THE TRANSURANIUM ELEMENTS

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I. INTRODUCTION

Since the discovery of artificial radioactivity, a new significance has become attached to certain ideas which have persisted in the minds of scientists for a considerable period of time. What is the maximum possible number of elements? Do elements exist beyond uranium in the atomic series? If there are elements beyond uranium, what are their properties? The advent of artificial radioactivity revived research along these lines, which has resulted in the synthesizing of elements of atomic number greater than 92.

The chemical literature contains reports of the detection of transuranium elements based on chemical separations and physical measurements of materials obtained from minerals, none of which has been verified. Conjectures as to the total possible number of elements have been recorded from time to time. Predictions as to the properties of elements beyond uranium have been made by different investigators. Finally, because radioactivity may be induced in atoms, it has been possible to produce certain of the transuranium elements in very minute amounts and to determine some of their properties, but several very important points remain to be solved.

Uranium has been known for a long time, Klaproth having first reported its discovery in 1789. Over a century later, in 1895–96, came the discovery of radioactivity by Becquerel. Investigation of this new property of matter showed that it was exhibited by uranium as well as by several other atomic species. Ultimately the three major disintegration series were distinguished one from the other, and uranium was named as the parent substance of one series. The idea still persisted, however, that perhaps in earlier times there were other radioactive substances now nonexistent, or that perhaps elements exist whose radioactive properties are such that the methods of detection and measurement in use have not been sensitive enough to permit their discovery.

II. CLAIMS REPORTED FOR THE EXISTENCE OF ELEMENT 93 IN MINERALS

The possible existence of element 93, a transuranium element, has been twice reported since the advent of x-ray spectroscopy as a method of analysis. A series of articles by Druce and Loring (25, 56, 57, 58) appeared in 1925 and 1926, reporting their investigations on manganese materials. Primarily they were interested in detecting elements 43 and 75. Before making their spectroscopic analyses it was necessary for them to calculate the characteristic x-ray lines for several of the heavier elements. Using the values given by Siegbahn for the known elements, they determined the characteristic lines for elements 43, 75, 85, 87, 93 and for the radioelements of the different disintegration series. Table 1 gives a summary of these values. In their discussion foreshadowing element 93, they suggested that two very distinct lines observed on two x-ray films might possibly be the $L\alpha_1$ and the $L\beta_1$ lines of the highest member of the manganese group. The observed wave lengths of these lines were 0.895 and 0.693 Å. as compared with the estimated values of 0.8936 and 0.697 Å., respectively. No definite claim for the detection of element 93 was advanced.

In 1934 considerable interest was manifested in an announcement by Odolen Koblic (50, 51, 52, 53), director of the National Uranium and Radium Plant in Joachimsthal, Czechoslovakia, that an element possessing an atomic weight greater than that of uranium had been discovered in Joachimsthal pitchblende. The name "bohemium" and the atomic number 93 were assigned to the newly found element. It was said to have an atomic weight of 240, based on the analysis of its silver salt, $Ag\mathit{93}O_4$. Koblic had reasoned that uranium was not the last member of the atomic series but that an element 93 must exist as the parent substance of protoactinium and the actinium series. Such an element would thus be a congener of manganese and rhenium. Its most stable compounds would be analogous to the perrhenates. $H\mathit{93}O_4$ would thus be a very strong acid. Its sodium salt would be very soluble, whereas the analogous thallous salt would be difficultly soluble. These two facts would permit isolation of the element, if it occurred in a mineral. With this in mind, Koblic treated the material obtained in the decomposition of the pitchblende. The mineral is used normally for the production of uranium and radium preparations. The first stage in breaking it down consists of treatment with soda ash and leaching. Koblic reasoned that the mother liquor from this alkali treatment should contain the element 93, so he evaporated a large quantity of the liquor to a small volume. The crystalline deposit thus secured contained the excess soda ash, saltpeter, and other soluble salts. The clear filtrate was further concentrated to remove additional amounts of soluble salts. The clear mother liquor obtained from this treatment was acidified with nitric acid and then treated with an excess of silver nitrate. The precipitate obtained was removed by filtration, and the clear filtrate treated with thallous nitrate. A red, crystalline precipitate formed, which was the expected $Tl\mathit{93}O_4$. This salt was converted into the more soluble silver salt, of which 115 mg. was obtained. From the

TABLE 1

The characteristic x-ray lines of elements 71 to 93, as calculated by F. H. Loring

(Reprinted from Chem. News **131**, 339 (1925))

1	2	3	4	5	6	7	8	9		10		11		12		13	
ELEMENT	AT. NO.	$\sqrt{\nu/R}$ †	DIFF.	λ Å U $L\alpha_1$ ‡	λ Å U $L\alpha_2$	MEAN α_1 α_2	λ Å U $L\beta_1$	FILM NO. 1.		FILM NO. 2.		FILM NO. 3.		FILM NO. 4.		FILM NO. 5.	
(◆ ———)	93	32·053		0·8877	0·8995	0·8936	0·697							0·895	0·693	0·900	—
URANIUM	92	31·673	·380	0·90833	0·92014												
	91	31·294	·379														
THORIUM	90	30·915	·379	0·95342	0·96524												
	89	30·536	·379														
	88	30·157	·379														
EKA-CÆSIUM	87	29·778	·379	1·0276	1·0394	1·0335	0·837					1·032	—	1·040	—	1·040**	—
	86	29·399	·379														
EKA-IODINE	85	29·020	·379	1·0820	1·0938	1·0879	0·891							1·086	0·895	—	0·900
	84	28·641	·379														
BISMUTH	83	28·262	·379	1·14115	1·1533												
LEAD	82	27·884	·378	1·17202	1·18352												
THALLIUM	81	27·503	·381	1·20971	1·21603												
MERCURY	80	27·125	·378	1·2385	1·2497												
GOLD	79	26·749	·376	1·27355	1·28489												
PLATINUM	78	26·374	·375	1·31008	1·32121												
IRIDIUM	77	25·997	·377	1·34854	1·35939												
OSMIUM	76	25·621	·376	1·38816	1·3982												
DVI-MANGANESE*§	75	25·246	·375	1·4298	1·4408	1·4353	1·2358	1·43	—	1·43	1·233	1·434	1·232			1·428	—
TUNGSTEN	74	24·869	·377	1·47348	1·48452												
TANTALUM	73	24·499	·370	1·51825	1·5294												
HAFNIUM‖	72	24·125	·374	1·56607	1·57704												
LUTECIUM¶	71	23·751	·374	1·61551	1·62636												
* THE HIGHEST MEMBER OF THE MANGANESE GROUP.								OTHER LINES		OTHER LINES		OTHER LINES		OTHER LINES		OTHER LINES	
† ν = FREQUENCY α_1 LINES. RYDBERG'S CONSTANT = 109737 = R.								Mn Kα & β		Cu Kα & β		Cu Kα & β		Cu Kβ		Cu Kα 1·537	
‡ $10^8/[(\sqrt{\nu/R})^2 \times R]$ = WAVE-LENGTH, λ, IN Å UNITS.								Fe Kα & β				Br-Ag Abs.		λ = 1·045		Cu Kβ 1·389	
§ OR RHENIUM.								Cu Kα & β						λ = 0·912		λ = 0·950	
‖ OR CELTIUM (Ct).														λ = 0·763		λ = 0·916 Br Ab	
¶ OR CASSIOPEIUM (Cp).																λ = 0·735	
** WIDE STRONG LINE. Br $K\alpha_2$ = 1·04172. Br $K\alpha_1$ = 1·03768.																λ = 0·651	
←- - - - - - - -→ = SAME VALUE ENTERED TWICE.																	

ratio $Ag\mathit{93}O_4$:AgBr, an atomic weight of about 240 was obtained. Koblic hence assumed that he had been successful in isolating the element 93 which he had predicted, and made his report in the chemical journals. He estimated that "bohemium" occurred to the extent of about 1 per cent in pitchblende, which would be a very large quantity of an element which had escaped detection for such a long period of time.

Subsequent x-ray and arc emission spectroscopic analyses failed to confirm the presence of any new element, but the presence of tungsten was unmistakable. Later tungsten was found in the material prepared by Koblic, the analyses having been made by I. Noddack (67). She reported that the preparation was a mixed salt of silver and thallium vanadates and tungstates in an excess of tungstic acid. The erroneous atomic weight obtained was due to the fact that the silver salt thought to be $Ag\mathit{93}O_4$ was in reality a complex silver tungstate. The unusual behavior of tungstates in acid media may be the reason for the observed reactions.

It is to Koblic's (54, 15) credit that he withdrew his claim for the existence of element 93 in the pitchblende.

III. THE NUMBER OF POSSIBLE ELEMENTS

Some of the discussions which have been reported relative to the total number of possible elements should be considered before reviewing the physical and chemical properties that might be expected for the transuranium elements.

In 1889, in his address on "The Chemical Problems of Today," Victor Meyer (65) stated that in order "to cast a glance upon general chemical studies which lie some years behind us, we must, above all, consider one of the most far-reaching discoveries of our epoch, *the revelation of the natural system of the chemical elements.* We owe this to the far-seeing Demetrius Mendeléeff. By the side of the titanic figure of the Russian scholar we see the Englishman, Newlands, and our own countryman, Lothar Meyer, successfully co-operating in the foundation and the structure of this work. What these men created has since become generally known: *they showed that the properties of the elements are functions of their atomic weights.* Mendeléeff taught us to predict the existence and the properties of chemical elements as yet unknown with a certainty. . . . The natural system has imposed upon us a problem of the greatest significance in the new determination of atomic weights, the numerical values of which are now of increased interest. But numerous other problems are presented by the new system of the elements. Above all, we are at a loss to discern the cause of the inner nexus of the elements as the system offers it. Also by diligent work, the less studied elements must be properly brought within the system. Fortunate circumstances may allow us to discover the numerous elements indicated by the periodic law. Here let us note a peculiar coincidence. We know today about seventy elements, but Mendeléeff's table indicates so far two small periods of seven each and five large ones of seventeen elements respectively. To these must be added hydrogen forming a 'group' in itself.

