Title
SOME REMINISCENCES OF THE EARLY DAYS OF CALIFORNIUM

Author
Ghiorso, A.

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Lawrence Berkeley Laboratory
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A. Ghiorso

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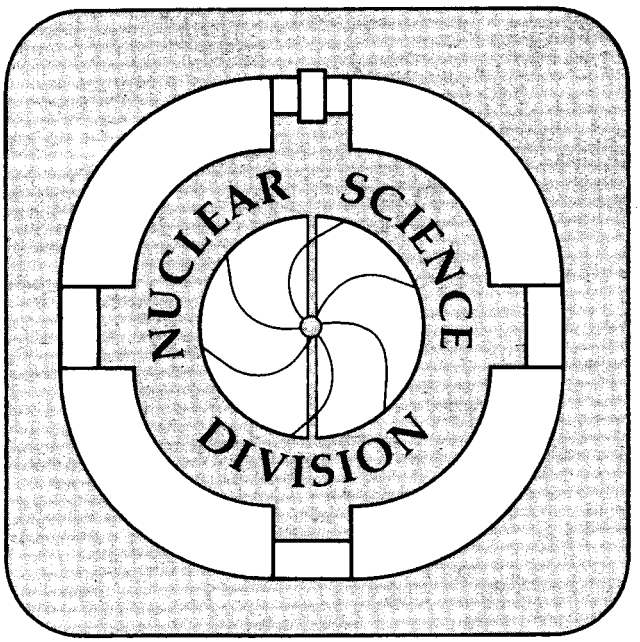

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DISCLAIMER

SOME REMINISCENCES OF THE EARLY DAYS OF CALIFORNIUM*

ALBERT GHIORSO
Lawrence Berkeley Laboratory, University of California,
Berkeley, California 94702

Today we take for granted the availability of ^{252}Cf which, due to the marvelous ingenuity of Ron Macfarlane, is used to produce the complicated spectra from bio-molecules. But it was not always so. Today I would like to take you back more than 30 years to the time when ^{252}Cf was discovered.

On 4 December 1952 Glenn Seaborg received a curious teletype message from James Beckerly, the Director of the Office of Classification in Washington:

> "Radiochemical data on recent Eniwetok test indicates presence of some unique heavy element isotopes such as Pu-244. We do not want to release any information on the properties of these isotopes, even their existence, at this time even though information is declassifiable under guide. Accordingly you are requested to withhold publication of any information on the existence and properties of isotopes present in debris samples and consider such information as secret, restricted data. This prohibition applies even when information is dissociated from test. Please inform those in laboratory who might have access to these data."

Glenn showed the teletype to the late Stan Thompson and me and we puzzled together for a while about what the message meant. Obviously something very startling had happened to produce the ^{244}Pu isotope in one fell swoop in the Eniwetok ("Mike") hydrogen bomb test explosion of 1 November 1952 (Fig. 1). Since we were not connected in any way with the bomb test program we had no way of finding out the details of what had happened. For a number of years Stan had been bombarding ^{239}Pu enclosed in "napkin rings" in the Materials Testing Reactor (MTR) near Idaho Falls, Utah in the hope of producing heavier nuclides by slow neutron capture reactions coupled with beta decay. So far we had not gotten beyond ^{242}Pu. What had happened and what did it mean for the future of our research program? Somehow we had to get involved or we would be left far behind.

By the next morning I had come up with the wildest idea of my career and I got together with Stan to try it out on him. First I assumed that ^{238}U was

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*Dedicated to Prof. R. D. MacFarlane on the occassion of his 50th birthday and presented at a symposium held in his honor at College Station, TX USA 15-18 May 1983.

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Fig. 1: A view of the Mike Explosion

present in the bomb as a major component and that somehow in a fraction of a microsecond it would be subjected to so many neutrons during the enormous explosion (15 Megatons!) that it would successively capture many of them to produce extremely heavy uranium isotopes. These would be highly beta unstable and thus quickly decay to elements with higher atomic number such as ^{244}Pu. I assumed that the yields of such nuclides as a function of mass number would vary logarithmically downward as shown in Fig. 2. To establish the slope of this line I then assumed that the yield of ^{244}U would be 10^{-3} that of ^{239}U. This number was just a guess based on a general knowledge of the sensitivity of mass spectrometry techniques known at that time - they had detected ^{244}Pu with certainty in the presence of ^{239}Pu, I knew, so I picked 10^{-3} as being reasonable. Finally, I assumed that a bomb fraction of about 10^{14} atoms was obtainable. This number was based on my experience gained two or three years before in analyzing some Russian test debris.

