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Dec. 20, 1942

AN ACCOUNT OF THE DISCOVERY  
AND EARLY STUDY OF ELEMENT 94

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The discovery of element  $93^{239}$  as the daughter of  $U^{239}$  produced by the capture of a neutron in  $U^{238}$  was made by E. McMillan and P. H. Abelson. In their published report<sup>1</sup> they described a method of separating this new element from the other known elements based on chemical properties which they had studied using tracer quantities of element 93. The essential feature of their chemical separation was the existence of two oxidation states of 93, the lower having an insoluble fluoride and the other, reached by treating the lower state with a powerful oxidizing agent, bromate ion, being soluble in the presence of hydrofluoric acid.

Classification changed to

by authority of Letter from Dir of Ops. A.E.C. Washington  
B. F. Bell  
on 6-12-58 Date Person making change

In their report McMillan and Abelson pointed out that since their preparations of 93 decayed by beta-particle emission, element 94 was almost certainly present as the decay product. However, they were unable to demonstrate the presence of this 94; that is, no radioactive decay with corresponding observable radiation was found by them.

In June, 1940, E. McMillan bombarded a uranium target with deuterons in the 60-inch cyclotron, and by the method of McMillan and Abelson separated chemically the 93 fraction. By means of an absorption curve he found that its beta-spectrum contained a component of 1 Mev upper energy limit, which is more energetic than the beta-rays from the previously discovered isotope,  $93^{239}$ . His decay curve for this sample indicated a half-life of about 1 day. Also, he found a growth of alpha-particle activity in the sample at a rate not inconsistent with a half-life of roughly 1 day for its parent. He measured

1 E. McMillan and P. H. Abelson, Phys. Rev. 57, 1185-1186, June 15, 1940.

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approximately the range of these alpha-particles; and used the activity as a tracer in a few preliminary studies of the chemical properties of the alpha-radioactive element.

About December, 1940, after McMillan had left Berkeley for M.I.T., G. T. Seaborg wrote to him and obtained permission to go ahead with this work. Then, working with A. G. Wahl (at that time a graduate student in chemistry) and J. W. Kennedy, he obtained another bombardment of deuterons on uranium; the 93 fraction was separated and studied as before. Again the 1 Mev beta-particles were found, but in this case the decay curves (taken with and without absorbers) showed only a 2.3-day half-life. After the beta-activity was largely decayed, alpha-particle radioactivity was found in the sample. Using this alpha-activity as a tracer, some chemical studies were made. It was found that this new substance did not volatilize with  $OsO_4$  in an acid solution containing bromate ion, did not deposit on copper by chemical deposition, did not precipitate with hydrogen sulfide in acid solution, did coprecipitate with cerium fluoride from hydrofluoric acid solution, and was not oxidized by bromate ion in cold sulfuric acid solutions to a state with a soluble fluoride. These few observations showed that this alpha-emitter was chemically different from most of the previously known alpha-active substances (thorium and samarium were not eliminated as possibilities). A preliminary investigation of the range of these new alpha-radiations was made; the range was found to be very close to that of polonium. All this work was done in December, 1940.

The same group of workers got a second uranium plus deuteron bombardment on January 13, 1941. Careful study showed that the energetic beta-component of the 93 fraction decayed with a half-life of 2.1 days (different from the

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2.3 day half-life of  $93^{239}$ ). Moreover, the growth of alpha-particle activity was found to follow a 2 day half-life. It was therefore concluded that this alpha-emitter was a decay product of a new beta-active isotope of element 93 and thus was a new element with atomic number 94. (Approximately a year and a half later this particular isotope of element 94 was identified as  $94^{238}$  by J. W. Kennedy, M. I. Perlman, E. Segre and A. C. Wahl; see report A-207.)

The chemical properties of the new element have been the subject of continuous investigation since its discovery. By the end of February, 1941, the original workers had noted at least two further properties: that  $94$  formed an insoluble iodate, and that  $94$  had a higher oxidation state (reached through the action of peroxydisulfate ion, often called persulfate ion, with silver ion as catalyst) which was soluble in the presence of hydrofluoric acid. This latter property is of special importance in separating it from other elements. A summary of all these results from the deuteron bombardment of uranium is believed to be in the files for future Physical Review publication, in the form of a report signed by Seaborg, McMillan, Wahl and Kennedy.