"By addition of these figures, $2 \times 7 + 5 \times 17 + 1$, we obtain exactly the number 100.

"It is true that no one can say whether the missing elements will really be discovered or if further new periods might not be indicated by which this number, 100, would be exceeded. But as far as positive data are at hand, they indicate exactly the number mentioned and nothing points beyond it. An odd coincidence which seems to ally the number of the existing elements with the number of our fingers."

This prediction of Victor Meyer's has not been exceeded. However, since then has come the discovery of the rare gases and of the radioactive elements. Moseley's law has permitted the determination of the number of rare earth elements that should be expected. The concept of isotopes has also entered the picture. All of these ideas have affected scientific thought, leading ultimately to the development of the Bohr theory and the different wave and quantum mechanics which have been the vogue during the past two decades.

The expounding of the Bohr theory brought forth several reports on the total possible number of elements. This was probably due in part to the new theory, and in part to the fact that investigators, following the discovery of both the electron and radioactivity, had called attention to the possibility that radioactive disintegration might be due to disturbing effects of the electrons around the atom, since ordinary mechanics and electrodynamics failed to account for radioactive decay. On the basis of the quantum postulates and the Bohr atom, Rosseland (71) attempted to show that in uranium the electrons in the first quantum group are located at a very short distance from the nucleus. In other words, for uranium, the constantly increasing attraction by the nucleus and the progressive repulsion set up by the increase in the number of electrons in the atom causes a contraction of the innermost electronic orbits so that they approach, in size, the order of magnitude of the nucleus itself, namely about 1.1^{-13} cm. Any further attraction of the innermost electrons would then cause them to be brought inside the nucleus. Rosseland, using certain of the concepts of the Bohr theory, attempted to show that this relationship existed at atomic number 92.

Another approximate upper limit can be determined by Bohr's method, which depends upon a peculiarity of the relativistic energy levels of the hydrogen atom. The inner electrons of a heavy atom are supposed to be hydrogenic, so the equation

$$1 + \frac{E}{m_0 c^2} = \left(1 + \frac{\alpha^2 Z^2}{[n_r + (k^2 - \alpha^2 Z^2)^{\frac{1}{2}}]^2}\right)^{-\frac{1}{2}}$$

may be derived. In the term $(k^2 - \alpha^2 Z^2)^{\frac{1}{2}}$ of this expression, when the term Z becomes sufficiently large an imaginary number results. At that point, therefore, a stable grouping could no longer persist. When $n = 1$ and $Z = 137$, this condition is fulfilled, indicating that no atom of atomic number greater than 137 can exist.

This method of calculation has been criticized on the basis that modifications of the laws of force must be considered when such small distances are involved. Kossel (55) assumed that the magnetic attraction of two electrons is proportional to r^{-4} and the electrostatic repulsion to r^{-2}. This indicates again that should the K-orbit become too small, an electron might fall into the nucleus, reducing the nuclear charge.

Similar calculations made by Flint and Richardson (37), on the basis of certain considerations of relativistic mechanics, indicated that there is a minimum possible radius for a circular inner electronic orbit, this limit being reached when $Z = 98$.

Erwin Meyer (64) discussed, in 1927, the limits of the periodic system with notations as to the results of other investigators. Using the older concept that electrons were located in spheres around the nucleus and that the radii of the spheres were proportional to the squares of the quantum numbers, he pointed out that the sum of the squares of the numbers from 1 to 6, thus $1 + 4 + 9 + 16 + 25 + 36$, gives the number 91. Then by including also the second electron of helium, which he stated was associated with the hydrogen electron in the innermost sphere, one obtains the number 92 as the limit of the atomic series. Uranium then would have, according to his concept, a nucleus with six surrounding shells. After an explanation of his point of view, he went on to say "that a hyper-uranium or eka-uranium has no place in the periodic system and that the highest atomic number of 92 is no accidental number."

Von Weinberg (79), in an article entitled "On the Origin of the Chemical Elements," stressed the point that the building-up process of the elements proposed by Bohr does not consider uranium as being the natural limit in the series of the chemical elements, but that one can on the contrary predict the properties of elements which are still heavier, should they exist.

Calculations and important contributions have been made by Eddington, Bohr, Sommerfeld, and many other scientists in attempting to solve the puzzle of the possible number of elements. Many of them have found that the number 137 satisfies the conditions for the ultimate breaking down of the atomic system. This number has, as a result, become a magic number in discussions of this problem.

It is seen therefore that the ideas as to the total number of possible elements are variant, the suggested values ranging from 92 to 137. It was the thought that transuranium elements may exist, at least hypothetically, that furnished one of the motives in the attempt to produce transuranium elements by bombardment with particles of high energy.

IV. PROPERTIES OF THE TRANSURANIUM ELEMENTS

A. Possible electronic configurations

With these ideas in mind, perhaps the next logical step is to consider the properties which might be predicted for the transuranium elements. In simple fashion this could be done by studying one of a number of different forms of the periodic classification and predicting properties for the elements. On the other hand, if tables of electronic configurations are studied first, it becomes possible to indicate the most probable structure for the atoms, and hence the most likely manner in which the elements beyond uranium may be arranged in a periodic table.

Naturally no spectroscopic data are available, a fact which also holds true for several of the other elements listed in the natural radioactive series. Any proposed scheme of electronic configuration for elements beyond element 92 must be pure conjecture, but these conjectures should permit a fairly accurate estimate of the properties of the elements.

The first question is that of deciding what might be the atomic number of the next rare gas. After that value has been chosen it is possible to arrange the elements for the series between radon and the "hypothetical rare gas" in various combinations and to guess the most probable arrangement.

One method of estimating the atomic number of the next rare gas is to apply Rydberg's rule, even though there is no definite evidence to show that it might hold for elements beyond uranium. This generalization gives the atomic number of the rare gases as equal to twice the sum of the squares of the ordinal numbers, 1, 2, 3, etc. For helium it is 2×1^2; for neon, $2(1^2 + 2^2)$ or 10; for argon, $2(1^2 + 2^2 + 2^2)$ or 18; and so on, until for radon it is $2(1^2 + 2^2 + 2^2 + 3^2 + 3^2 + 4^2)$ or 86. Should this series continue in this same manner, then the atomic number of the next "rare gas" would be determined by the series $(1^2 + 2^2 + 2^2 + 3^2 + 3^2 + 4^2 + 4^2)$ and would be 118.

A second method of arriving at this same number would be to observe the regularities in the electronic configurations for the rare gases. All of these elements have two electrons in the *K*-orbit. With the exception of helium, all the known rare gases have eight electrons in the outer completed orbit, and eight in the second or *L*-orbit. The other orbits possess either eighteen or thirty-two electrons. Table 2 gives the electronic configurations for the known rare gases as well as for the hypothetical element No. 118. The last value was determined in this instance by assuming that the repetition of 32 would follow for this last rare gas, in exactly the same fashion that 8 and 18 had repeated in the earlier sequences. This indicates, therefore, that the next series in a periodic classification would consist of the same number of elements as the series beginning with cesium.

The next question is to decide how the elements between element 86, radon, and element 118 should be arranged in a periodic classification. The series beginning with cesium consists of thirty-two elements and includes the very interesting group of the rare earth elements. The series beginning with radon likewise would have thirty-two elements. It might also have a new "rare earth" group.

TABLE 2

Electronic configurations of the rare gases

GAS		*K*	*L*	*M*	*N*	*O*	*P*	*Q*
He	2	2						
Ne	10	2	8					
A	18	2	8	8				
Kr	36	2	8	18	8			
Xe	54	2	8	18	18	8		
Rn	86	2	8	18	32	18	8	
?	118	2	8	18	32	32	18	8

In the accompanying tables of electronic configurations (tables 3 and 4), it is seen that transition groups occur with the elements 21 to 28 and 39 to 46, inclusive. In these instances it is obvious that the building-up process is not occurring in the outermost electronic group but rather one shell deeper in the atom. It is also observed that the same sort of change begins to take place beginning with lanthanum, No. 57, and ends with platinum, No. 78, which is a homolog of palladium, No. 46. However, there is one very striking difference in this series of elements. *First*, the series is longer than any of the preceding. *Second*, beginning with cerium, No. 58, the newly added electrons do not take their places in either of the outer two electronic groups, but rather fall into a shell lying deeper within the atom, namely the 4*f* shell. This type of change, in which an inner electronic group is being completed, gives rise to what is known as an "inner transition group." This building up of an inner electronic group as indicated affords an explanation for the very definite similarities in the

TABLE 3 (PART 1)

Electron configurations and term types for the ground states of the elements

(Numbers and symbols in parentheses are uncertain)

