

cyclotron was built toward the end of the decade of the 1950's and the early 1960's, he took over as the Director and served in that capacity until last year, and now he is back at the 88-inch cyclotron. Bernie, could we hear from you as to what you remember about those days.

Reminiscences

Bernard G. Harvey

Thank you, Glenn. I have to correct one remark that you just made though. It's true that when I came to the Lab in 1953 I did have only a temporary appointment, and that remained true until about three weeks ago, when I received a letter from President Saxon telling me that at last my appointment has been made permanent. I must say during those almost 26 years, the world's longest temporary appointment, I never really worried about when it was going to run out.

Well, I think the key to the successful experiment that you saw in the film and about which Albert and Glenn have talked was really technique. There were several new techniques that had to be developed and we were lucky, (or perhaps we weren't lucky, we were smart), that they all happened to come together at the right time.

First of all, we had to make the recoil target. My memory, and perhaps Greg can correct me if I'm wrong, is that the electroplating technique that we used was developed by Alfred Chetham-Strode who, alas, is no longer with us. Without that technique, as Albert said, the evaporation from a hot filament technique didn't work because of all other materials that were evaporated on

the surface of the target. The electroplating method worked beautifully and it gave a very high yield which, of course, was very important for handling such a rare product as the einsteinium target material. So that was one technique that we just happened to have available to us at the crucial moment.

The other technique that we worked hard on, and that we also needed for this experiment, was the ion exchange separation on the Dowex 50 column. It was Greg's idea to use alphahydroxyisobutyric acid as the eluant. We tried many different eluants and we tried all kinds of conditions. I believe that Greg and I ran an average of three ion exchange columns a day, testing different techniques, over a period of many months in order to be sure that when we started a separation we would know exactly what was going to happen. In fact we knew to one drop where each element would appear. When I first came to Berkeley, there were frequent surprises where sometimes curium, for example, would come off at drop 150 and sometimes it would come off after about 150 liters of eluant. Such an uncertainty, of course, was quite impossible to live with when dealing with an experiment as difficult as the mendelevium experiment. But we really had that down cold; we refined every little detail of the technique of ion exchange columns, and as a result of that we were able to tell exactly where the interesting activities would appear. In fact we got so good at it that I remember working up one of these plutonium samples that had been in the Materials Testing Reactor where one had many curies of fission products and curium and other heavier elements. After we ran the first ion exchange Dowex 50 column to separate out the uninteresting americium and curium from the interesting heavier things, we wanted to separate the really heavy elements one from the other in a second Dowex 50 column, but there was so little curium left after the first pass, we had to put some back in as a

tracer so that we would know what was going on in our column. So, the key to it all was the ion exchange, the electroplating, and, of course, the development of the cyclotron to which Albert referred.

I think there was another very important factor in the success of this experiment. It was that the group of people working on it were highly compatible one with the other and we all worked pretty hard. Albert made the remark that he's lost weight since those days and, seeing the film, I think I can say the same of myself. We needed it perhaps. But it was a good group, we were good friends, we worked very hard on this and many other experiments, and it was a lot of fun; it really was.

One final comment, I'm not sure I ever saw the film series on the television screen. The reason was that KQED, which was then a very new educational television station in the Bay Area, showed it at a most unfortunate time of day, and although my wife tried hard to get the children to watch their father, she was usually unsuccessful because the time of the film coincided with something more interesting, perhaps the Lone Ranger. Or perhaps it was at 6 a.m.—I don't remember. The children certainly had their priorities which were not necessarily those of the makers of the film. So, Albert, Glenn, Greg, it was a lot of fun, and I must say I thoroughly enjoyed working with you. I wish we could do it again.

Comments Following B. Harvey's Talk

Dr. Seaborg

Thank you, Bernie.

The Lone Ranger T.V. show was difficult to compete with. I remember once I appeared on the KQED program, Science in Action, as a guest. It was broadcast at some prime time like 7 p.m., and after the show was over I drove home. As I drove into my driveway, a number of kids in our neighborhood came running over and said "Oh, Glenn, we just saw you on T.V." I said, "Well, how did you like it?" They said, "Oh, we changed over to the Lone Ranger."

Introduction of Gregory Choppin

Dr. Seaborg

Next, I have the pleasure of introducing the discoverer of alpha-hydroxyisobutyrate as an eluting agent. This, by the way, is still the best eluting agent so far as I know for the separation of the actinides, 25 years later. I do not know why you don't get busy, Gregory, and develop something better.

Gregory Choppin joined our group in 1953, coming up from the University of Texas. He immediately began to work in the heavy isotope field with the heavy isotope people. As I recall, he was involved in the identification of the neutron-produced isotopes from the MTR at Arco, Idaho; isotopes like berkelium-249, californium-249, -250, -251, -252, einsteinium-253, -254,

fermium-255, and so forth. And then he went on to this mendelevium experiment and finished his work with us in 1956. In the meantime, he has become one of the world's leading investigators in the chemistry, especially the ion exchange chemistry, of the actinide elements, having done some of the nicest work in the world in this field.

Gregory, it is now your turn.

Reminiscences

Gregory R. Choppin

I am grateful to Professor Seaborg for inviting me to participate in this anniversary symposium on mendelevium. Particularly helpful is the fact that he put my talk after that of Al Ghiorso and Bernie Harvey, who have recalled rather well the significant aspects of those days. Obviously, this relieves me of the obligation to say anything significant.

When I arrived at the Rad. Lab. in June, 1953, it was with an incredible vacuum of knowledge about nuclear chemistry in general and heavy elements in particular. For the first few months I could not remember which was element 94 - americium or curium - since, obviously, the element after uranium, element 93, was plutonium. You perhaps can imagine how it was to be around Stan Thompson, Al and Bernie as they threw around phrases like citrate columns, 6.44 MeV, 15 barns, etc. That Glenn accepted me as a post doctoral assistant with my background in liquid ammonia research is still a surprise but I have always been grateful. Thank you, Glenn, very much.

So many things happened during the three years I worked with Stan, Al and Bernie. It was long hours, with quick transitions between many projects and fast trips to the M.T.R. in Idaho. But it was even more a time of learning from three amazing teachers. A particular memory was one trip to the M.T.R. to process the irradiated californium sample from the "Mike" shot debris. We were trying to "rediscover" element 100 so its formation could be published. Bernie and I opened the aluminum rabbit under the water in the reactor pit and discovered it was empty - no little quartz capsule with the Cf and, hopefully, the Fm.

The day before we had cleaned the trench of debris and, in doing so, had found a small quartz capsule which we stored in a bucket in the trench in case anyone came looking for their lost experiment. Stan looked at this capsule and proclaimed it our Cf sample! In fact, I doubt he had seen the Cf capsule when Bernie and I prepared it 3 months earlier. Based on Stan's certainty, the capsule was returned to the reactor for several days reactivation. Meanwhile the four of us went to Sun Valley to ski and to avoid being in touch with Glenn who was more nervous about such things. We returned, betting on whether the capsule was, indeed, the Cf sample or, maybe, cobalt - in which case we might spread ^{60}Co around. Of course, Stan's chemical intuition (or luck) held and we could report the formation of element 100.

In the experiments to make element 101, my primary responsibility involved the chemical separation for which we developed alpha-hydroxyisobutyrate as an eluant from columns of cation exchange resin. We needed something better than citrate or lactate to separate a few atoms of Md from Fm. It seemed to me that α -But (our shorthand for α -hydroxyisobutyrate) might give us better separations. Stan agreed and called to campus, where an

old bottle with about 2 grams was found in the stockroom. Bernie and I, with the help of Docia McKenna and Marge Nervik, began studies which eventually led to performing several hundred column elutions over the next few months. For about the first month we had only those 2 grams, so we had to recover and reuse the eluant solutions. Effects of α -But concentration, pH, temperature, column dimensions, loading technique, flow rate and drop size were among the variables we studied until we had a system which allowed us to separate a few atoms of Md and to predict to the exact drop when those atoms would elute. We only had 5 fission counters to use for the peaks of the 101 and 100 elution as well as for the valley before, between and after the peaks. An additional problem was that Es was the last actinide we could use to calibrate our elutions, thus we had to rely on the rare earth analogs of Fm and Md - Er and Tm. Obviously, we solved our problems or we would not be celebrating this anniversary for another year or more.

One of my strongest memories of element 101 is not related directly to the discovery experiments, but rather to the repeat experiments that we did about 6 months later. At that time Otto Hahn had come to the United States on a visit as a result of an invitation by Glenn Seaborg and E.O. Lawrence. Hahn was giving a seminar in the chemistry department, and Glenn mentioned to him that element 101 was being rediscovered that night. He expressed an interest in watching some of the experiment so we were told he would probably come around for a few minutes. Hahn came, watched and poked into everything and stayed and stayed. Lawrence and Seaborg were getting a little tired and suggested leaving, but Hahn said he wished to watch some of the decays. He was like a 5-year-old kid in his curiosity, and it was very impressive to see such an eminent scientist still excited about an experiment. This was quite an

experience for me because there I was in this very small counting room which was half Molar in Nobel prize winners.

In conclusion, this is an opportunity to express my great personal debt to Stan, Al, Bernie, and Glenn from whom I learned so much. Their consideration and friendship are the best memories of those days.

Glenn would come to the lab each afternoon and casually ask how things were going. I soon learned that he understood all and forgot nothing. The luncheon discussions in his office taught me much besides heavy element chemistry.

From Stan Thompson I learned that intuition beats careful thinking much of the time. Stan must have been one of the world's greatest instinctive chemists. Time and again the unexpected would happen and Stan would pull something out of his memory to suggest we try. Of course, as a new Ph.D. I knew Chemistry and I would often explain why that would not work. But we would try it and it did work. Stan taught me not to rely too much on books, but instead, to go ahead and try an unlikely technique or experiment. Working with Stan Thompson was one of the finest things that has happened to me.

I learned from Al that enthusiasm and energy are absolute prerequisites if you are going to get very far in research. I was constantly amazed that he never ran out of ideas, he never ran out of energy, and he was always arguing about everything. Along the way he taught me a great deal about politics. After cyclotron runs sometimes Al and Bernie would drive me home. Often we would stop, about 2-3 a.m., for coffee and Al would lecture me that I had to be more liberal in my thinking. I have tried to keep this as well as the other lessons that Al taught me as to how to go about doing science while enjoying it thoroughly.