By extrapolating this highly speculative line out to mass 254, assuming that as many as 16 neutrons had been captured, I predicted a yield of roughly 10^{-8} of that of mass 239 and visualized these ultra-heavy atoms decaying all the way up to element 100! The heaviest element known at this time was element 98, californium, with the neutron deficient isotopes 244, 245, and

246. A reasonable prediction based on our systematics for the nuclide $^{254}100$ seemed to be that it would be an alpha emitter with a half-life of about a month. With such a half-life we would get an activity of roughly one alpha per minute even though the explosion had occurred more than a month earlier.

Thompson was immediately enthusiastic about this unlikely scenario and we went to Glenn to talk about it. He thought that the idea was too fantastic to

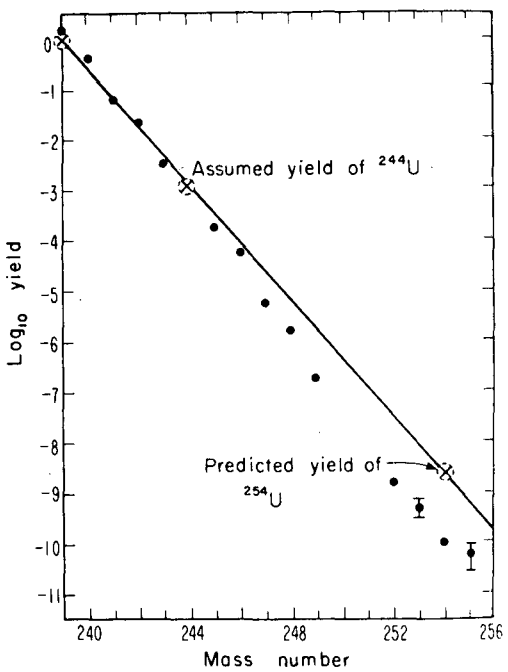

Fig. 2: Yield prediction for ^{254}U

be worth considering. Both he and Perlman thought that it was even highly unlikely that uranium would bind neutrons out that far and that any effort would be futile. But Stan and I were young and not to be deterred, fortunately, so we plunged ahead - just in case!

The new Livermore Laboratory was just being set up at this time with Ken Street heading its Nuclear Chemistry Department. Street had worked with us on the discoveries of elements 97 and 98 just a couple of years before so Stan had no difficulty in enlisting his cooperation. In order for Ken's group to practice their bomb analysis techniques they had received one of the filter papers that had been flown through Mike's mushroom cloud and Street was willing to let us have half of it immediately for our exploration.

With the helpful advice of Bill Crane who had just transferred from Stan's group to Street's, Thompson and Garry Higgins set to work (around the clock) to separate out chemically suitable fractions which would contain any actinide elements with atomic number higher than 94, plutonium. Two days after their strenuous efforts had been completed I started to analyze samples from their cation elution column using the same Frisch-gridded ionization chamber and 48-channel pulse analyzer that we had used to discover the alpha emissions from elements 97 and 98.

To our astonishment there was about a count per minute of an activity that we had never seen before. It had an alpha particle energy of 6.6 MeV and it eluted just ahead of a 6.1-MeV activity which we assumed was due to a new californium isotope. Clearly we were looking at the emissions from a new element. Whether it was due to element 99 or element 100 could not be determined chemically from this particular experiment because the elution column had not run as well as it normally did. We only had one detection chamber so we concentrated our counting on the 6.6 MeV fraction. We soon found that a small number of spontaneous fissions (SF) were also being emitted as well as alpha particles!

At that time SF activity was extremely rare and I remember going to a lot of trouble to prove (by means of absorption in thin foils) that these events were indeed due to fission. Immediately we jumped to the reasonable conclusion that the 6.6 MeV activity must be due to element 100 because we did not expect an odd Z element to have a prominent SF-decay branch. Thus it was that we temporarily assumed that our find must be my predicted $^{254}100$. And this was only eight days after I had proposed the wild idea!

But it was not to be quite so straightforward as that. We procured some more material from Tracerlab, a commercial firm which had a contract to work on weapons diagnostic problems, and made some new separations. This time we made some ^{246}Cf in the 60" Cyclotron and added it to the material so that we would have an unambiguous tracer for californium. By 20 December 1952 we had shown that the 6.6 MeV activity was due to the new element, 99 not 100, since it eluted like its Rare Earth homolog, holmium, rather than erbium. But what about the SF activity?

The spontaneous fissions were found now to be coming entirely from the Cf fraction - those in the first run were due to a "tailing" of californium into the element 99 fraction because of the imperfectly run column. Element 100 was to be found later when Stan, Gary, and other chemists processed large amounts of coral containing fallout from the huge test so that we could look with greater sensitivity for mass 255.

The new Cf sample was quite interesting, consisting principally of 6.1-MeV alphas and SF activity. We followed the activities for decay and, after a few

months it was obvious that the fraction had several constituents.