ELEMENT	K	L		M			N				O					GROUND TERM
	1s	2s	2p	3s	3p	3d	4s	4p	4d	4f	5s	5p	5d	5f	5g	
1. H	1															$^2S_{1/2}$
2. He	2															$^1S_0$
3. Li	2	1														$^2S_{1/2}$
4. Be	2	2														$^1S_0$
5. B	2	2	1													$^2P_{1/2}$
6. C	2	2	2													$^3P_0$
7. N	2	2	3													$^4S_{3/2}$
8. O	2	2	4													$^3P_2$
9. F	2	2	5													$^2P_{3/2}$
10. Ne	2	2	6													$^1S_0$
11. Na	2	2	6	1												$^2S_{1/2}$
12. Mg	2	2	6	2												$^1S_0$
13. Al	2	2	6	2	1											$^2P_{1/2}$
14. Si	2	2	6	2	2											$^3P_0$
15. P	2	2	6	2	3											$^4S_{3/2}$
16. S	2	2	6	2	4											$^3P_2$
17. Cl	2	2	6	2	5											$^2P_{3/2}$
18. A	2	2	6	2	6											$^1S_0$
19. K	2	2	6	2	6		1									$^2S_{1/2}$
20. Ca	2	2	6	2	6		2									$^1S_0$
21. Sc	2	2	6	2	6	1	2									$^2D_{3/2}$
22. Ti						2	2									$^3F_2$
23. V						3	2									$^4F_{3/2}$
24. Cr						5	1									$^7S_3$
25. Mn						5	2									$^6S_{5/2}$
26. Fe						6	2									$^5D_4$
27. Co						7	2									$^4F_{9/2}$
28. Ni						8	2									$^3F_4$
29. Cu	2	2	6	2	6	10	1									$^2S_{1/2}$
30. Zn							2									$^1S_0$
31. Ga							2	1								$^2P_{1/2}$
32. Ge							2	2								$^3P_0$
33. As							2	3								$^4S_{3/2}$
34. Se							2	4								$^3P_2$
35. Br							2	5								$^2P_{3/2}$
36. Kr							2	6								$^1S_0$
37. Rb	2	2	6	2	6	10	2	6			1					$^2S_{1/2}$
38. Sr	2	2	6	2	6	10	2	6			2					$^1S_0$
39. Y	2	2	6	2	6	10	2	6	1		2					$^2D_{3/2}$
40. Zr									2		2					$^3F_2$
41. Cb									4		1					$^6D_{1/2}$
42. Mo									5		1					$^7S_3$
43. Ma									(5)		(2)					$(^6S_{5/2})$
44. Ru									7		1					$^5F_5$
45. Rh									8		1					$^4F_{9/2}$
46. Pd									10							$^1S_0$
47. Ag	2	2	6	2	6	10	2	6	10		1					$^2S_{1/2}$
48. Cd											2					$^1S_0$
49. In											2	1				$^2P_{1/2}$
50. Sn											2	2				$^3P_0$
51. Sb											2	3				$^4S_{3/2}$
52. Te											2	4				$^3P_2$
53. I											2	5				$^2P_{3/2}$
54. Xe											2	6				$^1S_0$

TABLE 3 (PART 2)

Electron configurations and term types for the ground states of the elements

(Numbers and symbols in parentheses are uncertain)

ELEMENT	K	L	M	N				O					P						Q	GROUND TERM
				4s	4p	4d	4f	5s	5p	5d	5f	5g	6s	6p	6d	6f	6g	6h	7s	
55. Cs	2	8	18	2	6	10		2	6				1							${}^7S_{1/2}$
56. Ba	2	8	18	2	6	10		2	6				2							${}^1S_0$
57. La	2	8	18	2	6	10		2	6	1			2							${}^2D_{3/2}$
58. Ce	2	8	18	2	6	10	(1)	2	6	(1)			(2)							$({}^3H_4)$
59. Pr							(2)	2	6	(1)			(2)							
60. Nd							(3)	2	6	(1)			(2)							
61. Il							(4)	2	6	(1)			(2)							
62. Sm							6	2	6				2							${}^7F_0$
63. Eu							7	2	6				2							${}^8S_{7/2}$
64. Gd							7	2	6	1			2							${}^9D$
65. Tb							(8)	2	6	(1)			(2)							
66. Dy							(9)	2	6	(1)			(2)							
67. Ho							(10)	2	6	(1)			(2)							
68. Er							(11)	2	6	(1)			(2)							
69. Tm							13	2	6				2							${}^2F_{7/2}$
70. Yb							14	2	6				2							${}^1S_0$
71. Lu	2	8	18	2	6	10	14	2	6	1			2							${}^2D_{3/2}$
72. Hf								2	6	2			2							${}^3F_2$
73. Ta								2	6	3			2							${}^4F_{3/2}$
74. W								2	6	4			2							${}^5D_0$
75. Re								2	6	5			2							${}^6S_{5/2}$
76. Os								2	6	6			2							${}^5D^4$
77. Ir								2	6	7			2							${}^4F$
78. Pt								2	6	9			1							${}^3D_3$
79. Au	2	8	18	2	6	10	14	2	6	10			1							${}^2S_{1/2}$
80. Hg													2							${}^1S_0$
81. Tl													2	1						${}^2P_{1/2}$
82. Pb													2	2						${}^3P_0$
83. Bi													2	3						${}^4S_{3/2}$
84. Po													2	4						${}^3P_2$
85. —													2	5						${}^2P_{3/2}$
86. Rn													2	6						${}^1S_0$

chemical and physical properties of the rare earth elements, of which this inner transition group is composed. It explains also the "lanthanide contraction," which in turn may be used to explain the similarity of properties of elements 72 to 78 with those of the elements which are their lower homologs, numbers 40 to 46, respectively.

TABLE 4

A possible electronic configuration for elements 86 to 118

ELEMENT	K	L	M	N				O				P				Q	
				4s	4p	4d	4f	5s	5p	5d	5f	6s	6p	6d	6f	7s	7p
86. Rn	2	8	18	2	6	10	14	2	6	10		2	6				
87.	2	8	18	2	6	10	14	2	6	10		2	6			1	
88. Ra	2	8	18	2	6	10	14	2	6	10		2	6			2	
89. Ac	2	8	18	2	6	10	14	2	6	10		2	6	1		2	
90. Th	↑	↑	↑											2		2	
91. Pa														3		2	
92. U														4		2	
93. E-Re														5		2	
94. E-Os														6		2	
95. E-Ir														7		2	
96. E-Pt														9		1	
97. E-Au												2	6	10		1	
98. E-Hg												2	6	10		2	
99.						32			18		1	2	6	10		2	
100.											2					2	
101.											3					2	
102.											4					2	
103.											5					2	
104.											6					2	
105.											7					2	
106.											8					2	
107.											9					2	
108.											10					2	
109.											11					2	
110.											12					2	
111.											13					2	
112.								2	6	10	14					2	
113.																2	1
114.																2	2
115.																2	3
116.																2	4
117.	↓	↓	↓													2	5
118. E-Rn	2	8	18	2	6	10	14	2	6	10	14	2	6	10		2	6

For the period beginning with element 87, ekacesium, there is again the possibility of a recurrence of a transition group exactly similar to those of the preceding periods. Since the number of elements in this period is postulated as the same for the series beginning with cesium, the important question at this point is whether there will be an "inner transition group" similar to the rare earth group. A definite answer in this case will naturally depend on the energy relationships for the addition of electrons to the various shells, and as yet there is positively no information to establish this point.

Referring to the electronic configuration tables, it is noted that calcium has two electrons in the 4*s* orbit. Magnesium, likewise, has two electrons in its outermost orbit. Should the building-up process continue beyond calcium, as it did in the case of magnesium, the new electrons should drop into the 4*p* shell. However, the energy relationships calculated from spectroscopic data show that the electrons go into the 3*d* shell for the elements scandium to nickel. These elements make up the transition group whose properties are somewhat more similar than those of the other members of this series. The elements gallium to krypton constitute the group where the electrons are again adding to the outer or 4*p* shell, the rare gas configuration being completed with krypton.

In the next period an exactly similar situation exists. The electrons are first added, for rubidium and strontium, to the 5*s* shell. With yttrium, however, the next electron adds to the 4*d* and not to the 5*p* shell. The elements yttrium to palladium make up this second transition group. The 5*p* electrons are added from indium to xenon, which is the rare gas completing this period.

In the next period, exactly the same sort of change starts to take place. For cesium and barium the electrons are added in the outermost or 6*s* group. For lanthanum the energy relationship is such that the third electron being added drops into the 5*d* instead of into the 6*p* group. This is analogous to the preceding groups.

However, with cerium a new type of change occurs. The electron which is being added to the structure in this case does not drop into either the 6*p* or the 5*d* shell but rather deeper into the atom, namely, into the 4*f* shell. This addition of electrons to the 4*f* shell continues until that group is completed with fourteen electrons. With lutecium we have again a structure analogous to that shown by yttrium, in that the innermost orbital groups are all completed, namely, the *K*, *L*, and *M* groups for yttrium and the *K*, *L*, *M*, and *N* groups for lutecium.

The elements cerium to ytterbium are known as an "inner transition" group. The very definite similarities in the chemical and physical properties of the rare earth elements are due to this change in electronic structure which is taking place deeper in the atom, and to the fact that the outermost electronic configurations, which define the valence relationships, are essentially identical. The "lanthanide contraction," often used to explain the similarities in the properties of the elements in this portion of the atomic series, is due to this electronic change occurring in the 4*f* shell rather than in the outer portion of the atom.

Following the rare earth group the changes again take place in the outer orbital groups, as was the case in the previous periods, the normal transition group ending with platinum, the higher homolog of palladium.

With the beginning of the series starting immediately after radon, with ekacesium, No. 87, the question arises relative to the occurrence of an "inner transition" group before the next rare gas. As outlined above, it is assumed that the next rare gas configuration will be attained with element 118. For those energy relationships which have been determined, this period begins in exactly the same fashion as all of the preceding, that is, the first two electrons go into the outermost electronic group, the 7*s* in this instance. The evidence also indicates that the next electrons are added to the 6*d* shell and not to the 7*p* shell. This is analogous to the first two transition periods. This type of structure probably holds as far as uranium, although spectroscopic data for this portion of the atomic series are far from being complete. Therefore, anything that is said about this portion of the table is, to a great extent, mere conjecture.

At this point the question of another "inner transition" group built up in a fashion like the rare earth group must be considered. One should ask whether such a transition group might begin with thorium. This element occupies a position in this last period similar to that of cerium in the preceding group. Likewise one might think such a group could begin with ekarhenium, just following uranium. Perhaps the transition group would occur later in the atomic series, and perhaps not at all.