But if I had come to believe that instinct and enthusiasm were enough, it would have been an inadequate education, so I have always been grateful that Bernie was part of the group. First, although he was quite experienced, he had just come to Berkeley. So, as the new members of the group, we developed a close relation - possibly as a slight defense against the two old pros - Stan and Al. Bernie combined careful, logical thinking and deep understanding. The same precision, care, and understanding were evident in his laboratory work.

So each of my co-workers affected me greatly and I thank them for that. I also thank them for three exciting years during which mendelevium was born.

Comments Following G. Choppin's Talk

Dr. Seaborg

Thanks, Greg. I'm glad you brought up this capacity of Stan Thompson for intuitive thinking in chemistry. He had the best--and I hesitate to say this in German because Mike Nitschke is here--"chemisches gefuhl" of anybody that I know. That was true throughout his career, especially at Chicago, at the Metallurgical Laboratory, when we developed the chemical process for the isolation of plutonium. This was of extraordinary importance for the free world at that time. He was the moving spirit behind that process, the originator of the process which wasn't supposed to work. Our main problem at Chicago was that we were working on something that was considered to be impossible. It just didn't follow the principles of chemistry, but Stan kept saying that

this'll go, it's o.k. And it was probably the only process that we could have put through in the time at our disposal.

Also, your mention of Otto Hahn reminds me of another story. In 1966 when he must have been 87 or 88 years old, he came to Vienna to receive the Fermi Prize, and I had the responsibility and pleasure as Chairman of the Atomic Energy Commission to bestow this upon him in a huge ceremony. After the ceremony and later in the evening, when most of us were getting kind of tired and wanted to retire, he wanted to go out to the bar. And then, believe me, he did have an eye for the girls. And I think we finally had to say, you know, Professor Hahn, don't you think maybe it is getting a little late for a man of your age. I guess he finally humored us and called it quits.

I should mention that Greg Choppin traveled the farthest of anybody in coming to this meeting. He is at Karlsruhe on a sabbatical leave this year at the Institute for Transuranium Research, so we appreciate very much his taking the trouble to come this distance.

Introduction of E. Kenneth Hulet

Dr. Seaborg

And now, we are going to get on to the serious part of the program. This is the part where we are going to hear about the chemical and nuclear properties of mendelevium, and the first speaker for that portion of the program is going to be Kenneth Hulet. Kenneth Hulet received his bachelor's degree from Stanford University, with his major in chemistry, in 1949. Then he came to work for us in the Nuclear Chemistry Division of the Radiation Laboratory rather soon after he received his degree, and he worked for a while, as I recall it, as a sort of technician in Stanley Thompson's group. But quickly, he showed his capability, was accepted as a graduate student, and went on into the graduate program. He was nominally my graduate student but he did his work with Stan Thompson, and for that matter more or less independently. He got his degree in 1953 and soon went to the Livermore Laboratory where he has had a distinguished career of research on the chemical properties and the nuclear properties of the actinide elements, particularly the heavy actinide elements. He has been one of the key persons in the discovery and the elucidation of the plus 2 state of the actinide elements, which is observed as you go up toward the top of the series. He has been involved in the synthesis and the characterization of the nuclear properties of a number of the heaviest actinide isotopes.

Ken Hulet...

Chemical Properties of Mendeleevium

E. Kenneth Hulet

In the earlier observances in this series commemorating the discovery of new elements, a profusion of information on the chemical properties of the elements 97 (Bk), 98 (Cf), 99 (Es), and 100 (Fm) was available 25 years after their discovery. Many properties obtainable from bulk samples had been measured and we therefore knew of the structure of their metals and simple compounds, their vapor pressures, densities, magnetic susceptibilities, and energy levels in their atoms and molecules. Such information was obtainable only because milligram amounts of Bk, Cf, and Es could be manufactured by neutron irradiation of the lighter actinides. Beginning with Md, a greatly reduced amount of knowledge will ever become available for the reason that the synthesis of these heaviest of the known elements is only possible by ion bombardment of lighter actinide target nuclei. Even with the most intense ion beams and the largest available quantities of target isotope, about 10^6 atoms at a time is all the Md that can be produced for chemical studies. This lack of sufficient sample size coupled with the very short lifetimes of the few atoms produced has severely restricted the gathering and broadening of our knowledge concerning the properties of Md and the heavier elements. To illustrate, the literature contains a mere eleven references to the chemical studies of Md, and none of these deal with bulk properties associated with the element bound in solid phases.

The isotope ^{256}Md is nearly always employed for chemical studies of this element. Besides having a convenient half-life of 77 min, this nuclide can be

made with millibarn cross sections by a number of nuclear reactions between light or heavy ions with actinide target nuclei. We have found that the bombardment of fractions of a microgram of ^{254}Es with intense alpha-particle beams will produce $\sim 10^6$ atoms of ^{256}Md in one to two hours of irradiation time. The ^{256}Md is most easily detected through spontaneous fission arising from the ingrowth of its electron-capture daughter ^{256}Fm . A difficulty with using spontaneous-fission counting to determine the Md content of samples is that the growth and decay of fission radioactivity in each sample must be followed with time in order to resolve the amounts of Md and Fm initially present. However, alpha-particles of a distinctive energy coming from a 10% alpha-decay branch can also be used to identify ^{256}Md in a mixture of actinide tracers.

Mendelevium and Fm metal were found to be more volatile than other actinide metals. In the numerous thermochromatographic studies by Zvara and coworkers,¹ the evaporation of Fm and Md tracer from molten La at 1150°C was compared with the behavior of other selected lanthanides and actinides. The volatility of Md and Fm was found to be greater than that of Cf, Cf was about equivalent to Yb and Eu, and all were much more volatile than Am. The volatilities are correlated by the number and energy of the valence bands minus the energy needed to promote electrons to the valence bands in the metals. Therefore, within the normally trivalent lanthanides and actinides, the more volatile elements are associated with the divalent metals. The unusual volatility of Fm and Md was then construed by Zvara as evidence for divalency in the metallic state.

There are no experimental verifications of the electronic structure of Md, but this has been calculated by several methods to be $5f^{13}7s^2$ in which the ground state level is $^2F_{7/2}^2$.

The separation of Md from the other actinides can be accomplished either by reduction of Md^{3+} to the divalent state³ or by chromatographic separations with Md remaining in the tripositive state. Historically, Md^{3+} has been separated in columns of cation-exchange resin by elution with α -hydroxyisobutyric acid solutions.⁴ This method is still widely used even though extraction chromatography requires less effort and attention to technique. Horwitz and coworkers⁵ developed a highly-efficient and rapid separation of Md^{3+} by employing HNO_3 elutions of columns of silica powder saturated with an organic extractant, bis(2-ethylhexyl)phosphoric acid. The separation of Md from Es and Fm could be completed in under 20 minutes and had the advantage of providing final solutions of Md free of complexing agents that might be an interference in subsequent experiments.

When the divalent state of Md was first discovered, extraction chromatography was used to prove that the behavior of Md^{2+} was dissimilar to that of Es^{3+} and Fm^{3+} .³ The extractant, bis(2-ethylhexyl)phosphoric acid (HDEHP), has a much lower affinity for divalent ions than it does for the tri- and tetravalent ones. Thus, the extraction of Md^{2+} is much poorer than the extraction of the neighboring tripositive actinides as indicated by the results shown in Table 1. This became the basis for a separation method in which tracer Md in 0.1M HCl is reduced by fresh Jones' Reductor in the upper half of an extraction column containing HDEHP absorbed on a fluorocarbon powder in the lower half. Mendeleevium, in the dipositive state, is rapidly eluted with 0.1M HCl whereas the other actinides are retained by the extractant. The separation is

Table 1. Comparison of the extraction behavior of tracer Es, Fm, and Md after treatment with various reducing agents. The column-elution method of extraction chromatography was used with the extractant, bis(2-ethylhexyl)phosphoric acid (HDEHP) adsorbed on a column bed of a fluoroplastic powder. (Reprinted with the permission of Science: Copyright 1967 by the American Association for the Advancement of Science).

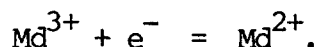
CONDITIONS FOR REDUCTION	STANDARD POTENTIAL OF REDUCING AGENT (volts)	% NON-EXTRACTED BY HDEHP COLUMN	
		Md	Es-Fm
Zn(Hg) AMALGAM, 80° ~20 min, 0.1 M HCl; Zn(Hg) AMALGAM IN UPPER HALF OF EXTRACTION COLUMN	+0.763	77	<0.10
0.01 M Eu ²⁺ , 0.1 M HCl, ~2-3 min, 80°; Zn(Hg) AMALGAM IN UPPER HALF OF EXTRACTION COLUMN	+0.43	75	<0.10
0.6 M Cr ²⁺ , 0.1 M HCl, ~2 min, 25°C; EXTRACTION COLUMN PRE-WASHED WITH 0.6 M Cr ²⁺ IN 0.1 M HCl	+0.41	99	0.56

quickly performed, but the Md contains small amounts of Zn^{2+} from the Jones' Reductor and also Eu^{2+} , which was added prior to the elution to prevent reoxidation of Md^{2+} by the extractant.

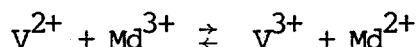
The solution chemistry of the trivalent oxidation state has not been investigated beyond its behavior in the separation procedures described above. All observations indicate that Md^{3+} is a "normal" actinide with an ionic radius slightly less than that of Fm. As might be expected, attempts to oxidize Md^{3+} with sodium bismuthate failed to show any evidence for Md^{4+} .³

The divalent oxidation state was the first found for any member of the actinide series^{3,6} and, therefore, stirred a strong theoretical and experimental effort to establish the reasons for the unexpected stability of this state in Md, and subsequently, in the adjacent actinides. We shall summarize the interpretations for divalency in the heaviest actinides in a later section, but in this section, only the known properties of Md^{2+} will be presented.

In the earliest experiments with Md^{2+} , rough measurements were made of the reduction potential for the half-reaction



The first measurement gave a reduction potential of -0.2 V with respect to the standard hydrogen electrode.³ This value was obtained from determining the equilibrium concentration of each metal ion in the reaction



and then calculating the equilibrium constant. After entering the equilibrium constant into the Nernst equation, it was found that V^{3+} was a better reducing agent than Md^{2+} by about 0.07 V. In other experiments, Mály observed the

complete reduction of Md^{3+} with V^{2+} but the reduction was incomplete when Ti^{3+} was used.⁶ From these observations, he concluded the standard reduction potential of Md^{3+} was close to -0.1 volt. The standard potentials obtained by both groups are in reasonable agreement and, most importantly, they conclusively show that the stability of Md^{2+} is greater than any lanthanide(II) ion. This finding was surprising since divalency in the lanthanides is mainly associated with the special stability given by the half-filled and fully-filled f -electron shell. Divalent Md ions are at least one electron short of the stable $5f^{14}$ configuration.

