

Chapter 1

Introduction

1.1. The Pretransuranium Story

The transuranium story began in Italy soon after the discovery of the neutron in England and of artificial radioactivity in France. In their original work in 1934 in Rome, E. Fermi, E. Amaldi, O. D'Agostino, F. Rasetti, and E. Segrè^{1,1} bombarded uranium with neutrons and obtained a series of β -particle-emitting radioactivities. On the basis of the periodic table of that day (Fig. 1.1), they were led to believe that the first transuranium element, with atomic number 93, should be chemically like rhenium (i.e., eka-rhenium, or Eka-Re), element 94

PERIODIC TABLE - BEFORE WORLD WAR II

1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	(43)	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57-71 La-Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	(85)	86 Rn
(87)	88 Ra	89 Ac	90 Th	91 Pa	92 U	(93)	(94)	(95)	(96)	(97)	(98)	(99)	(100)				
		57 La	58 Ce	59 Pr	60 Nd	(61)	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu	

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Fig. 1.1. Pre-World War II periodic table. Parentheses indicate elements undiscovered at that time.

like osmium (Eka-Os), and so forth. Therefore, they assigned a 13-min activity to element 93. To quote from the above-mentioned paper entitled “Possible Production of Elements of Atomic Number Higher than 92”:

“This negative evidence about the identity of the 13-min activity from a large number of heavy elements suggests the possibility that the atomic number of the element may be greater than 92. If it were element 93, it would be chemically homologous with manganese and rhenium. This hypothesis is supported to some extent also by the observed fact that the 13-min activity is carried down by a precipitate of rhenium sulphide insoluble in hydrochloric acid. However, as several elements are easily precipitated in this form, this evidence cannot be considered very strong.”

Soon thereafter, in a paper^{1,2} entitled “*Über das Element 93*” (“On Element 93”), Ida Noddack took issue with this interpretation, suggesting that the radioactivities observed by Fermi *et al.* might be due to elements of medium atomic numbers:

“Es wäre denkbar, das bei der Beschiessung schwerer Kerne mit Neutronen diese Kerne in mehrere grössere Bruchstücke zerfallen, die zwar Isotope bekannter Elemente, aber nicht Nachbarn der bestrahlten Element sind.” [One could think that in the bombardment of heavy nuclei with neutrons these nuclei disintegrate into several larger fragments which, although they are isotopes of known elements, are not neighbors of the irradiated elements.]

However, this paper, which intimated the possibility of the nuclear fission reaction, was too revolutionary and was not taken seriously. In addition, experiments in Germany during the following years by O. Hahn, L. Meitner, and F. Strassmann (Fig. 1.2) appeared to confirm the Italian interpretation and for several years the “transuranium elements” were the subject of much experimental work and



Fig. 1.2. Fritz Strassmann, Lise Meitner, and Otto Hahn (left to right), Mainz, Germany, 1956.

discussion. In a typical paper by Hahn, Meitner, and Strassmann,^{1,3} part of a series they published during 1935–1938, they reported a 16-min ${}_{93}\text{Eka-Re}^{237}$, 2.2-min ${}_{93}\text{Eka-Re}^{239}$, 12-h ${}_{94}\text{Eka-Os}^{237}$, 59-min ${}_{94}\text{Eka-Os}^{239}$, 3-day ${}_{95}\text{Eka-Ir}^{239}$, and 12-h ${}_{96}\text{Eka-Pt}^{239}$.

In 1938, I. Curie and P. Savitch,^{1,4} working in Paris, found a product of 3.5-h half-life that seemed to have the chemical properties of a rare earth, but they failed to give a rational interpretation of this astonishing discovery. Their paper had the title “*Sur la Nature du Radioélément de Période 3.5 Heures Formé dans L’Uranium Irradié par les Neutrons*” (“On the Nature of a Radioactive Element with 3.5-Hour Half-Life Produced in the Neutron Irradiation of Uranium”), and included the following:

“Nous avons montré qu’il se forme dans l’uranium irradié par les neutrons un radioélément de période 3.5 heures dont les propriétés chimiques sont semblables a celles des terres rares. Nous la désignerons ci-dessous par la notation $R_{3.5h}$. $R_{3.5h}$ se sépare nettement de Ac, allant en tête, de fractionnement, alors que Ac va en queue. Il semble donc que ce corps ne puisse être qu’un élément transuraniens possédant des propriétés très différentes de celles des autres éléments transuraniens connus, hypothèse qui soulève des difficultés d’interprétation.” [We have shown that

in the neutron irradiation of uranium a radioactive element with a half-life of 3.5 hours is produced, with chemical properties similar to those of rare earths. In the following we will refer to it as $R_{3.5h}$. $R_{3.5h}$ separates cleanly from Ac by going to the 'head' (beginning) of the fractionation while Ac goes to the 'tail' (end). It seems, therefore, that this species cannot but be a transuranic element having properties very different from those of the other known transuranic elements, a hypothesis that raises interpretational difficulties.]