In the transition groups it is observed that, after two or three electrons become associated with a group, their presence tends to stabilize that particular structure; additional electrons then tend to add to that same orbital group until a completed shell is formed. This is the case with the 3*d* electrons for the group beginning with scandium, the 4*d* electrons following yttrium, and the 4*f* electrons for the rare earth elements. Following actinium (No. 89) and up to uranium (No. 92), the meager amount of spectroscopic data available seems to indicate that the electrons are adding to the 6*d* shell rather than dropping deeper into the atom. Might it not be logical to think that, because of the stabilizing influence of these first three or four electrons, the electrons shall continue to take their places in this 6*d* orbital grouping until it is completely filled? If such should be the case, then a normal transition group would begin with actinium and end with ekaplatinum (No. 96). From this point of view, therefore, consideration of a new "inner transition" group early in this period is eliminated. It does not eliminate the possibility of a second "inner transition" group at a later point in the atomic series. Comparison of tables 3 and 4 shows the very decided difference in the type of structure of these super-heavy atoms with respect to the location of the inner transition or "rare earth type" elements.

Two Bohr–Thomsen tables are given here for comparison. One of these is that proposed by Bohr (table 8), in which it is indicated that an inner transition group analogous to the rare earth group begins with element 94. The other (table 7) has been drawn up according to the above conjecture, which obviously may be entirely wrong, since so little is known of the energy relationships which might be expected in this proposed portion of the atomic series.

The building-up process continues, as indicated in the table of electronic configurations (table 4), so that the 7*s* and 6*d* electrons are added first. Elements 97 and 98 are assigned structures which are analogous to those of gold and mercury. With element 99 the next electron to be added is placed in the 5*f* group. The 6*s*, 6*p*, and 6*d* and 7*s* shells remain as they were. Electrons are added continuously to the 5*f* shell, analogous to the filling in of the 4*f* electrons in the rare earths. Thus an "inner transition group" is being built up, similar to the rare earths in that the electrons are added to an inner electronic group but different in that it is not part of an ordinary transition group which has already been started.

To be sure, the chemical and physical properties of elements 99 to 112 would be very similar, and probably like ekamercury, number 98. Their properties will be summarized later, in a discussion of two different types of the periodic table, one of which will correspond to this type of electronic configuration. With element 113, electrons again should add to the outermost electronic grouping, giving the elements analogous to the series thallium to radon, respectively. This change would end with No. 118, the next (hypothetical) rare gas.

B. Periodic classifications including the transuranium elements

In considering all possible arrangements several periodic tables were prepared, four of which are reproduced here. Two of these (tables 5 and 7) correspond to the electronic configuration proposed above (table 4), whereas the other two (tables 6 and 8) correspond to an arrangement where the "inner transition" group is a portion of a normal transition group. In these last two tables the "inner transition" group begins with ekaosmium ($Z = 94$).

These periodic classifications permit the preparation of a summary of the physical and chemical properties of the elements beyond uranium in

TABLE 5

A periodic classification which corresponds to the electronic configuration given in table 4 (see table 6)

																H 1	He 2
Li 3	Be 4											B 5	C 6	N 7	O 8	F 9	Ne 10
Na 11	Mg 12											Al 13	Si 14	P 15	S 16	Cl 17	A 18
K 19	Ca 20	Sc 21	Ti 22	V 23	Cr 24	Mn 25	Fe 26	Co 27	Ni 28	Cu 29	Zn 30	Ga 31	Ge 32	As 33	Se 34	Br 35	Kr 36
Rb 37	Sr 38	Y 39	Zr 40	Cb 41	Mo 42	Ma 43	Ru 44	Rh 45	Pd 46	Ag 47	Cd 48	In 49	Sn 50	Sb 51	Te 52	I 53	Xe 54
Cs 55	Ba 56	*	Hf 72	Ta 73	W 74	Re 75	Os 76	Ir 77	Pt 78	Au 79	Hg 80	Tl 81	Pb 82	Bi 83	Po 84	Ab 85	Rn 86
V 87	Ra 88	Ac 89	Th 90	Pa 91	U 92	E-Re 93	E-Os 94	E-Ir 95	E-Pt 96	E-Au 97	☒	113	114	115	116	117	118
*Rare Earths:			La 57	Ce 58	Pr 59	Nd 60	Il 61	Sm 62	Eu 63	Gd 64	Tb 65	Dy 66	Ho 67	Er 68	Tm 69	Yb 70	Lu 71
☒	Possible Transition Group		E-Hg 98	99	100	101	102	103	104	105	106	107	108	109	110	111	112

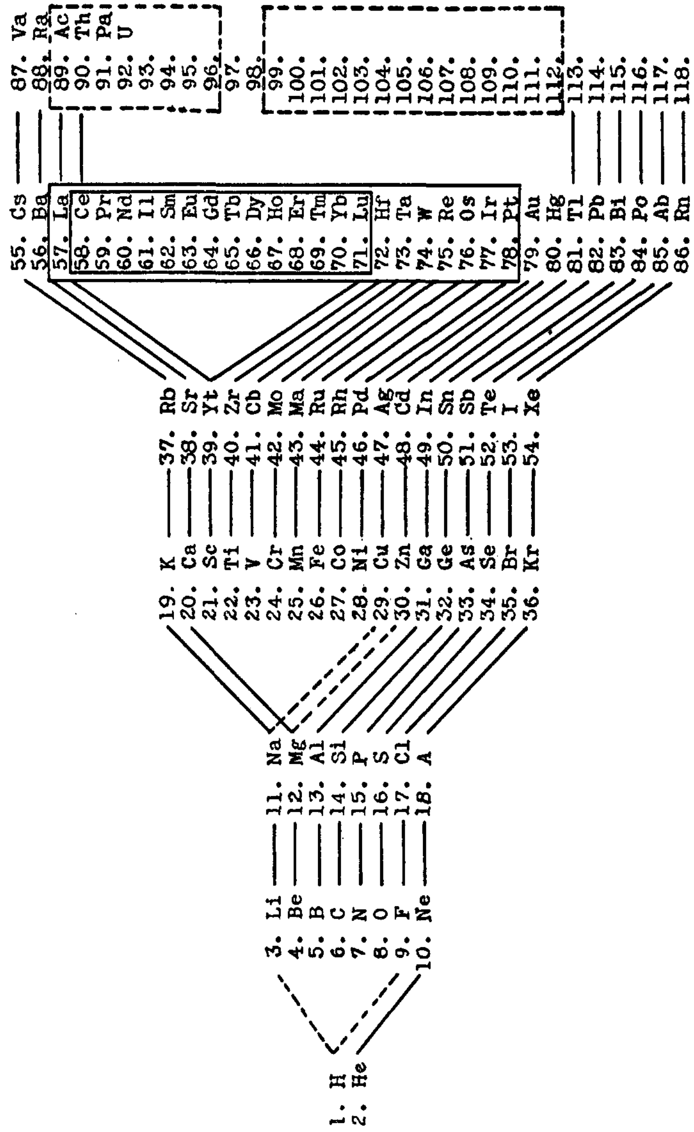
TABLE 6

A long-type periodic table with an "inner transition" group within a normal transition group (see table 8)

																H 1	He 2
Li 3	Be 4											B 5	C 6	N 7	O 8	F 9	Ne 10
Na 11	Mg 12											Al 13	Si 14	P 15	S 16	Cl 17	A 18
K 19	Ca 20	Sc 21	Ti 22	V 23	Cr 24	Mn 25	Fe 26	Co 27	Ni 28	Cu 29	Zn 30	Ga 31	Ge 32	As 33	Se 34	Br 35	Kr 36
Rb 37	Sr 38	Y 39	Zr 40	Cb 41	Mo 42	Ma 43	Ru 44	Rh 45	Pd 46	Ag 47	Cd 48	In 49	Sn 50	Sb 51	Te 52	I 53	Xe 54
Cs 55	Ba 56	*	Hf 72	Ta 73	W 74	Re 75	Os 76	Ir 77	Pt 78	Au 79	Hg 80	Tl 81	Pb 82	Bi 83	Po 84	Ab 85	Rn 86
V 87	Ra 88	Ac 89	Th 90	Pa 91	U 92	E-Re 93	☒	109	110	111	112	113	114	115	116	117	118
*Rare Earths:			La 57	Ce 58	Pr 59	Nd 60	Il 61	Sm 62	Eu 63	Gd 64	Tb 65	Dy 66	Ho 67	Er 68	Tm 69	Yb 70	Lu 71
☒ Possible Transition Group			E-Os 94	95	96	97	98	99	100	101	102	103	104	105	106	107	108

TABLE 7

A Bohr-Thomsen table corresponding to the electronic configurations of table 4

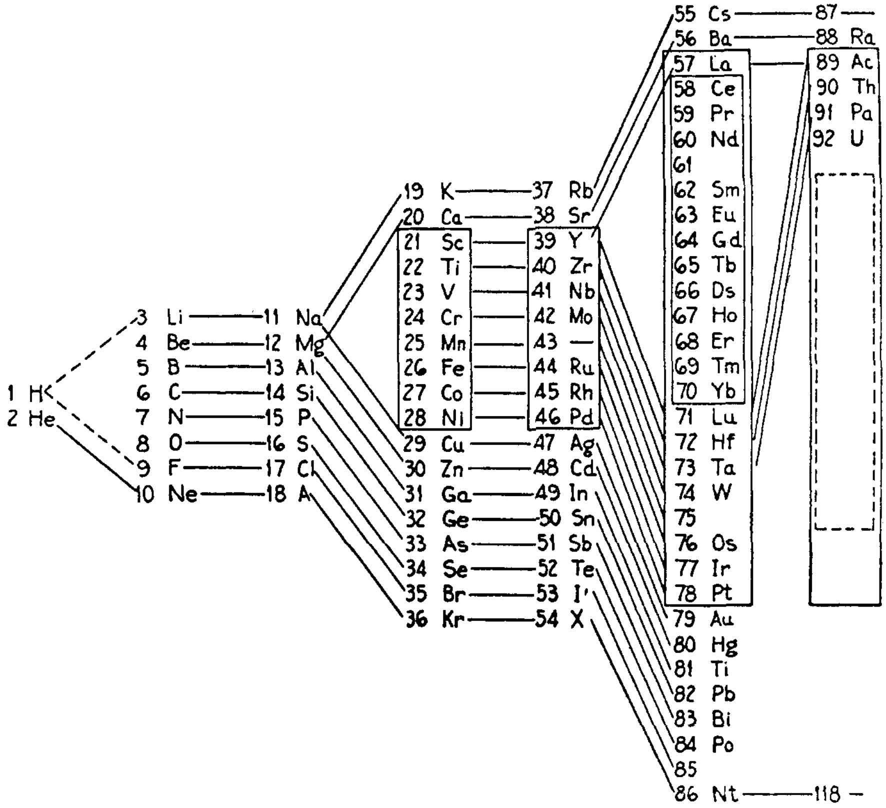


the atomic series. The only possibilities to be considered here will be the following: (*1*) Elements 87 to 118 as a period, with elements 94 to 108 as an "inner transition" group; and (*2*) elements 87 to 118 as a period, with elements 98 to 112 as an "inner transition" group.