Additional experiments which may not be clearly relevant to the divalent oxidation state include the reduction of Md^{3+} to $Md(Hg)$ by sodium amalgams and by electrolysis.⁷ Both the extraction experiments with Na amalgams and the electrolysis at a Hg cathode indicated a large enrichment of Md in the Hg phase relative to that of Np , Pu , Am , Cm , and Cf . The percentages of Es and Fm in the sodium amalgam were not greatly different from the percentage of Md . But a clear enrichment of Md was obtained in the electrolysis experiments as shown in Fig. 1. The initial rate of amalgamation is much larger for Md than for Es and Fm .

Recently, new electrochemical experiments were carried out with Md in which controlled-potential electrolysis was used to study the reduction of Md^{3+} to the metallic state in a Hg amalgam.^{8,9} Half-wave potentials were measured by radiocoulometry and radiopolarography in the presence of noncomplexing and weak and strong complexing agents. The radiopolarogram obtained for Md in a noncomplexing media is presented in Fig. 2. The half-wave potential for Fm was remeasured at the same time as that of Md because of its presence as a decay product of ^{256}Md . The results showed that the reduction

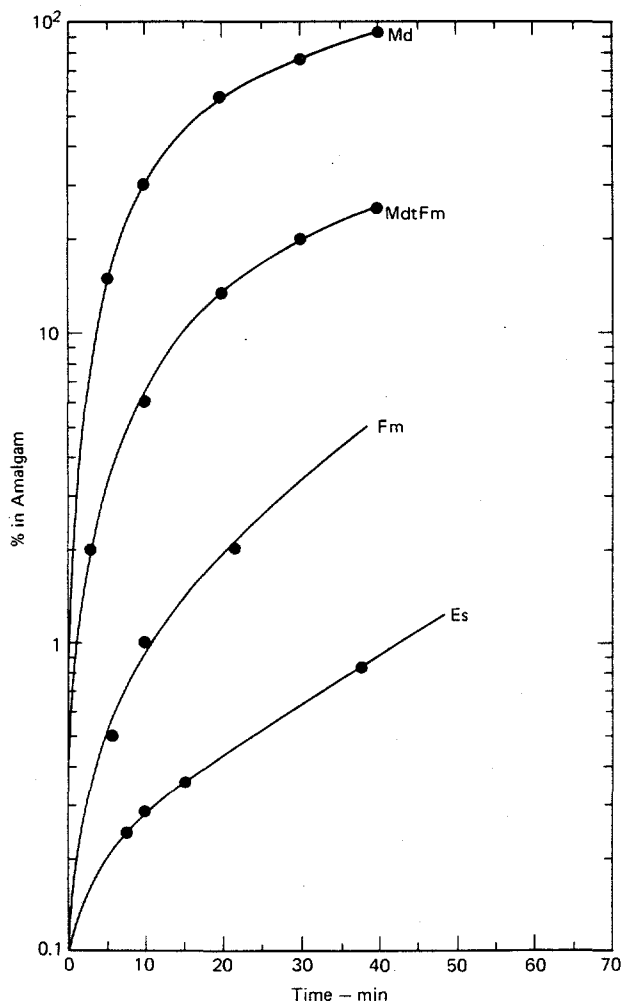


Fig. 1. Percentage of actinide tracers electrodeposited in mercury as a function of time and passed charge. Current density used in this experiment was 5 mA/cm^2 . (Reprinted with the permission of J. Mály and Pergamon Press (Ref. 7)).

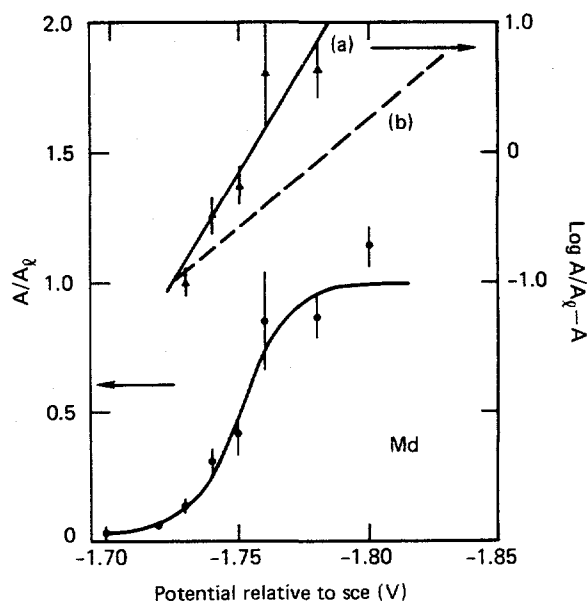
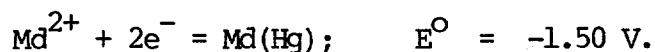


Fig. 2. Distribution of Md as a function of applied voltage between mercury in a dropping Hg cathode and 0.1 M tetramethyl ammonium perchlorate at $\text{pH} = 2.4$. The slope of the logarithmically-transformed line indicates the number of electrons exchanged in the electrolysis reaction. The slope of line (a) is 30 mV , and (b) is 60 mV , which corresponds to a one-electron reduction. (Ref. 9).

potential of Md is about 10 mV more negative than Fm and that no significant difference is observed upon changing the medium from ClO_4^- to Cl^- . In citrate solutions, a shift of 90 mV was obtained for Md which is about the same shift seen with Fm and Ba ions in a citrate medium. The slope of the logarithmically transformed wave was 30 mV for Md and Fm.

The electrochemical reaction taking place at a reversible electrode can be deduced from the slope of the polarographic wave. Specifically, the number of electrons exchanged at the electrode, based on the Nernst equation, is obtained from this slope. From the analysis of the polarograms, there were three electrons involved in the electrochemical reduction of the trivalent ions of the elements Am through Es and only two electrons for the reduction of Fm and Md. This implies that Md^{3+} was first reduced to Md^{2+} before being further reduced to the metal. The III \rightarrow II reduction step is not detected by this radiopolarographic technique because both the III and II ions are in the solution phase; whereas, the measured parameter is the distribution of the tracer between the aqueous and Hg phase.

These results demonstrate that the electrochemical behavior of Md is very similar to that of Fm and can be summarized in the equation



The half-wave potentials measured by this method include the amalgamation potential of the metal-mercury reaction. The amalgamation potential was estimated to be 0.90 V by using the metal radii as a correlating parameter and interpolating within a series of divalent elements with known amalgamation potentials.¹⁰ This correlation is shown in Fig. 3. The standard electrode potential is then given as -2.40 V for the $\text{Md}^{2+} + 2e^- = \text{Md}^0$ reaction. The

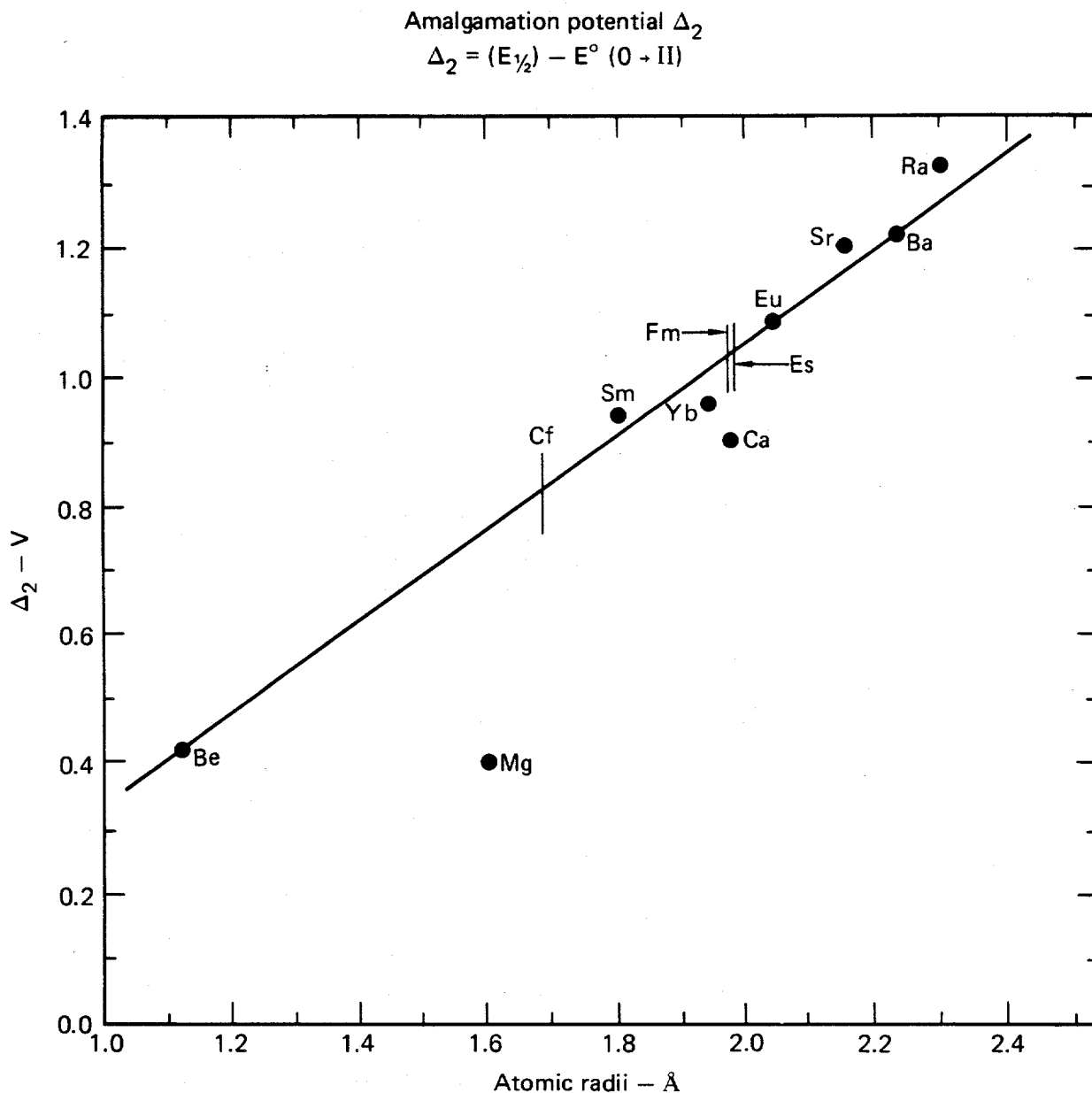


Fig. 3. Amalgamation potentials, Δ_2 , derived from experimental data are plotted as a function of the atomic (metallic) radii. The amalgamation potential for Fm is obtained by using an estimated radius. (Partially redrawn from Ref. 10).

author's estimated 5 mV accuracy for the measured half-wave potential seems reasonable, but there is a much larger uncertainty in the estimated amalgamation potential. Because the amalgamation potential represents a large correction in obtaining the standard potential, caution should be exercised in combining this standard potential with other data to calculate additional thermodynamic properties.