Then came the breakthrough. Early in 1939, Hahn and Strassmann,^{1,5} on the basis of experiments performed in December 1938, and with interpretive help from Meitner, who had been forced to leave Germany, described experiments in which they had observed barium isotopes as the result of bombardment of uranium with neutrons. This historic paper had the title "*Über den Nachweis und das Verhalten der bei der Bestrahlung des Urans mittels Neutronen entstehenden Erdalkalimetalle*" ("On the Identification and the Behavior of Rare Earth Metals Produced in the Neutron Irradiation of Uranium") and contained the following conclusion:

"Als Chemiker müssten wir aus den kurz dargelegten Versuchen das oben gebrachte Schema eigentlich umbenennen und statt Ra, Ac, Th die Symbole Ba, La, Ce einsetzen. Als der Physik in gewisser Weise nahestehende "Kernchemiker" können wir uns zu diesem. allen bisherigen Erfahrungen der Kernphysik widersprechenden, Sprung noch nicht entschliessen. Es könnten doch noch vielleicht eine Reihe seltsamer Zufälle unsere Ergebnisse vorgetäuscht haben." [We, as chemists, based on the briefly described experiments, should rename the above-mentioned scheme and replace Ra, Ac, Th with the symbols Ba, La, Ce. As nuclear chemists, being in some respects close to physics, we have not yet been able to take this leap, which contradicts all previous experiences in nuclear physics. It could be that a series of strange coincidences could have mimicked our results.]

Subsequent work showed that the radioactivities previously ascribed to transuranium elements are actually due to uranium fission products, and hundreds of radioactive fission products of uranium have since been identified. Thus in early 1939 there were again, as five years earlier, no known transuranium elements. During these five years Seaborg developed an increasing interest in the transuranium situation. When as a graduate student he gave his required annual talk at the College of Chemistry's weekly Research Conference in 1936, he chose the transuranium elements as his topic, describing the work of Hahn, Meitner, and Strassmann referred to earlier.

During the two years following Seaborg's seminar talk in 1936 and before the discovery of fission, his interest in the neutron-induced radioactivities in uranium continued unabated and, in fact, increased. He read and reread every article published on the subject. He was puzzled by the situation — both intrigued by the concept of the transuranium interpretation of the experimental results and disturbed by the apparent inconsistencies in their interpretation. He remembers discussing the problem by the hour, with Joseph W. Kennedy, a colleague in research, often in the postmidnight hours of the morning at the old Varsity Coffee Shop on the corner of Telegraph and Bancroft Avenues near the Berkeley campus, where they often went for a cup of coffee and a bite to eat after an evening spent in the laboratory.

Seaborg first learned of the correct interpretation of these experiments, that neutrons split uranium into two large pieces in the fission reaction, at the weekly Monday night seminar on nuclear physics conducted by Ernest O. Lawrence in Le Conte Hall. On this exciting night, January 30, 1939, they heard the news from Germany of Hahn and Strassmann's beautiful chemical experiments. He recalls that at first the fission interpretation was greeted with some skepticism by a number of those present, but, as a chemist with a particular appreciation for Hahn and Strassmann's experiments he felt that this interpretation just had to be accepted. He remembers walking the streets of Berkeley for hours after this seminar in a

combined state of exhilaration in appreciation of the beauty of the work and of disgust at his inability to arrive at this interpretation despite his years of contemplation on the subject.

In investigations by Edwin M. McMillan^{1.6} at Berkeley and others elsewhere, one of the radioactivities produced in the neutron bombardment of uranium was found to behave differently than the others. This beta radioactivity with a half-life of about two days did not separate by recoil from thin layers of uranium, as did the energetic fission products, when uranium was bombarded with slow neutrons. Segrè, thinking that this was an indication that it might not be a fission product, performed a chemical separation to see if it was a transuranium element *produced* as the daughter of the previously well-known β -particle-emitting ^{239}U . He was unsuccessful and published a Letter to the Editor^{1.7} entitled “An Unsuccessful Search for Transuranic Elements,” identifying the two-day radioactivity as being due to a rare earth element because of his lack of chemical expertise.

1.2. Early Days at the Berkeley Radiation Laboratory

The invention of the cyclotron at Berkeley by Ernest O. Lawrence and M. Stanley Livingston in 1931^{1.8} started a new era in physics by making a giant leap possible with a relatively simple device. Up to this time progress in the production and study of artificial radioactivities was very slow, because although α -emitting sources prepared from natural ores were used initially in the discovery of artificial radioactivity by the Joliot-Curies, the intensities that could be achieved in this manner were very small. The nonresonant accelerators that soon became available provided much current but could not attain sufficiently high energies to do very much in the way of interesting chemistry or physics. The cyclotron, on the other hand, was a relatively simple device and could be constructed inexpensively by physicists anywhere in the world with only modest help from the Berkeley “cyclotroneers.”