Recently von Grosse (76) has written regarding the expected chemical properties of elements 93 and 94. He considered two possibilities,—one with elements 93 and 94 as homologs of rhenium and osmium, the other with a new "inner transition" group beginning with uranium as the first member, just as cerium is the first member of the rare earth group in the previous period.

TABLE 8

A Bohr–Thomsen table with an "inner transition" group within a normal transition group



With respect to the first possibility, von Grosse states, "Ekarhenium (En) will have a valency from a maximum of 7 down to 2 and even 1. The highest oxide, En_2O_7, will be stable, like Re_2O_7, and unlike Mn_2O_7; it will be volatile and probably sublime or melt below 375°. Hydrogen will reduce it at elevated temperatures first to lower, non-volatile oxides and later probably to the metal itself. It will form in water solution the acid,

$HEnO_4$, with water-soluble salts like $KEnO_4$, $Ba(EnO_4)_2$, etc. Since the basicity increases with the period number, $HEnO_4$ must also be basic enough to form water-soluble salts with strong acids (HNO_3, HCl, etc.), analogous to uranyl salts. In any case it will remain in solution in highly acid media. Ekarhenium hexa- or hepta-fluoride (EnF_7) will be very volatile and will be immediately hydrolyzed by water into HF and hydrated oxides.

"Ekaosmium will also have different valences, 8 being the highest. The highest oxide—the tetroxide, $EOsO_4$—will sublime and boil below 200°, dissolve in water and alkalies similarly to OsO_4, but will be a weaker oxidizing agent than OsO_4.

"The highest halides, especially the fluorides, will be volatile and readily hydrolyzable."

The above properties were derived on the basis of an electronic configuration as given in table 9. With respect to the second possibility, which is in accordance with the Bohr theory that "a second group of rare elements" exists, von Grosse gives the electronic configurations shown in table 10, and states that "we may assume that the filling of this lower quantum level has begun already in uranium; in this case the electron distribution would be given by Table III (i.e., table 10) according to the most reliable present knowledge.

"Uranium would then correspond to cerium in the first group of rare earths and elements 93 and 94, also 95 and 96, etc., would all have properties very similar to element 91, just as the chemical properties of Pr, Nd, Sm, etc., are practically identical with La, and elements 93, and 94 in that case, would *not have* the properties of ekarhenium and ekaosmium.

"It might also be that the filling up of the n_5 quantum level will begin at element 93, or 94, or even later; in the first case element 93 will be similar to uranium, just as Ce resembles La, and only in special cases resemble rhenium (like Ce → Zr). A definite decision as to the real nature of these elements could be made after calculating the energy levels of the transuranium elements."

Naturally, as von Grosse also states, if the energy levels of the last three elements in the periodic table (thorium, protoactinium and uranium) were known exactly, it would be possible to predict with more certainty what the behavior of the transuranium elements should be.

1. Elements 87 to 118 as a period, with elements 94 to 108 as the transition group

(*a*) The chemistry of elements 87 to 92 does not need to be considered.

(*b*) *Element 93, ekarhenium.* (*1*) Ekarhenium should exhibit valences from 7 down to 1 or 2. (*2*) The highest oxide of element 93 should be more stable than Re_2O_7; it should be definitely unlike Mn_2O_7 it; should be

TABLE 9*

Electronic configurations of elements 91, 92, 93, and 94

(The electron distribution of the elements given is based on the analogy with their lower homologs)

QUANTUM NUMBERS n……	1	2		3			4				5				6				7	HIGHEST VALENCY
l……	*s*	*s*	*p*	*s*	*p*	*d*	*s*	*p*	*d*	*f*	*s*	*p*	*d*	*f*	*s*	*p*	*d*	*f*	*s*	
ELEMENTS																				
91 Et	2	2	6	2	6	10	2	6	10	14	2	6	10		2	6	3		2	5
92 U	2	2	6	2	6	10	2	6	10	14	2	6	10		2	6	4(5)		2(1)	6
93 En	2	2	6	2	6	10	2	6	10	14	2	6	10		2	6	5(6)		2(1)	7
94 Eo	2	2	6	2	6	10	2	6	10	14	2	6	10		2	6	6(7)		2(1)	8

* Formulated by von Grosse (76).

TABLE 10*

Electronic configurations according to the Bohr theory

QUANTUM NUMBERS n……	1	2		3			4				5				6				7	HIGHEST VALENCY
l……	*s*	*s*	*p*	*s*	*p*	*d*	*s*	*p*	*d*	*f*	*s*	*p*	*d*	*f*	*s*	*p*	*d*	*f*	*s*	
ELEMENTS																				
91 Et	2	2	6	2	6	10	2	6	10		2	6	10		2	6	3		2	5
92 U	2	2	6	2	6	10	2	6	10		2	6	10	1	2	6	4		1	6
93	2	2	6	2	6	10	2	6	10		2	6	10	2(1)	2	6	4(5)		1	5(6)
94	2	2	6	2	6	10	2	6	10		2	6	10	3	2	6	4		1	5
95	2	2	6	2	6	10	2	6	10		2	6	10	4	2	6	4		1	5
etc.																				

* Formulated by von Grosse (76).

definitely volatile, subliming or melting between 300–400°C. This oxide should be easily reduced to lower oxides and then to the metal. (*3*) Ekarhenium would probably be placed along with arsenic or bismuth in the electromotive force series table. (*4*) The chlorides of higher valence should be more stable than those of rhenium, and very definitely more stable than the higher chlorides of manganese. They should be volatile at moderate temperatures. (*5*) The sulfide should form in acid solution in an analogous fashion to ReS_2. (*6*) The metal should dissolve in aqua regia, nitric acid, or other strong oxidizing media to give the acid, $HEnO_4$. (*7*) The element probably would be radioactive.

(*c*) *Element 94.* This element should be somewhat similar in behavior to osmium and should be designated as ekaosmium. It would be the first member of a new inner transition group. (*1*) Like osmium, element 94 might exhibit a valence of 8 as a maximum. (*2*) Its oxide, $94O_4$ should be very volatile, and could probably be steam-distilled from an acid medium. The oxide should react with water to form an acid, and with alkalies. It should be a weaker oxidizing agent than OsO_4. (*3*) The highest halides should be volatile and hydrolyzable. (*4*) The metal should be obtained by reduction more easily than osmium. (*5*) The element should be placed below osmium in the electromotive force series table. Hence platinum and gold should displace it easily from solution. (*6*) The element should form a sulfide in acid solution in similar fashion to rhenium, ekarhenium, osmium, and platinum. (*7*) The metal should have a relatively high melting point, probably about 2000°C. (*8*) If existent in nature, the element should probably be in the free form or as an alloy with other precious metals.

(*d*) *Elements 95 to 108.* (*1*) These elements should all be very similar in properties to ekaosmium, hence their properties would be analogous to those mentioned in the preceding section. The chief differences would be of degree. All of them would probably exhibit a maximum valency of 8. Each of the elements should form higher oxides which would be reasonably volatile, and would differ sufficiently so that separations could be effected. The separations would be difficult. (*2*) The elements should stand below platinum in the electromotive force series. (*3*) They should form compounds like the chloroösmiates, with the probability that perhaps eight groups might be coördinated. (*4*) Undoubtedly, there would be a shrinkage in atomic and ionic sizes of these elements analogous to the "lanthanide contraction" in the rare earth group. This would mean that the properties of the members of this group and of the elements immediately following would vary in a fashion somewhat similar to the changes noted for the rare earths and the elements following (hafnium, tantalum, etc.).

(*e*) *Elements 109 to 110.* These would be homologs of iridium and platinum. Should a contraction of atomic and ionic sizes take place, analogous to the "lanthanide contraction," these elements should be very similar to iridium and platinum in chemical properties, but very different in physical properties such as density, melting point, etc.

(*f*) *Elements 111 to 118.* These would be analogs of the elements gold to radon, respectively. Their properties should be like those of the elements of the previous period (gold, mercury, . . . radon), because of the volume contraction mentioned above. (Zirconium and hafnium, columbium and tantalum, etc., are closely related for the same reason.)

2. Elements 87 to 118 as a period, with elements 89 to 97, inclusive, as a normal transition group, and elements 98 to 112 as an "inner transition" group

This arrangement corresponds to the electronic configurations given in table 4 and in the periodic tables (tables 5 and 7). This particular grouping differs from the preceding in that the "inner transition" group does not build up until after the normal transition group has been completed. Hence, there is a marked difference from the rare earth group, which is an "inner transition" within a normal transition group.