In addition to the di- and trivalent ions of Md, a stable monovalent ion was reported by Mikheev et al. in 1972.¹¹ This oxidation state was indicated in the cocrystallization of Md with CsCl and RbCl after the coreduction of Md^{3+} and Sm^{3+} with Mg in an ethanol-7 M HCl solution. Mendelevium was also found enriched in Rb_2PtCl_6 precipitates, a specific carrier for the larger ions of the alkali metals. These results were explained by a stabilization of the monovalent ion due to completing the f shell which would give the $5f^{14}$ electronic configuration.

These experiments were recently repeated and a series of new ones were performed in which attempts were made to prepare Md^+ by reduction with $SmCl_2$ in an ethanolic or fused KCl medium.¹² After the reductions, the coprecipitation behavior of Md was compared with the behavior of tracer amounts of Es, Fm, Eu, Sr, Y, and Cs. A large number of experiments showed that Md consistently followed the behavior of Fm^{2+} , Eu^{2+} , and Sr^{2+} rather than the behavior of Cs^+ . The most telling experiment was the precipitation of Rb_2PtCl_6 after reduction of Md^{3+} with Sm^{2+} . The distribution of the tracer elements between the precipitate and an ~85% ethanol solution is given in the form of a ratio in Table 2. These results clearly demonstrate that Md did not coprecipitate with Rb_2PtCl_6 , whereas virtually all of the Cs did so. The

overall conclusion of this work was that Md cannot be reduced to a monovalent ion with Sm^{2+} , and therefore, the earlier claim for Md^+ , was unsubstantiated.

Table 2. Distribution of tracer elements after reduction with Sm^{2+} and coprecipitation with Rb_2PtCl_2 in ~85% ethanol. (Ref. 11).

Distribution ratio for						
Fm	Md	Eu	Sr	Y	Es	Cs
0.004	0.005	0.006	0.012	0.017	0.033	110

This same conclusion was reached also by Samhoun et al.⁸ and David and coworkers⁹ on the basis of their electrochemical investigations of Md, which we described earlier. If the potential for the reaction $\text{Md}^+ + e^- \rightarrow \text{Md}$ was more positive than -1.5 V, it would have been observed in the electrochemical reductions. Furthermore, the logarithmic slope of the Md reduction waves could not be fitted to a slope of 60 mV expected for a one-electron change. And lastly, the shifts in potential caused by complexing Md with either citrate or chloride ions were consistent with it being a divalent ion and not with it being either a cesium-like or silver-like ion.

The attempts to produce a monovalent state have the positive effect of setting limits on its stability. From the limits obtained, we can then make an estimate of the stability of the $5\underline{f}^{14}$ configuration relative to the $5\underline{f}^{13}7\underline{s}$. Presumably, the $\underline{f}^{13}\underline{s}$ configuration lies lower in energy than the \underline{f}^{14} because there is no obvious stabilization of a monovalent state due to a possible

closing of the $5f$ shell. The divalent ion is at least 1.3 V more stable than the monovalent.

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Introduction of Darleane Hoffman

Dr. Seaborg

Our next speaker is Darleane Hoffman of the Los Alamos Scientific Laboratory. She is much too young for me to have known her as long as I have known the previous speakers. Darleane received her Ph.D. degree from Iowa State College; I guess it is Iowa State University now. Her first position was actually with the Oak Ridge National Laboratory where she worked on the ill-fated aircraft nuclear propulsion project. As a matter of fact, that was one of the first things that I went to President Kennedy about when I became Chairman of the Atomic Energy Commission. I said that I didn't think that this project was a good way to spend money. Very soon after that the project was terminated.

Darleane went from Oak Ridge, I believe, directly to the Los Alamos Scientific Laboratory, where she worked in the radiochemistry group, became the associate group leader very soon, and turned her attention to heavy isotopes, actinide isotopes, and in particular to an elucidation of the fission reaction. I would rate her as one of the world's leading authorities in the investigation of nuclear fission. She has discovered this interesting phenomenon of symmetric fission in the heavy fermium isotopes, fermium-258, fermium-259, and so forth. She and her co-workers made the interesting discovery of plutonium-244 in nature. Due to Darleane, we can now no longer speak of the 92 natural elements; we have to say the 92 plus plutonium natural elements. Recently, and I do not know whether this was for the good of science, or even for the good of Darleane, she assumed the position as Director

of the Chemistry and Nuclear Chemistry Division at Los Alamos. Nevertheless, she is keeping her hand in scientific work, and we have the pleasure of having her give a little bit of her time in association with our research group here at Lawrence Berkeley Laboratory.

Nuclear Properties of Mendelevium

Darleane C. Hoffman

Sup

Mendelevium is a particularly interesting element from the standpoint of its nuclear and fission properties. Isotopes from mass 248 to 259 are known—most of these are neutron deficient and decay by alpha emission or electron capture. The heaviest known isotope of mendelevium, ^{259}Md , is nearly on the line of beta stability and decays¹ predominantly via spontaneous fission (SF) with a half-life of 95 minutes. So far, no heavier isotopes of mendelevium have been detected. Viola et al.² estimate a beta-decay energy of about 0.7 MeV and an electron-capture (e.c.) decay energy of about 0.5 MeV for ^{260}Md , while Myers³ estimates 0.5 and 1.5 MeV, respectively. A beta-decay energy of about 0.15 MeV² to stable³ is estimated for ^{261}Md , which both^{2,3} predict will be stable toward e.c. decay. Assuming first-forbidden beta decay with a log ft of 6, the half-lives for beta decay would then be from 2 to 7 hours for ^{260}Md and 50 days to stable for ^{261}Md . The electron-capture half-life for ^{260}Md would be from 1 to 10 hours. Thus for ^{261}Md and heavier isotopes the half-lives will be determined by SF decay since, as discussed later, it is expected to be very short. These data are summarized in Table 1.

Table 1. Predicted decay energies and half-lives for heavy Md isotopes.

Isotope	260	261	262
Q_{β} (MeV)	$0.5^3 - 0.7^2$	0.15^2 to stable ³	$1.1^3 - 1.7^2$
$T_{1/2}$	2 - 7 h	50 d to stable	5 m - 1/2 h
Q_{ec} (MeV)	$0.5^2 - 1.5^3$	stable ^{2,3}	$0.9^3 - \text{stable}^2$
$T_{1/2}$	1 - 10 h	stable	4 h - stable
SF	≈ 200 d	2 - 200 μ s	≈ 0.2 s

The even-mass isotopes of mendelevium, like those of einsteinium, are expected to exhibit isomerism due to combination of the 101st proton (assigned as the 7/2-(514) proton state) with low-lying, high-spin neutron states such as 7/2+(613) and 9/2+(615). Isomers are currently known for ^{254}Md and ^{258}Md . The 55-d alpha-emitting isomer of ^{258}Md is believed to be the ground state and has been given an 8- assignment; no e.c. decay⁴ was observed. It is most likely composed of the low-lying 7/2-(514) proton and the 9/2+(615) neutron states. A 43-m isomer of ^{258}Md has been produced⁵ by the (α ,n) reaction on ^{255}Es and is believed to decay by e.c. capture. It is probably the 1- level formed by the combination of the same two single particle states and should be excited by perhaps 100 keV over the ground state.

A high-spin (7-, 8-) isomer of ^{256}Md , resulting from the coupling of the 7/2-(514) \downarrow proton with the 7/2+(613) \uparrow or possibly the 9/2+(615) \downarrow neutron states, should also be expected. The decay energy of the 7- state can be estimated to be ≈ 100 keV more than for the known 76-m ^{256}Md ground state (0-) which decays primarily by e.c. capture, but it probably has a considerably

longer e.c. half life due to the necessity for decay to a high-spin state in the ^{256}Fm daughter, such as the known 6+ level at 332 keV or the 8+ level at 563 keV which are populated⁶ by the decay of the 7.6-h high-spin isomer of ^{256}Es . It might also decay by alpha emission to ^{252}Es . A concerted search for this and other isomers of the even-mass Md isotopes should be made.

The production of 43-m ^{258}Md allowed studies⁵ of the SF properties of its 0.380 ms ^{258}Fm daughter to be performed which showed that SF of ^{258}Fm resulted in a narrowly symmetric fragment mass distribution with unusually high total kinetic energy. (See Figs. 1 and 2.) The only other spontaneously fissioning isotope known to exhibit similar properties is 1.5-s ^{259}Fm . (See Figs. 3 and 4.) So far, SF decay of mendelevium has only been observed¹ for 95-m ^{259}Md . Its SF properties have turned out to be quite unusual in that although its fragment mass distribution is highly symmetric, its total kinetic energy of 189 MeV is not anomalously high (see Fig. 4) as is the case for ^{258}Fm and ^{259}Fm . The full width at half maximum of the total kinetic energy distribution of 104 MeV is unusually large compared to those for other spontaneously fissioning nuclides as shown in the summary given in Table 2. Hulet et al.⁹ have suggested that this relatively low total kinetic energy may be due to the emission of a $Z = 1$ particle which then allows the remaining mass to divide into two $Z = 50$, closed-proton shell fragments. If confirmed, this would be another dramatic demonstration of the strong effect exerted on low-energy fission by the fragment shells. Further examination of the fission properties of still heavier Md isotopes would be of particular interest.