In the alpha particle analysis the 6.1-MeV peak remained essentially constant but at 6.6 MeV, the same energy found in the element-99 fraction, a new peak grew into the spectrum. We deduced correctly that ^{253}Cf had been produced in the explosion and was a beta emitter with a half-life of about a month which was decaying to $^{253}99$ with a similar half-life. The SF activity was found to decay with a half-life of about two months and its yield suggested that it must be ^{254}Cf assuming that its principal decay mode was by spontaneous fission. After many months the SF decay curve had flattened out to show a longer-lived component with the same half-life as the 6.1-MeV alphas. We decided on the basis of yield that this activity must be due to ^{252}Cf.

By 1954 we had mapped out the region involving elements 97-100 as it was produced in the Mike explosion. It is amazing how close my intuitive guess concerning the mass yield curve came to reality (Fig. 3). Could the new

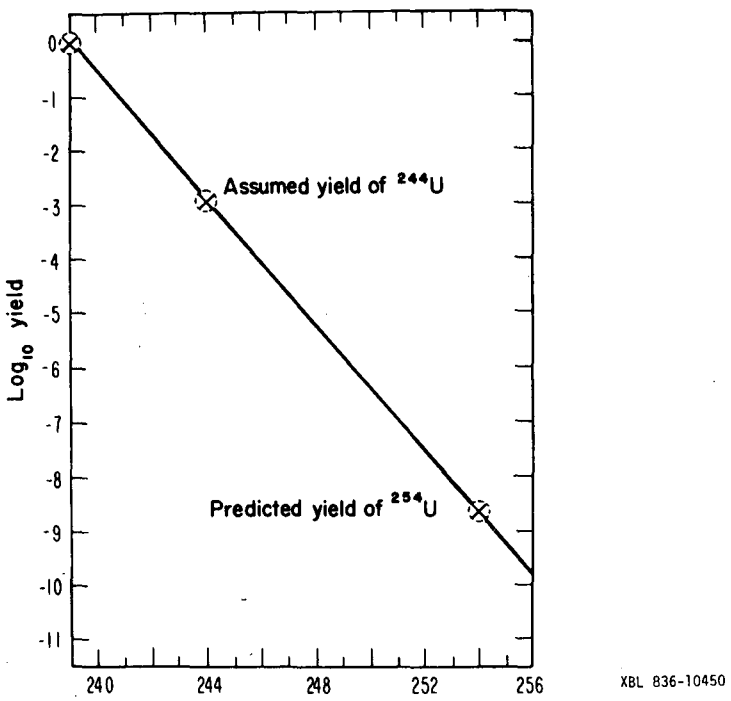

Fig. 3: Comparison of actual with predicted yields

elements be produced with slow neutrons? With the new knowledge and chemical experience gained in these experiments we were able to show that they <u>were</u> produced in the "napkin rings" with a very tiny yield at that time. Another few years would be necessary before the buildup in the neutron reactors of the

heavier masses in the curium isotopes had become sufficient to produce amounts of these elements comparable to that available from Mike (Fig. 4).

In 1957 it became clear to many of us that a cooperative program should be established in the U.S. to produce very large quantities of the heaviest elements -- as much as a gram of ^{252}Cf was envisioned! Glenn T. Seaborg was the key figure in getting this program started so that when he became Chairman of the Atomic Energy Commission in 1961 it was guaranteed that the Transplutonium Program would have the necessary support and guidance. Early

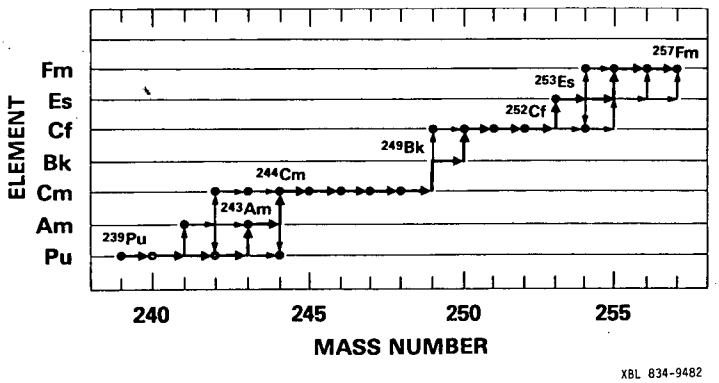

Fig. 4: Slow neutron production of the heavy elements in reactors

along the Transplutonium Committee with which I have been associated since its inception, was set up to help guide the project and allocate the rare and valuable nuclides to the proper scientific channels.

And, to complete the circle, that is how today you scientists can be routinely supplied with californium sources.

ACKNOWLEDGMENT

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UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720