Summarizing this arrangement, it is seen that (*a*) elements 87 to 92 are the elements ekacesium to uranium. (*b*) Element 93 would be a homolog of rhenium, or ekarhenium; element 94 would be a homolog of osmium, or ekaosmium; element 95 would be a homolog of iridium, or ekairidium; element 96 would be a homolog of platinum, or ekairidium. The normal transition group would end at this point. (The transition would begin with No. 89, actinium.) Element 97 would be a homolog of gold, or ekagold. (*c*) Elements 98 to 112 would be an inner transition group, all of the elements having very similar properties. (*d*) Elements 113 to 118 would be analogs of the elements from thallium to radon, respectively.

(*a*) *Elements 87 to 92,* inclusive, are the elements ekacesium, radium, actinium, thorium, protoactinium, and uranium. It will not be necessary to comment on their properties.

(*b*) *Element 93.* The properties of this element would be as listed above (see section 1(b)).

Element 94. The properties of this element would be as listed above (see section 1(c)). The essential *difference* from the preceding case (page 108) is that there would not be a group of fifteen elements possessing very similar properties analogous to ekaosmium.

(*c*) *Elements 95 to 97.* These elements are analogs of iridium, platinum, and gold. They would differ from iridium, platinum, and gold, just as these differ from ruthenium, rhodium, and palladium.

(*d*) *Elements 98 to 112.* These elements form an inner transition group. Ekamercury would be the first member. It would be followed by fourteen elements of very similar properties. All of these elements should be very noble in character, being liquids or low-melting solids. The second member would probably show valences of 2 and 3, and the others would have a normal valence of 2. Regularities within the group would show other valence relationships. A contraction of ionic and atomic volumes would undoubtedly occur with increasing atomic number.

(*e*) *Elements 113 to 118.* These elements are analogs of thallium to radon, respectively. Their properties would be very similar to those of thallium, etc.

(*f*) All of these elements might exhibit radioactivity.

V. ARTIFICIAL RADIOACTIVITY STUDIES ON URANIUM

Previous to 1934 experiments on artificial disintegration had always been considered negative in *two important respects. First,* the emission of high energy electrons as disintegration products had not been demonstrated. *Second,* evidence for the persistence of transformation processes after the removal of the source of bombarding particles had been noted. In 1933 and 1934 I. Curie and Joliot (17) reported that in an artificial disintegration the product atom need not always be a stable isotope, but that disintegration could occur with a relatively long mean-life and with the emission of light particles. As bombarding particles, they used the alpha particles from polonium. They found that the ejected particle usually was a positron. Similar results were obtained by other investigators when various elements were bombarded either with alpha particles or with artificially accelerated protons and deuterons.

Fermi reasoned, however, that neutrons should be very effective as bombarding particles, since the effect of Coulomb repulsion, an important factor in the use of charged particles for bombardment, should not be a deterring factor. Accordingly, he, with his associates, Amaldi, D'Agostino, Rasetti, and Segrè, proceeded to bombard systematically the various chemical elements with neutrons. In 1934 Fermi (27, 28, 29) made the first announcement that artificial radioactivity can be produced by neutron bombardment. Essentially his method consisted in using sealed glass tubes about 6 mm. in diameter and 15 mm. in length and containing radon and beryllium powder as a source of neutrons. The neutrons, passing through the glass walls, acted upon either the elements or the compounds containing them, placed about the radon–beryllium mixture.

Since the nuclei of the heavy radioactive elements exhibit a general instability, Fermi (8) deemed it advisable, as part of his investigation, to determine the effect of neutrons on thorium and uranium. It was found that the two elements could be very strongly activated, and the decay curves indicated that the nature of the induced radioactivity was rather complex. For thorium the initial experiments showed the presence of at least two active substances, one of a period less than 1 min., and the other of a period of about 15 min. With uranium the results were rather startling. The decay curves indicated that the several active constituents were present. The appearance of several half-life periods suggested either that the induced activity was due to different isotopic constituents of the element, or that, if only a single isotope was present, either alternative or successive disintegrations were occurring. Likewise the presence of these substances of different periods indicated the necessity of considering different nuclear reactions. The method utilized to clarify the various possibilities is dependent upon the chemical identification of the active elements.

A. Problems involved in the studies of artificial disintegration

A few preliminary remarks relative to the research methods and problems involved in a radioactivity study of this nature should be made. Three main problems must be kept in mind by the investigator.

First, there is a question of measurement. The detection and identification of the various active substances depends upon the determination of the magnitude and the type of radioactivity. This phase of the investigation also involves the chemistry necessary to purify the starting materials and the separations essential to prove that new atomic species have been produced.

Second, the effect of naturally radioactive substances which might be present in the materials under examination must be remembered.

Third, with uranium, the possibility of transuranium elements being formed looms as an important factor. Should such be the case, numerous questions come to mind, most of which can be summarized in one question, "What would be the chemical behavior of these elements?" The chemical properties which might be associated with these elements have been mentioned previously.

With respect to the first of these problems, it must be remembered that in all cases of artificial radioactivity, minute unweighable amounts of substances are being utilized. They can be detected only by the ionization which their radiations produce. To be sure, this must not be considered as an uncertainty in the proof of their existence. For example, there is no longer any question of the existence of the naturally active elements even though, in nearly all cases, their presence likewise is demonstrated only through the effect of their radiations.

Up to the present time artificially radioactive elements have been obtained in even smaller amounts than some of the naturally active elements, hence it would seem that circumstances will be extremely unfavorable for their separation and identification. However, to discuss a nuclear reaction satisfactorily, it is essential that the active elements involved be separated chemically. Such an analysis consists of adding to the irradiated material small amounts of elements as carriers which are either isotopic or very closely related chemically to the material being examined, and effecting a separation of the various constituents. The activity of the various fractions obtained can then be determined, and the active substance identified. The principle involved is that of the radioactive indicator method, which has been widely used in the past to observe the course of different chemical and physical processes. In this instance, however, the carriers are added to assist in separating the active constituents, instead of adding a radioactive material as an indicator for more common elements.

In the case of the artificially produced radioactive elements, it is also possible to add to the irradiated material (carefully freed of ordinary active materials previous to irradiation) an amount of an ordinary beta-active element. Then if the induced activity and the added activity (recognizable by its characteristic period) can be resolved by chemical means, it can be assumed that the two activities are not due to isotopes.

Direct applications of this principle will be illustrated during the discussion of the chemical separations involved in the study on uranium.

The methods of identification make use of the usual devices for observing radioactivity. Naturally they must be very sensitive, since the amounts of the materials and, frequently, the intensity of the radiations are very low. Geiger-Müller counters which register the number of radiation particles penetrating the tube per minute have been extensively used. For making direct conclusions concerning the nature and the speed of the emitted particles, the Wilson cloud chamber is extremely practical. To absorb certain of the radiations, screens have been used. The sensitive Hoffman type and Wulff type electrometers are extensively employed in different laboratories where this problem is being investigated.

Regardless of whether the material decomposing is a naturally occurring or an artificially produced element, the radioactive disintegration for a homogeneous substance always follows a simple exponential law. That is, in a given unit of time the same percentage of the atoms present will always decompose. Therefore, when the intensity of the activity is plotted logarithmically against the time, the "decay curve" obtained will be a straight line for a homogeneous substance.

For many elements the decay curves are simple exponentials. For others, however, they must be analyzed into two or more exponentials. Such is the case for uranium which has been activated. For two substances, each of which is decomposing at its own specific rate, the "decay curve" is the resultant of the two straight-line exponential curves for each of the two substances. For example, in the case of a mixture of two substances one of which has a very short half-life and is disintegrating to form the second, which has a very long half-life, the decay curve initially shows a very rapid decrease of activity with respect to the time, but eventually changes into a straight-line exponential curve which is characteristic for the longer-lived substance. By subtracting the value for the longer-lived substance it is possible to determine the value for the material with the short half-life, the resulting curve also being a straight line. Conversely, if a substance of a long half-life disintegrates into a very short-lived element, after attaining "radioactive equilibrium," only a straight line is obtained. In this case the presence of the short-lived substance can be established only through chemical separations followed by activity measurements.

In case there are more than two active substances present, the situation is even more complicated. The various processes of disintegration taking place must then be determined by pursuing measurements for longer periods of time, by making chemical separations, and, in the case of induced radioactivity, by varying the period of bombardment in order to change the relative quantities of the various materials which are being rendered active. In the case of uranium these last points are very important.

Referring back to the second problem previously mentioned, the effect of naturally active elements must be anticipated when observing the activated uranium. The naturally active substances might possibly interfere with the chemical separations necessary to prove the presence of elements possessing the induced activity. Their presence might also interfere with the determination of half-life periods and the estimations of the gamma radiations of the artificially produced atoms. On the other hand, the presence of naturally active substances might provide a radioactive indicator which would be useful in following the chemical separations.

A résumé of the three known radioactive series is given in table 11 and figure 1. Interesting facts are the following:

(*1*) UI and UII are isotopes. Both have exceptionally long half-life periods and are alpha-emitting. They will, therefore, not interfere with the detection of substances of relatively short half-life periods involving a matter of minutes, days, or weeks. Should an isotope of one of these forms be produced no difficulty will arise. When radioactive equilibrium for the above has been established, the quantity of UII is found to be very small.