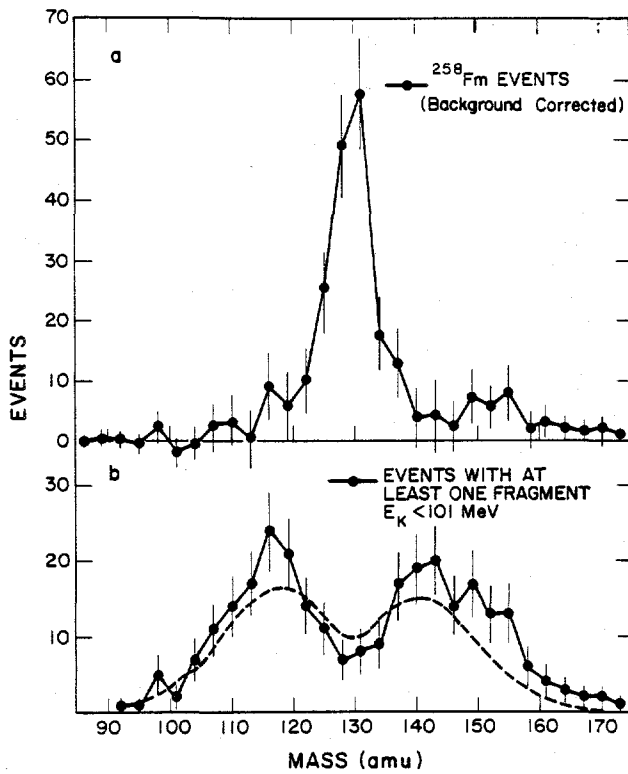


Fig. 1. Mass-yield distributions for SF events from the bombardment of ^{255}Es (Ref. 5). a) Mass distribution of ^{258}Fm resulting after subtraction of an estimated 65% background due to ^{256}Fm . b) Mass distributions of SF events with one fragment kinetic energy < 101 MeV, similar to that for ^{256}Fm shown by the dashed line.

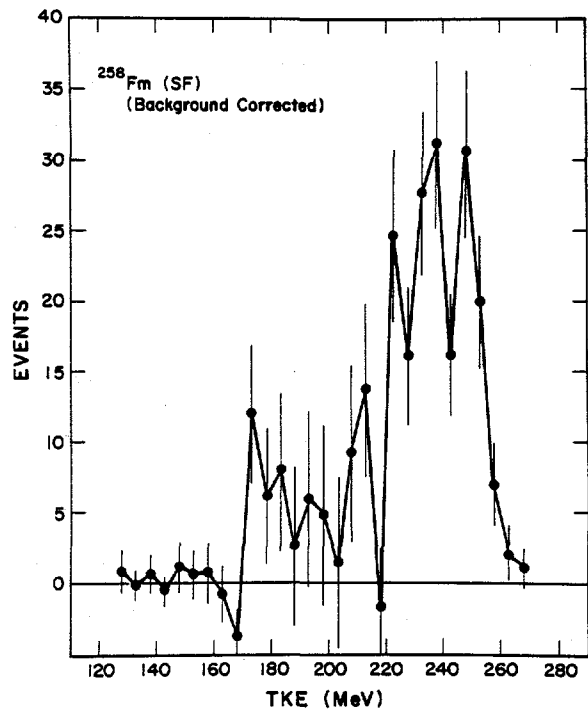


Fig. 2. Total kinetic energy distribution for ^{258}Fm (SF) after subtraction of background from ^{256}Fm (SF).

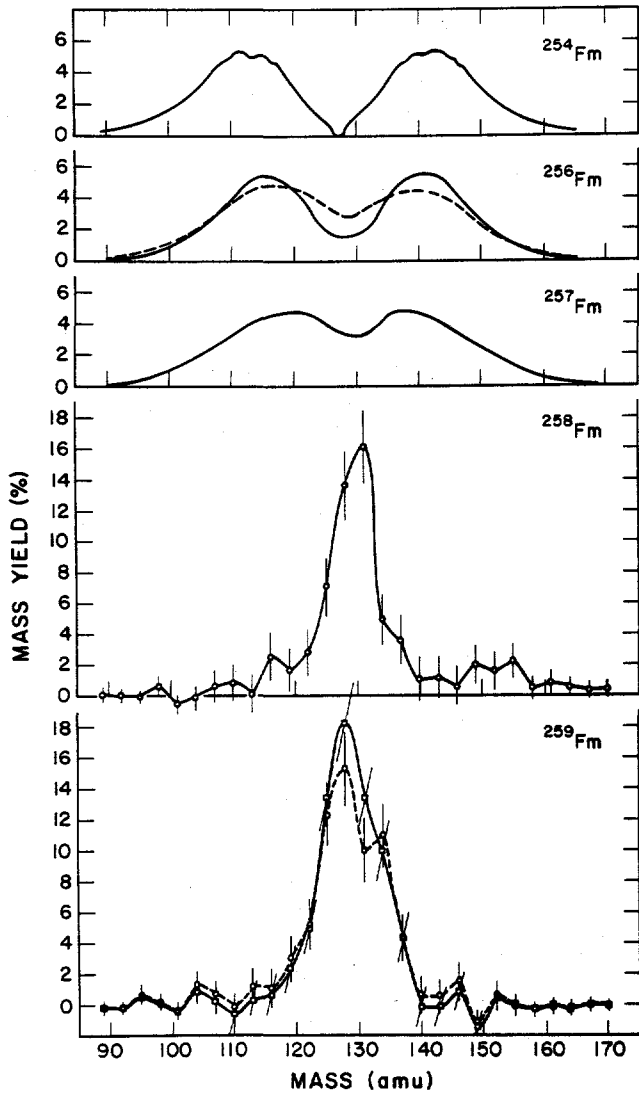


Fig. 3. Mass-yield distributions for SF of Fm isotopes (Ref. 5).

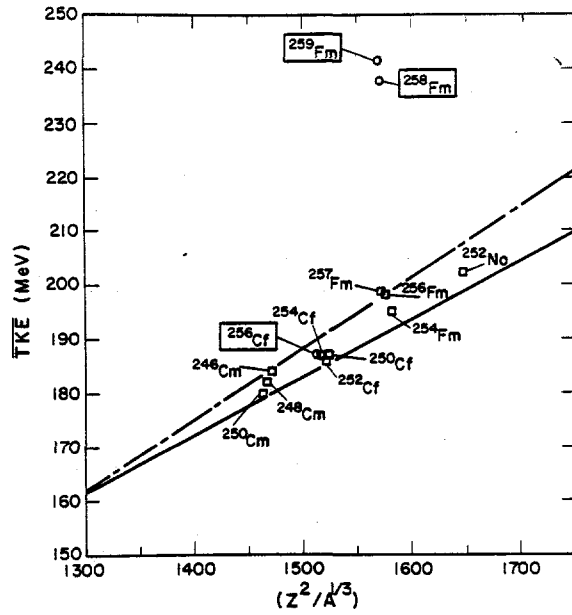


Fig. 4. Total kinetic energy vs. $Z^2/A^{1/3}$ for SF of heavy actinide isotopes. The dashed line is the fit of Unik et al.⁷ and the solid line is the fit of Viola et al.⁸

Table 2. Low energy fission properties of some heavy element isotopes.

Fissioning Nuclide	SF $T_{1/2}$ (Seconds)	Peak-to-Valley Ratio ^a	$\overline{\text{TKE}}$ (MeV)	$\sigma_{\overline{\text{TKE}}}$	$\overline{\nu}_T$
²⁵⁰ Cf	5.4×10^{11}	> 300 (RC)	187.0	11.3	3.49
²⁵⁰ Cf*	-	≥ 50 (RC)	189.1	13.0	-
²⁵² Cf	2.7×10^9	≥ 750 (RC)	185.7	11.6	3.73
²⁵² Cf*	-	≈ 20 (RC)	185	15.5	-
²⁵⁴ Cf	5.2×10^6	≥ 145 (RC)	186.9	11.8	3.89
²⁵⁶ Cf	7.4×10^2	Asymm. (SS)	189.8	14.6	-
²⁵³ Es	2.0×10^{13}	326 (RC)	191	13.4	-
²⁵⁵ Es*	-	≈ 8 (SS)	194.3	15.9	-
²⁴⁶ Fm	1.3×10^0	≥ 10 (SS)	199	14.8	-
²⁴⁸ Fm	3.8×10^1	≥ 10 (SS)	198	14.5	-
²⁵⁴ Fm	2.0×10^7	≈ 42 (RC)	195.1	11.7	3.96
²⁵⁶ Fm	1.0×10^4	12 (SS)	197.9	14.4	3.70
²⁵⁶ Fm*	-	2.5 (RC)	195.5	18	-
²⁵⁷ Fm	4.1×10^9	≈ 1.5 (SS)	197.6	15.3	3.77
²⁵⁸ Fm	3.8×10^{-4}	Symm., $\sigma = 8$ (SS)	238	14	-
²⁵⁸ Fm*	-	Symm., Broad (SS)	197	-	-
²⁵⁹ Fm	1.5×10^0	Symm., $\sigma = 11$ (SS)	242	21	-
²⁵⁹ Md	5.7×10^3	Symm., $\sigma = 13$ (SS)	189	44	-
²⁵² No	8.6×10^0	Asymm. (SS)	202.4	15.4	4.15

*Thermal-neutron induced fission

^aRC indicates radiochemical determinations;
SS indicates solid-state measurements.

The SF half-life of 95-m for ^{259}Md is unusually long compared to those for the even-even isotopes. For example, ^{258}Fm which has the same number of neutrons has a half-life of only 0.380 ms. The hindrance due to the odd proton, 7/2-(514), in Md is apparently sufficient to lengthen the half-life for ^{259}Md by more than 10^7 relative to ^{258}Fm . Such hindrances due to specific odd-proton or odd-neutron single particle states have been known for some time and have been discussed in detail by Randrup et al.¹⁰ The hindrance is typically of the order of 10^5 , but can be as small as 10 and as large as 10^{10} . If the 101st proton provides ^{260}Md with the same hindrance relative to ^{259}Fm (1.5 s) as for ^{259}Md relative to ^{258}Fm , then its SF half-life would be of the order of 200 d. It would then be expected to decay predominantly by beta or e.c. emission with a half-life of a few hours, depending on the decay energy, as discussed earlier. This would provide a means for studying the SF decay of the very short-lived ^{260}No daughter which has the same number of neutrons as ^{258}Fm and afford another assessment of the effect on the fission process of protons beyond $Z = 100$. ^{256}Cf , also having 158 neutrons but only 98 protons, shows an asymmetric mass distribution and "normal" total kinetic energy in contrast to the SF of ^{258}Fm . (See Figs. 5 and 6.)