Meanwhile, the probable preparation of an isotope of  $94$ , namely  $94^{239}$ , by the decay of  $93^{239}$  as reported by McMillan and Abelson had not gone unnoticed. Even though these atoms presumed present in the McMillan and Abelson samples had been observed in no way, on about December 15, 1940, E. Fermi, E. O. Lawrence and E. Segre held a discussion, in Pegram's office at Columbia University, on the possibility of making with a cyclotron enough  $94^{239}$  to measure some of its nuclear properties, and in particular to investigate the question of its fission with slow neutrons in view of its possible application to a chain reaction. This discussion followed some preliminary talks on the subject. Segre returned to Berkeley on January 1, 1941, to

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follow up that suggestion. He talked further with Lawrence, studied McMillan's and Abelson's work and made plans for the experiment. He asked Seaborg to cooperate in the project. On January 24 they wrote to Fermi indicating they were at work.

During approximately this same period Seaborg and Lawrence had received correspondence enclosing a report by Abelson on the usefulness of the  $94^{239}$  in supporting a chain reaction provided it fissioned with thermal neutrons with a favorable cross section, as there were strong theoretical reasons to believe. He wrote in this report (The Thermal Neutron Capture Cross Section of  $93^{239}$  and  $94^{239}$ ) that E. McMillan had been asked by the uranium committee to make measurements on these cross sections. McMillan, however, had been called to M.I.T. and suggested (in a telephone conversation) that Seaborg should "handle the chemical side of the problem...[and] the choosing of a physicist to take care of the physical side of the experiments. He further suggested that Professor Lawrence be personally interested in a supervisory capacity.

In this report, which appears to reflect the opinion of the uranium committee, Abelson writes:

Obviously, the results of these experiments will have a large bearing in the determination of the value of uranium power. It is probable that the cost of isotope separation will be great. The decision to spend perhaps a million dollars on a separation plant may well hinge on the results of these experiments.

With this background of requests for information on the fission cross section of  $94^{239}$ , Seaborg and Segre naturally collaborated in their efforts. On January 11, 1941, Segre wrote to Fermi giving some bombardment yields and asking for some needed uranium. He added that another person would be required for the experiments.

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After a February spent largely in practicing chemical procedures and determining yields, Kennedy was asked to join the group, and about March 1, 1941, the big bombardment of uranium (with neutrons) was made. The 93 fraction from this irradiation was separated by the method of McMillan and Abelson, with an ether extraction step added to remove most of the uranium at the start. This sample (A) in its rare earth carrier was studied and as its beta-activity decayed an alpha-activity was found to grow. This was of course the same isotope of 94 that McMillan and Abelson had prepared in essentially the same way, but now it was available in such quantity that its alpha-emission was observable. This  $94^{239}$  sample (A) was used during April by Seaborg, Segre and Kennedy to establish that the isotope gave fissions with slow neutrons. The sample seemed thick with rare earth carrier; and on May 12 it was given to Wahl to be separated from much of the rare earth by means of the chemical reactions of element 94 which he had learned by use of the other 94 isotope,  $94^{238}$ , as tracer. He "thinned" the sample down to about 200 micrograms of rare earth carrier and mounted it as sample B. This was used for final cross section measurements. Then, on about May 25, 1941, the results of this work, including the fission cross section for slow neutrons and the half-life for alpha-decay, were submitted in the form of a letter to Dr. L. J. Briggs signed by Seaborg, Segre, Kennedy and Lawrence. The body of this letter was issued from Washington as report A-35. At the present time, the letter is in the files of G. Breit awaiting eventual publication in the Physical Review.

During the summer of 1941 a larger sample of  $94^{239}$  was made in the cyclotron and chemically purified and mounted (in rare earth carrier) by Wahl. It was used in some fast neutron fission studies by Seaborg and

Segre (Report A-22). In the fall, this sample was used by Kennedy and Wahl in an investigation on its possible spontaneous fission; their results were reported around December 1, 1941, and appeared in the secret reports as A-68.

In the present year (1942) much larger samples of  $94^{239}$  have been prepared, both in the Berkeley 60-inch cyclotron and in the St. Louis cyclotron. The samples so made were so large that it has been possible to obtain and study them purified and without rare earth or other carrier. This work has been done independently by Wahl in Berkeley and by a group of chemists working under Seaborg in Chicago.

*Eugene L. Segev*

*Edwin M. McMillan*

s/ Joseph W. Kennedy

*Arthur C. Wahl*

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