(*2*) Dempster (22, 23, 24) has recently reported the amount of a third

TABLE 11

Half-life periods and radiations of the chief disintegration series

URANIUM RADIUM SERIES	T*	RADIATION	ACTINIUM SERIES	T	RADIATION	THORIUM SERIES	T	RADIATION
Uranium	4.4 × 10^9 yr.	α	Actino-uranium	ca. 4 × 10^8 yr.	α			
Uranium X_1	24.5 days	β	Uranium Y	24.6 hr.	β	Thorium	ca. 1.8 × 10^{12} yr.	α
Uranium X_2	1.14 min.	β, γ	Protoactinium	3.2 × 10^4 yr.	α	Mesothorium 1	6.7 yr.	(β)
Uranium Z	6.7 hr.	β						
Uranium II	3 × 10^5 yr.	α	Actinium	13.5 yr.	(β)	Mesothorium 2	6.13 hr.	β, γ
Ionium	8.3 × 10^4 yr.	α	Radioactinium	18.9 days	α	Radiothorium	1.9 yr.	α
Radium	1590 yr.	α	Actinium X	11.2 days	α	Thorium X	3.64 days	α
Radon	3.82 days	α	Actinon	3.92 sec.	α	Thoron	54.5 sec.	α
Radium A	3.05 min.	α	Actinium A	0.002 sec.	α	Thorium A	0.14 sec.	α
Radium B	26.8 min.	β, γ	Actinium B	36 min.	β, γ	Thorium B	10.6 hr.	β, γ
Radium C	19.7 min.	α, β, γ	Actinium C	2.16 min.	α, β	Thorium C	60.5 min.	α, β
Radium C″	1.32 min.	β	Actinium C″	4.76 min.	β, γ	Thorium C″	3.1 min.	β, γ
Radium C′	ca. 10^{-6} sec.	α	Actinium C′	0.005 sec.	α	Thorium C′	10^{-9} sec.	α
Radium D	22 yr.	β	Actinium D (lead)			Thorium D (lead)		
Radium E	5.0 days	β						
Radium F (polonium)	140 days	α						
Radium G (lead)								

* T = half-value period.

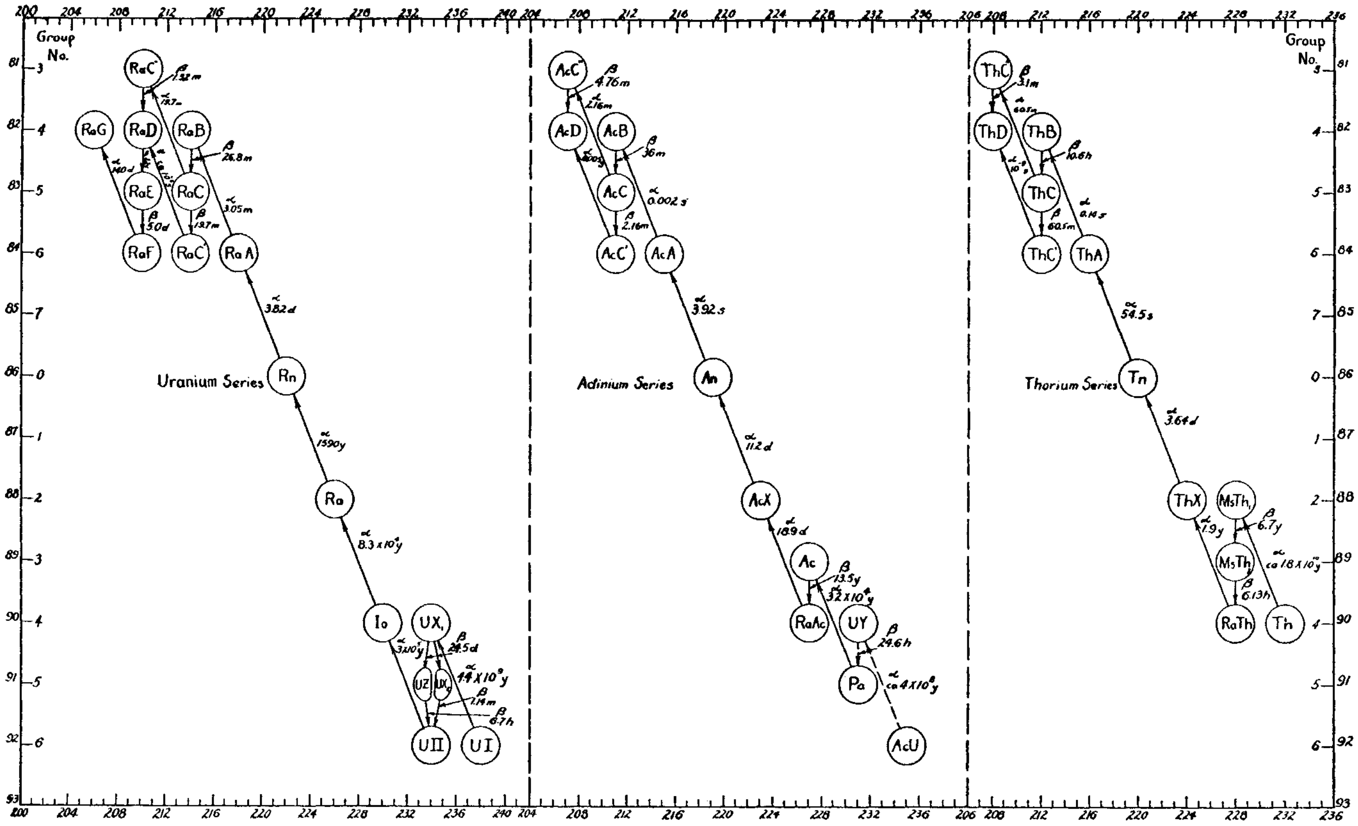


FIG. 1. The three natural radioactive series in an atomic number-atomic weight scheme. In α-emission the element is shifted four places to the left and two upward; in β-emission the element is shifted one place downward.

uranium isotope of atomic mass 235. Hence it is essential that this atomic species be considered in setting up the nuclear reactions taking place. The relative abundance of the three known uranium isotopes (UI^{238}, UII^{234}, U^{235}) is 1:0.007:0.00006.

(*3*) More troublesome are the three beta-emitting isotopes UX_1, UX_2, and UZ. UX_1 is an isotope of thorium ($Z = 90$; half-life = 24.5 days). UX_2 (68.4 sec.) and UZ (6.7 hr.) are both isotopes of protoactinium (No. 91). Unless these elements are removed, it will be found that their activity with respect to that of the artificially produced substances will be very appreciable. Even though they are removed before the start of an experiment, it must be remembered that within a few hours their concentration will again be relatively high compared to the artificially produced elements. Likewise, these elements are being formed constantly even during the bombardment of uranium. Hence if the irradiation is carried on for several hours, as is necessary to prove certain of the changes which are occurring, a considerable excess of UX and a very appreciable amount of UZ with respect to the activated materials may result during the bombardment period.

(*4*) Often, members of both the thorium and actinium series may have been present in the original source of the uranium salts. This implies that a careful purification of the uranium must be made.

With reference to the third question as to whether or not the transuranium elements might be formed, the following questions must be asked: (*a*) Should these elements be formed, can they be easily separated from uranium? (*b*) Should they be easily separated from the uranium, can they be separated without difficulty from one another? (*c*) Should they exhibit very short half-life periods, can chemical separations be performed rapidly enough to be of aid in establishing the exact nature of the disintegration series? (*d*) Should more than one transuranium element be formed, are they produced under identical conditions? In other words, will bombardment with slow and fast neutrons be of assistance in determining the disintegration processes? (*e*) Should elements of very long half-life periods, and in particular alpha-emitting elements, be formed, will it be possible to detect and identify them? (*f*) Would deuteron bombardment supplementary to the neutron bombardment be of assistance in identifying the nuclear reactions? (*g*) Since *K*-electron capture has been observed as taking place in the heavier elements in contrast to the emission of positrons, would such occur with uranium? (*h*) Is it possible that perhaps alpha particles, or other forms of radiations, are emitted only during the time of bombardment? Should such a change take place, the nature of the nuclear reactions noted after cessation of bombardment would probably be drastically different than if no special types of reactions are occurring only during the bombarding period.

B. The experimental results on bombarded uranium

With these questions in mind, a discussion of the work of Fermi, of Hahn, Meitner, and Strassman, of Curie and Savitch, and of others will be reviewed.

The initial report on the induced radioactivity of uranium was made by Fermi (8, 31) in 1934. Half-life periods of 10 sec., 40 sec., 13 min., and at least two other periods varying from 40 min. to 1 day were mentioned. It was stated that electrons had been detected and that Geiger-Müller counters were used to note the radiations.

Fermi (31, 20) and his associates, Amaldi, D'Agostino, Rasetti, and Segrè, cited evidence that the substance with the 13-min. half-life period could easily be separated from most of the heavy elements, and that it was not isotopic with uranium. The concentrated uranium nitrate solution used for the irradiation process was diluted with 50 per cent nitric acid. A small amount of a manganese salt was added and the manganese precipitated as the dioxide from a boiling solution upon addition of sodium chlorate. It was found that the manganese dioxide carried a large percentage of the material possessing the 13-min. half-life period. This indicated that the element with this characteristic was not isotopic with uranium. It is obvious why this particular reaction was chosen for the first determinations. Since the activity was due to a beta-particle emission, according to the Fajans–Soddy rule, an element of higher atomic number than 92 would be indicated. This assumes no alpha-particle emission. A higher congener of manganese and rhenium would thus be expected. The chemistry of manganese might possibly be adaptable, even though there would probably be a considerable difference in the chemical properties of manganese and ekarhenium. A review of the chemistry of rhenium, which should be more closely allied to an element 93, shows that this particular separation is by no means the best, even though for the original exploratory work it served a definite purpose.

Having proved that the 13-min. active substance was not isotopic with uranium, Fermi *et al.* (31) tested the possibility of isotopes of elements 90 (thorium) and 91 (protoactinium). The very short half-life period of UX_2 (68.4 sec.), an isotope of protoactinium, rendered this phase of the work more difficult. The irradiated uranium nitrate solution was mixed with some UX_1 and UX_2, whose activities produced about 2000 impulses per minute on the Geiger counters. Lanthanum and cerium were added to act as carriers for the UX in the chemical separations. The precipitation of manganese dioxide was carried out, using the chlorate oxidation in nitric acid solution. It was found that the precipitated manganese dioxide again carried only the 13-min. substance. No trace of UX_1 (24.5 days) was found in the precipitate. Likewise none of the UX_2 was noted, even though the measurements of the activity were made within 2 min. after the manganese dioxide was precipitated.