There is no neutron analogue for ^{261}Md in the Fm isotopes from which to scale its possible fission half-life, but if we use the reduction in half-life of ^{258}Fm relative to ^{256}Fm of 4×10^{-8} for the addition of two neutrons, to scale the 95-m half-life of ^{259}Md , then a half-life of 0.2 ms might be expected for ^{261}Md . Using the reduction in half-life between ^{257}Fm and ^{259}Fm would give a still shorter estimate of only 2 μs for ^{261}Md so studying its SF properties will be extremely difficult. However, ^{262}Md , which might have an SF half-life from 0.5 ms to 0.7s could furnish a still more neutron-rich

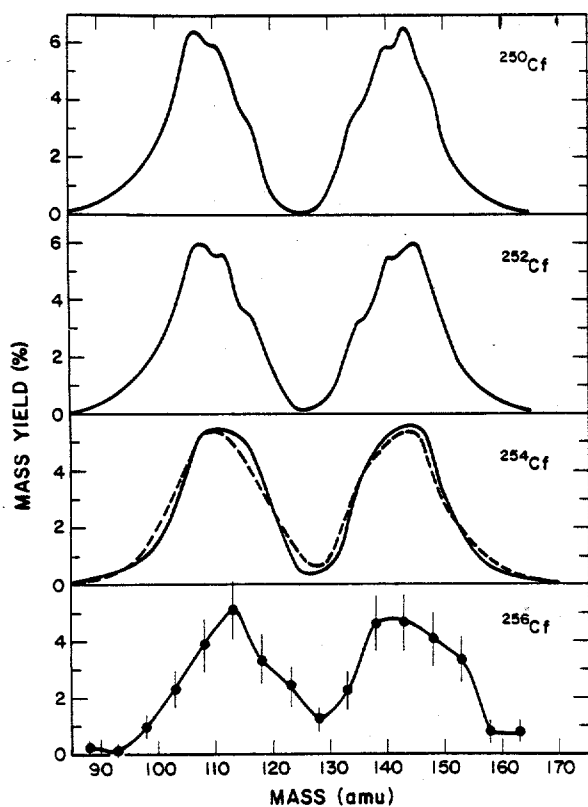


Fig. 5. Pre-neutron emission mass-yield distributions for SF of Cf isotopes (Ref. 5).

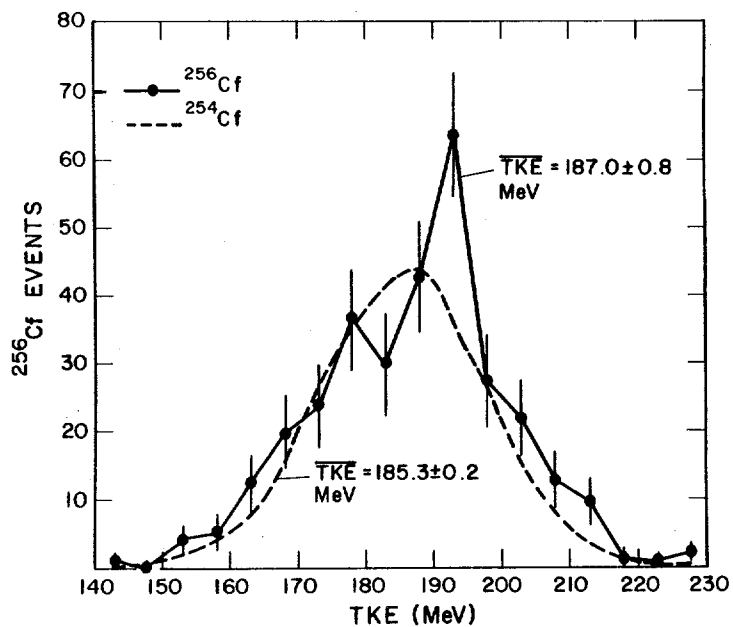


Fig. 6. Total kinetic energy distributions for SF of ^{254}Cf and ^{256}Cf with no correction for neutron emission. The average pre-neutron emission total kinetic energy is 188.8 ± 0.9 MeV for ^{256}Cf after correction for a long-lived background from ^{254}Cf and for neutron emission.

(N = 161) nuclide for study. This nuclide would be of particular interest since it could fission symmetrically into nuclei with more nearly the N = 82 closed-neutron shell configuration, but which would have an extra proton over two Z = 50 closed-shell fragments. Although studies of the SF properties of the heaviest Md isotopes will obviously be most difficult and challenging, they are critical in assessing the relative importance of the proton and neutron shell structure of the fragments on the fission process.

In summary, the heavy Fm isotopes (^{258}Fm and ^{259}Fm) so far appear to be unique in exhibiting very symmetric mass distributions and anomalously high fragment total kinetic energies. These effects appear to be associated with the Z = 100 proton configuration of the fissioning nuclide which can fission symmetrically into two fragments having the Z = 50 closed-proton shell configuration; ^{256}Cf which has the same number of neutrons but only 98 protons does not exhibit these fission properties. Furthermore, ^{259}Md , also having 158 neutrons but with 101 protons, fissions symmetrically but with a "normal" total kinetic energy. However, the total kinetic-energy distribution is extremely broad, perhaps indicating a range of fragment shapes at scission from spherical to highly distorted. It is extremely important to measure the properties of more nuclides with Z greater than 100 and N \geq 158 to check the relative importance of the fragment proton and neutron shells. New methods are needed for measurements of half lives of milliseconds or less and for providing positive identification of the Z and A of the fissioning species. Recent experiments¹¹ indicate that ^{259}Fm is produced with a 10 to 15 nb cross section in bombardments of ^{248}Cm with ^{18}O and that therefore extreme caution must be exercised in making assignments based on the assumption of compound nucleus formation. However, these direct transfer or deep inelastic reactions

appear to offer a good possibility for making the heavier isotopes of Md, No, and Lr for study by bombarding Bk or Es targets with ^{18}O . Such studies will be of utmost importance in furthering our attempts to understand the fission process.

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Introduction of Arnold M. Friedman

Dr. Seaborg

Our next speaker is Arnold M. Friedman. Arnie got his Ph.D. with Joe Kennedy at Washington University in St. Louis in 1953. I have an indirect connection with Arnie. Although Joe Kennedy didn't get his Ph.D. with me in a formal sense, he worked with me and I made the suggestion for his Ph.D. thesis problem. It was during the period when I was serving as personal research assistant to Gilbert Newton Lewis, and was not yet actually on the faculty here at Berkeley. So, in a sense, Arnie is one of my Ph.D. students once removed, or grand-student.

Arnie began to work, as soon as he received his Ph.D., at the Argonne National Laboratory. He was a member of the team from Argonne, along with Paul Fields and others, that was carrying on the investigation (and discovery) of heavy isotopes produced in the neutron bombardment of plutonium and heavier isotopes in the MIR at Arco. This took place at nearly the same time that we were identifying the isotopes that I enumerated earlier--berkelium-249 and so forth, on up to fermium. Arnie and Paul Fields and their co-workers were doing the same thing at the same time at the Argonne National Laboratory. He has continued his work in the heavy isotope region studying the nuclear and radioactive properties of the actinide isotopes and has specialized in this region for the last ten or fifteen years.

Arnie, would you come forward. He will speak on the radioactive properties of mendelevium whereas Darleane spoke on the nuclear properties; I think the line of demarcation is pretty thin.

Radioactive Decay of Md Isotopes*

Arnold M. Friedman

The radioactive decay of the known Md isotopes has two general characteristics which can be related to their nuclear structure. First, all of the even neutron species have a relatively small alpha branching ratio (~10%) and decay primarily by electron capture. Secondly, there are several known isomer pairs, all in odd neutron species. ~~In this paper we will attempt~~ ^{an} ^{is made} to determine the reasons for these empirical observations.

In Fig. 1¹ the proton level structure in the region of Z=101 is shown. The state $\frac{7}{2} - [514]$ is the ground state of the Z=101 species. In Fig. 2² the spectrum of the $^{250}\text{Cf} (\alpha, t) ^{251}\text{Es}$ proton transfer reaction is shown. As we can see the $\frac{7}{2} + [633]$ and $\frac{3}{2} - [521]$ bands are the ground states for the Z=97 and Z=99 species. In the Md isotopes the B_2 deformation apparently has decreased, so that the $\frac{7}{2} - [514]$ band is the ground state. As one can see from Figs. 1 and 2, there is about a 300 KeV energy gap between the $\frac{7}{2} -$ band and the ground state bands of the even-neutron einsteinium isotopes. Therefore, the unhindered alpha decay of even-neutron Md isotopes will always be to a band 300 KeV in excitation, and will consequently have a relatively long half-life. On the other hand, the electron capture decay will not have that energy hindrance and will proceed normally. Therefore, it is not surprising that the alpha branching ratios are small for the even-neutron isotopes.

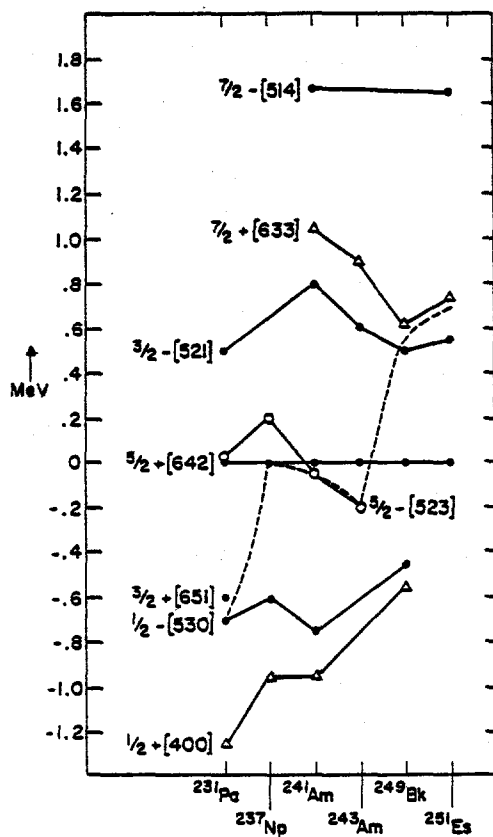


Fig. 1

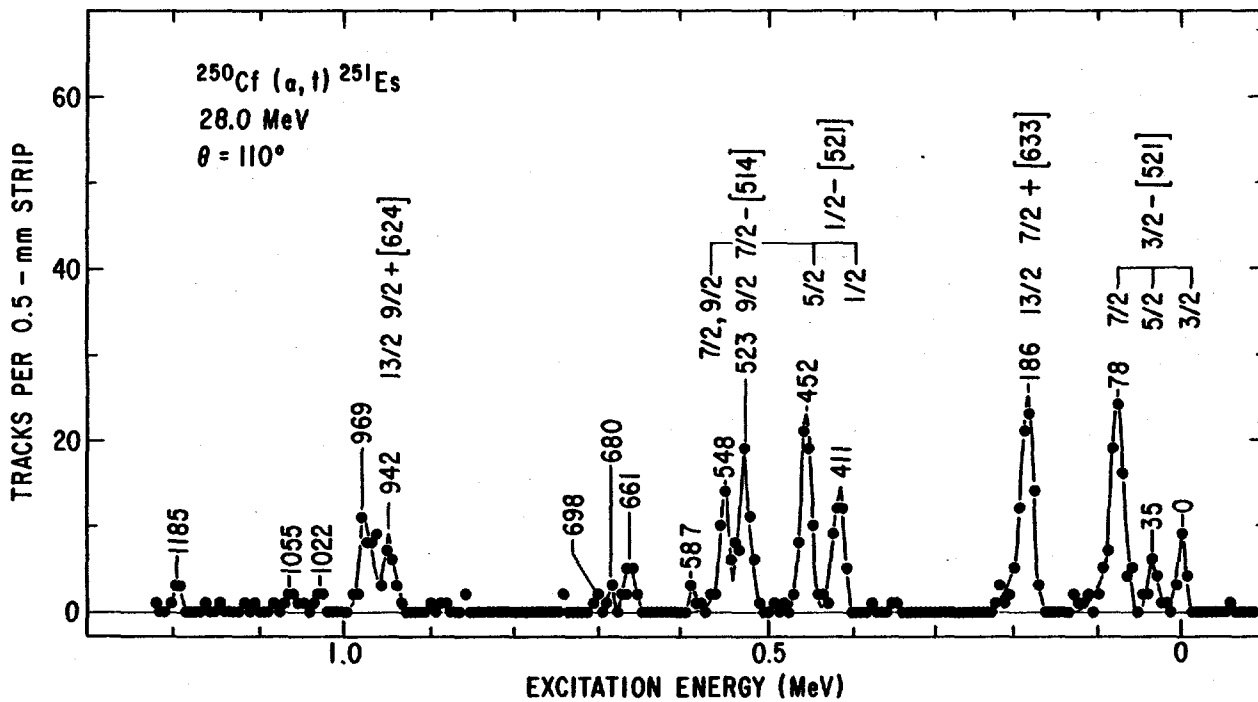


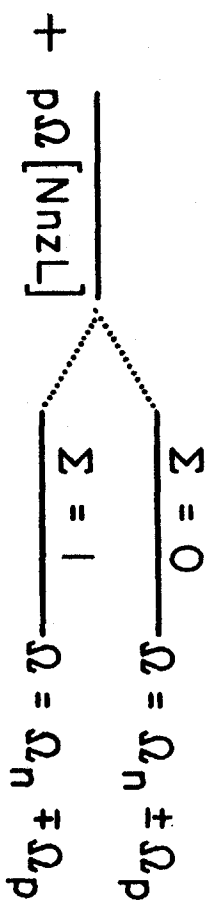
Fig. 2

Odd Neutron Species

The decay of the odd neutron species is characterized by the appearance of several isomer pairs. In order to understand these it may be best to review the manner of addition of two odd particle orbitals in deformed nuclei. Since, in deformed nuclei, there is a common reference axis (the major elliptical nuclear axis) for both particles, the projection of the angular momentum of the particles on this axis will have to add as parallel ($\Sigma=1$) or antiparallel ($\Sigma=0$). In Fig. 3 we can see the sum rules for this addition in deformed nuclei; in the case of odd-odd nuclei they yield the well known Gallagher-Moskowsky rule.

As an example of the above, we see in Fig. 4 the levels observed by Katori and Friedman³ in ^{242}Am by use of the $^{243}\text{Am}(d,t)^{242}\text{Am}$ reaction. These fit into three pairs of rotational bands whose base angular momentum (Ω) values are completely described by $\Sigma=0$ and $\Sigma=1$ bands of either the $\frac{1}{2} + [631]$, $\frac{5}{2} + [622]$ and $\frac{1}{2} - [501]$ neutron adding to the $\frac{5}{2} - [523]$ proton according to our rules.

In addition to the angular momentum values two other characteristics of the $\Sigma=0$ and $\Sigma=1$ bands in odd odd deformed nuclei can be seen in Fig. 3. The energy splittings of the $\Sigma=0$ and $\Sigma=1$ bands can be calculated by use of a variety of particle-particle interactions. These all yield, in general, a splitting of about 75 KeV, which is about the amount observed experimentally. In addition, the bands all occur at an excitation energy close to that of the various single neutron excitation energies in neighboring, isotonic, even proton, odd neutron nuclei. In Fig. 4 the excitation energies of the neutron single particle levels⁴ in ^{241}Pu are shown for comparison with those of the corresponding $\Sigma=1$ and $\Sigma=0$ bands in ^{241}Am .



nΩ [NnzL]

If $\Omega = L + \frac{1}{2}$ ↓

If $\Omega = L - \frac{1}{2}$ ↓

for ↓ p ↓ n or ↓ p ↓ n

$\Sigma = 1$ $\Omega = \Omega_n + \Omega_p$

$\Sigma = 0$ $\Omega = \Omega_n - \Omega_p$

for ↓ p ↓ n

$\Sigma = 1$ $\Omega = \Omega_n - \Omega_p$

Fig. 3

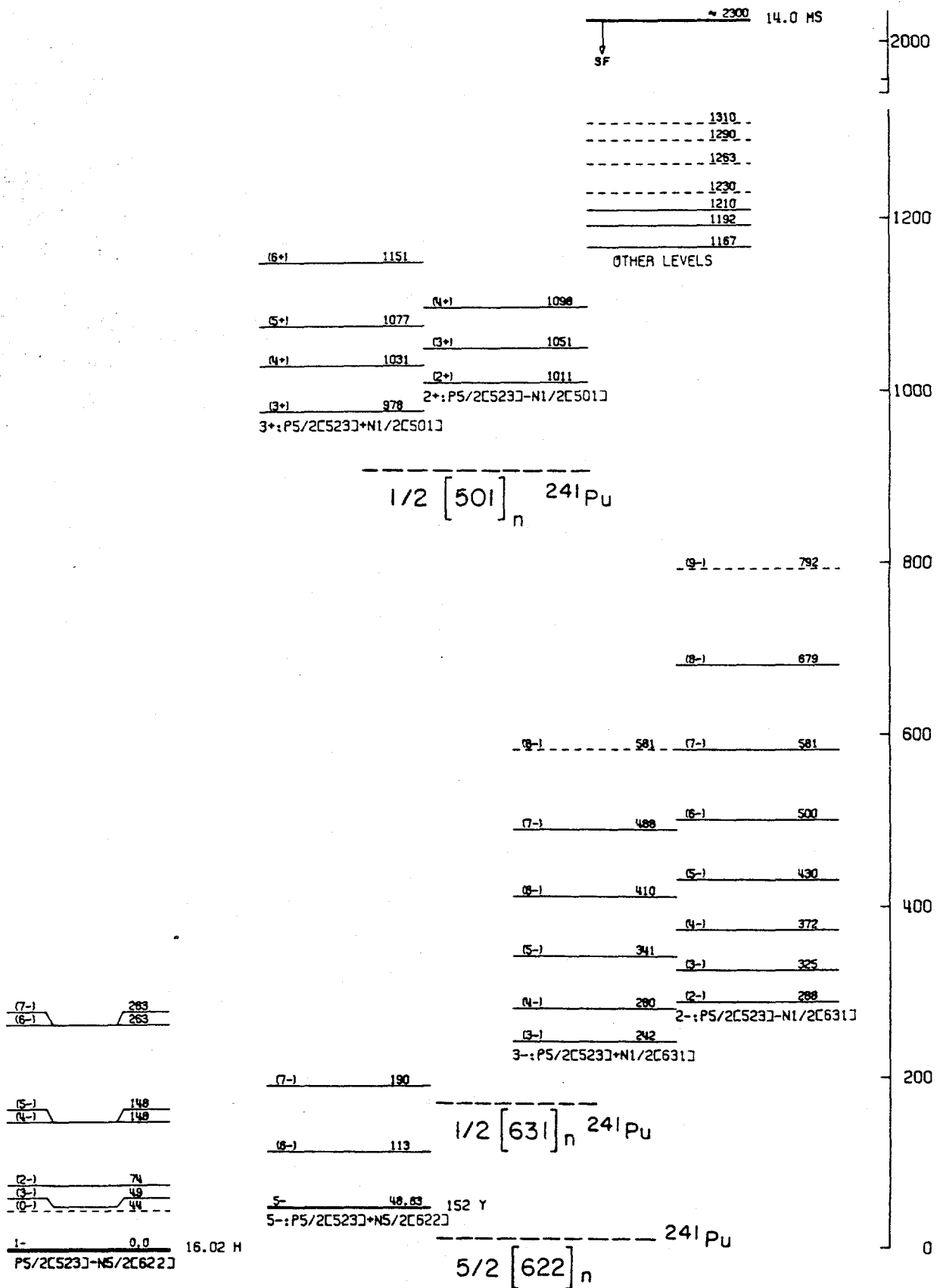


Fig. 4

In the region of 152-158 neutrons we find the neutron levels¹ shown in Fig. 5. At 153, 155, and 157 neutrons, i.e. in ^{254}Md , ^{256}Md , and ^{258}Md , we would expect that the lowest lying neutron levels would be the $\frac{1}{2} + [620]$, $\frac{7}{2} + [613]$ and $\frac{9}{2} + [615]$ levels successively. Therefore, we would expect that these would add to the $\frac{7}{2} - [514]$ proton level to form the $\Sigma=0$ and $\Sigma=1$ bands shown in Fig. 6.

In Fig. 6 we see that ^{254}Md should have two isomers, $\Sigma=1, \Omega\pi=3-$ and $\Sigma=0, \Omega\pi=4-$. Both of these should decay by electron capture rather than alpha emission, and indeed there are two known isomers of ^{254}Md which decay by electron capture with half-lives of 10 minutes and 20 minutes.

We note that for 157 neutrons, i.e. in ^{258}Md , the lowest lying neutron species is the $\frac{9}{2} + [615]$ which should form the $\Sigma=1, \Omega\pi=8-$ and $\Sigma=0, \Omega\pi=1-$ pair. The 8- isomer is the species found by Ahmad et al.² which decays by alpha emission and has a half-life of 57 days. The 1- isomer is the species, recently found by the Los Alamos group, that decays by electron capture and has a half-life of 60 minutes.

In the case of ^{256}Md (155 neutrons) the $\frac{7}{2} + [613]$ neutron adds to the $\frac{7}{2} - [514]$ proton and we would expect to have a $\Sigma=1, \Omega\pi=0-$ and $\Sigma=0, \Omega\pi=7-$ isomeric pair. The $\Sigma=1$ band is known and its alpha decay (10%) was characterized by Ahmad et al. as seen in Fig. 7. It is seen in the figure that the unhindered alpha decay proceeds to a $\Omega\pi=0-$ pair band in ^{252}Es as would be predicted by this assignment. To date, the $\Sigma=0, \Omega\pi=7-$ isomer has not been discovered. However, it should exist and should have an alpha decay half-life of at least several days. As we have seen, the decay characteristics of the Md isotopes can be readily related to the ≈ 300 KeV energy gap in the single particle

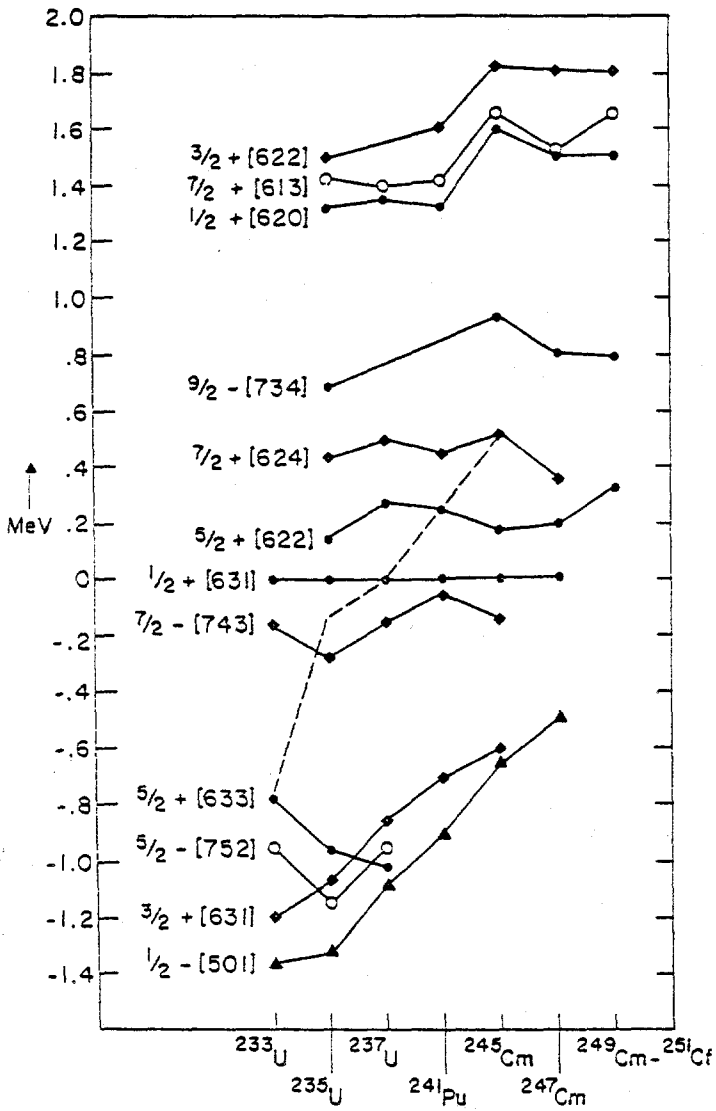


Fig. 5

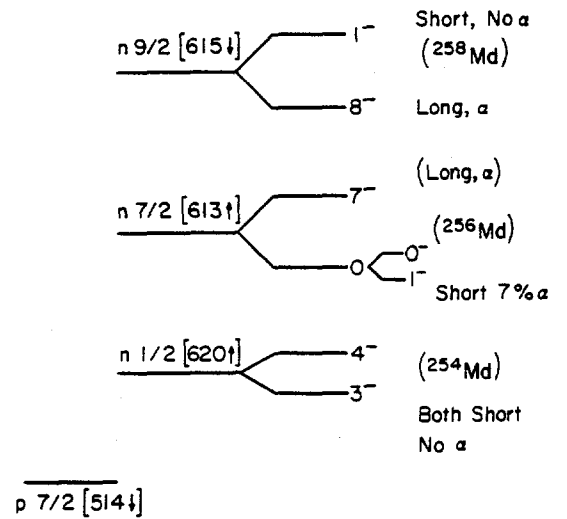


Fig. 6

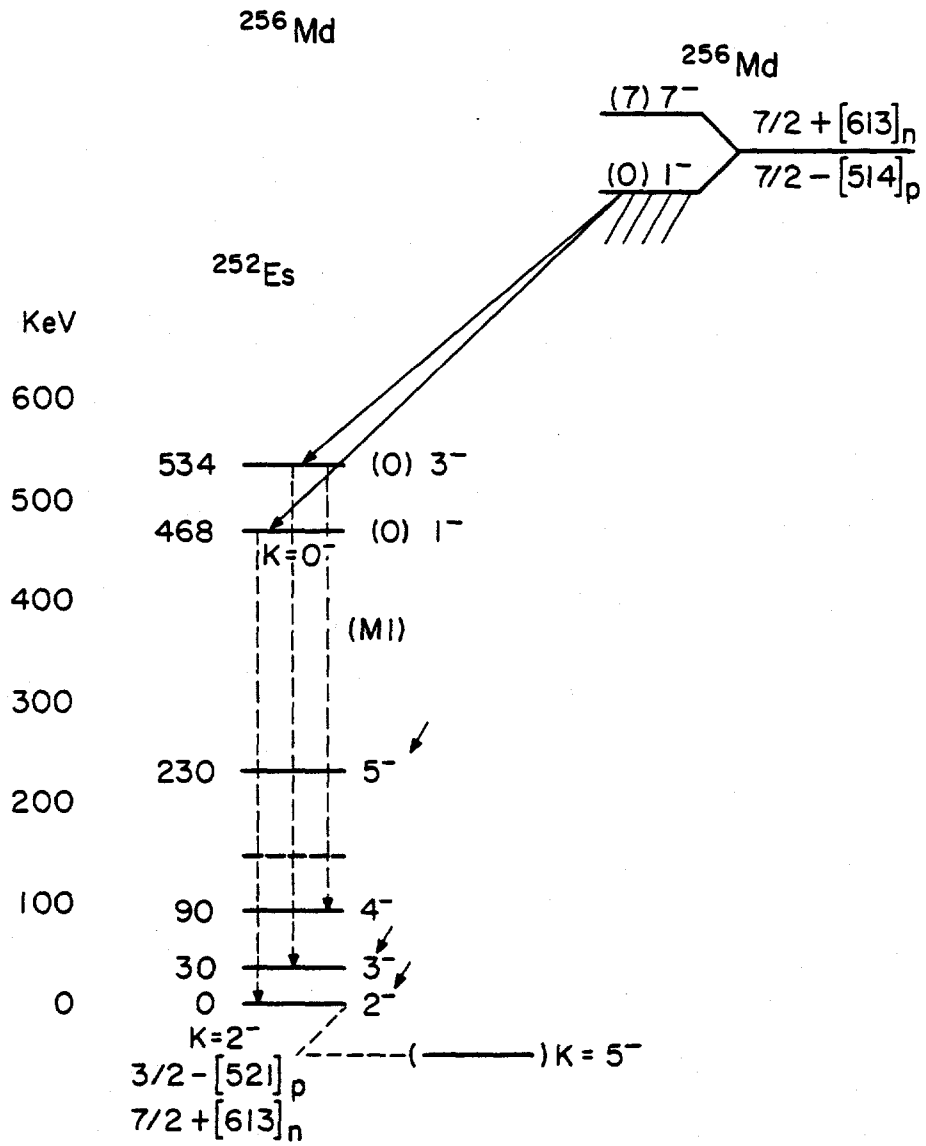


Fig. 7

proton energy spacings and to the high angular momentum of the lowest lying ($\frac{7}{2} - [514]$) proton orbital.

It may be of interest to note, as can be seen in Figs. 1 and 2, that a similar situation will occur in the $Z=105$ isotopes, where the $\frac{9}{2} + [615]$ proton is the lowest lying state. In the case of the $N=155$ and 157 species, mass 260 and mass 262 should have isomeric states which have long half-lives for alpha emission. However, these species may well decay primarily by spontaneous fission.

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* Work performed under the auspices of the Office of Basic Energy Services, Division of Nuclear Sciences, U.S. Department of Energy.

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Concluding Remarks

Dr. Seaborg

This brings us to the end of the 25th Anniversary of mendeleevium. I think, in summary, that mendeleevium is a very interesting element. It is the first element that has been discovered and produced on a one atom at a time basis. The techniques worked out for mendeleevium have served, broadly speaking, as a model for the first synthesis and identification of elements beyond mendeleevium--the recoil technique, the one atom at a time chemistry, and so forth. It is probably the first element in the sequence going up that won't be possible to isolate in weighable amounts. That may be too strong a statement if I am talking about forever. I realize that fermium has not yet been isolated in weighable amounts, but the potential is clearly there because you can make fermium-257 by neutron irradiation, so I think it is just a matter of time until somebody does that. Mendeleevium-258 is sufficiently long-lived to isolate in weighable amounts, but you cannot make it by neutron irradiation and the yield by any other nuclear transmutation reaction is very small. I think the yields are too small for us to contemplate doing that now. I do not know how it might be a hundred or thousand years from now, but now it looks like it will not be possible to isolate mendeleevium in weighable amounts.

Mendeleevium is an interesting element from the standpoint of its chemistry. It is the first element, as you proceed up the actinide series, where the dipositive state plays an important role. It has a lot of interesting nuclear properties; perhaps it isn't distinguished so much from its neighbors in that aspect, however. It was the first element, the first of the tran-

suranium elements, that had an important political impact. It made an actual contribution to the bettering of relations between the Soviet Union and the United States.

So I think, all in all, it is a very interesting element and one that is worth paying some attention to as we have today. And with that, the symposium draws to a conclusion and thank you all very much for coming.