The elimination of elements 88 (radium) and 89 (actinium) as the source of the activity was shown by adding $MsTh_1$ as a natural radioactive indicator, with barium and lanthanum as carriers, to the irradiated uranyl nitrate solution. The results were negative.

The Italian investigators likewise added inactive lead and bismuth to the irradiated uranium. The conditions for precipitating the manganese dioxide were then regulated so that the manganese could be precipitated without inclusion of either the lead or the bismuth. In each case the 13-min. active material precipitated with the manganese.

These experiments indicated the possibility that the substance possessing the 13-min. half-life period was not an isotope of uranium (92), protoactinium (91), thorium (90), actinium (89), radium (88), bismuth (83), or lead (82). Its chemical behavior excluded ekacesium (87) and emanation (86) from consideration.

Fermi and his associates (10) reasoned that the lack of positive evidence of a relationship of the 13-min. substance with any of the heavier elements indicated the possibility of an element of atomic number greater than 92. Further they suggested that if element 93 were formed, chemically it should be congener of manganese and rhenium, a hypothesis which they proved by showing that the 13-min. material coprecipitated with rhenium sulfide in hydrochloric acid solution. At first they ruled this evidence as being weak, since several elements can be precipitated with hydrogen sulfide under about these same conditions. The later splendid work of Hahn, Meitner, and Strassmann substantiated their conclusions, however.

Fermi and his associates (31) mentioned that the possibility of similar chemical properties for elements 93, 94, and 95 would render it very difficult to distinguish between these elements. (Reference to an earlier part of this paper will give more detailed information about the close relationship of these elements.)

In a later paper Fermi and his coworkers (34) announced the detection of an active substance of about a 90-min. half-life. They wrote, "A more effective reaction for obtaining the 13-minute active product is the following: irradiated uranium nitrate is dissolved in diluted hydrochloric acid; some rhenium nitrate is added, and then rhenium is precipitated as sulphide by addition of sodium thiosulphate. This precipitate carries about 50% of the activity; and sometimes more. The percentage of UX_1 and of UX_2

found in the rhenium precipitate varies also with the conditions of the reaction (particularly with the acidity), but can be made very low, probably less than 1%. It was actually possible to separate the 13-minute active product and to measure its period using uranium which had not been purified at all from UX. The 90-minute active product has apparently chemical properties very analogous to those of the 13-minute active product as in every type of reaction they are always obtained in about the same percentage. These activities seem, therefore, both to be due to products with atomic number higher than 92, and possibly to isotopes of a same element."

Von Grosse and Agruss (75, 76, 77, 78), on the basis of certain experiments on protoactinium, criticized Fermi's contention that the 13-min. active material was an element 93, and that the 90-min. material was material isotopic with the 13-min. substance. Their thought was that one should expect, according to the periodic law, different properties for the highest homolog of manganese from those assumed by Fermi. Accordingly von Grosse and Agruss (78) performed experiments using protoactinium as a radioactive indicator. In one test they precipitated the manganese from a solution containing uranyl nitrate, nitric acid, protoactinium, and manganese nitrate by the addition of sodium chlorate. The activity in the uranyl nitrate filtrate was precipitated with zirconium phosphate. The activities of both the manganese oxide precipitate and of the material recovered from the uranyl nitrate filtrate were determined. In a second experiment potassium perrhenate in a hydrochloric acid solution was mixed with a definite amount of protoactinium. The rhenium was precipitated as the sulfide, using sodium thiosulfate. After boiling the solution, the rhenium sulfide was filtered off, impregnated with zirconium oxychloride, fumed with sulfuric acid, and ignited, and then its alpha-activity was measured. The protoactinium in the filtrate was recovered with zirconium phosphate as in the first experiment. Table 12 shows the results of this investigation, which did not include any irradiated uranium as one of the materials to be treated. These results seem identical within experimental error. Von Grosse and Agruss suggested that Fermi's 13-min. active material would also show the characteristic reaction of element 91, namely the precipitation, together with zirconium, of the 13-min. substance by phosphoric acid from a strongly acid solution (20 to 30 per cent hydrochloric acid or nitric acid solution). They felt that Fermi's experiments with UX_2 were inconclusive, because the very short life of that radioactive material introduced very great difficulties in the chemical separations. They also hinted that the 13-min. material be named radiobrevium, which is an isotope of protoactinium.

Von Grosse and Agruss (76), however, withdrew their objections to

Fermi's conclusions, after Fermi announced that the zirconium phosphate precipitate produced according to the conditions outlined by von Grosse did not contain any of the 13-min. activity. The work of Hahn, Meitner, and Strassmann also emphasized the correctness of Fermi's conclusion.

I. Noddack (67) also criticized Fermi's original reports, on the basis that Fermi had not investigated whether or not elements other than protoactinium, thorium, actinium, radium, bismuth, and lead as well as emanation and ekacesium might possibly be responsible for the half-life periods and the chemical reactions which Fermi claimed for element 93. Accordingly she performed experiments using manganese nitrate in a nitric acid solution, to which she added small amounts of a large number of different elements, either in the form of soluble salts or as colloids. The resulting mixture was boiled and treated with potassium chlorate. The manganese dioxide precipitate contained titanium, columbium, tantalum, tungsten, iridium, platinum, gold, and silicon in practically quantitative amounts. Antimony, lead, bismuth, nickel, and cobalt were partly precipitated. She also precipitated manganese dioxide from a solution containing some polonium, finding that the polonium precipitated almost quantitatively with the manganese. Fermi did not attempt to prove that polonium was absent. I. Noddack also commented on the well-known fact that rhenium sulfide, like many other sulfides, can adsorb ions and hence might carry down the artificially radioactive substances during the precipitation. Her objections were explained later by Fermi as well as by Hahn, Meitner, and Strassmann.

TABLE 12

Results of an experiment using protoactinium as a radioactive indicator

	REACTION	
	Precipitation of manganese dioxide with sodium chlorate	Precipitation of rhenium sulfide with sodium thiosulfate
Behavior of element 91	99 to 100 per cent precipitated in hot 30 per cent nitric acid	40 to 60 per cent precipitated in hot 15 per cent hydrochloric acid
Behavior of Fermi's element 93	Large per cent of activity precipitated in hot concentrated nitric acid	About 50 per cent of activity precipitated in hot concentrated hydrochloric acid

The work of Hahn and Meitner (39 to 49, 59 to 63) has done much to clarify the position of elements 93 and 94. The initial step in the chemistry which they performed is important in the chemical separations made later in their analyses. Protoactinium, thorium, and uranium were found to be quantitatively precipitated by sodium hydroxide. The salts of the other elements of higher atomic number, as well as those of ekarhenium, should be soluble. Therefore, they added potassium perrhenate and platinum chloride to the irradiated UX-free uranium solution before the addition of sodium hydroxide. The mixture was filtered to remove uranium and any protoactinium which might be present. The filtrate, which had been made slightly acid, was treated with hydrogen sulfide. The precipitated platinum sulfide was removed by filtration. The resulting filtrate was then treated with an equal volume of 37 per cent hydrochloric acid, and the rhenium sulfide precipitated by further addition of hydrogen sulfide. They found that both the 13-min. and the 90-min. active substances were precipitated with the platinum, and that the rhenium sulfide was inactive. If only the rhenium were added, then both active materials went with the rhenium sulfide, although in smaller amounts. They also showed that if UZ (No. 91) were added, no precipitation of this element took place with the sulfide, hence disproving the contention of von Grosse and Agruss (75) that the active materials were isotopes of protoactinium. The fact that the 13-min. and the 90-min. materials precipitated most readily under the conditions for precipitating the platinum sulfide indicated that these substances were probably more like the platinum group metals than like rhenium and probably belonged to elements beyond element 93.

Hahn and Meitner (43) were particularly careful to prove the exclusion of element 91 (protoactinium). To irradiated UX-free sodium uranate dissolved in dilute hydrochloric acid they added about 10 mg. of platinum and rhenium, 5 mg. of zirconium, and some UZ. The amount of UZ (β-emitting, 6.7 hr.) corresponded in its intensity to about an equivalent of the amount of UZ in 150 g. of uranium. A sodium hydroxide precipitation was carried out, the resulting precipitate and filtrate being treated separately. The filtrate, after being made slightly acid, was saturated with hydrogen sulfide. It was noted that the 13-min. and the 90-min. substances concentrated with the platinum sulfide in a yield of about 30 to 34 per cent. The precipitate from the hydroxide containing the uranium was dissolved in 25 per cent hydrochloric acid, and an additional 5 mg. of zirconium added. The UZ was precipitated with the zirconium phosphate. A portion of this precipitate was measured in the electroscope, and the remainder with a counter. The agreement obtained between the treatment with the zirconium phosphate and the platinum sulfide precipitation showed that the 13-min. and the 90-min. substances certainly contained less than 1/1000 of the amount of the UZ originally added as an indicator. This showed in a very direct manner that both of these materials were not isotopic with element 91, as had been suggested by von Grosse.

As Hahn and Meitner (44) continued their investigations of the exact nature of the disintegrations taking place in the bombarded material, it was observed that the half-life value of 90 min. was probably in error and should be regarded as 100 min., even though there was a qualification that perhaps this activity was due to two substances, one of a shorter half-life than 100 min. and the other perhaps of about 2 to 3 days. Subsequently it was determined that the 100-min. activity was due to a mixture of a 59-min. and a 66-hr. substance. Later Hahn, Meitner, and Strassmann (47) reported evidence for the existence of a substance with a half-life of about 12 hr., whose properties indicated that it was a platinum group homolog.

At this same time the German investigators (47) reported a characteristic distinction between the platinum homologs and ekarhenium, based on the behavior of these materials toward metallic platinum or platinum saturated with hydrogen. Experiments were conducted on unbombarded uranium in equilibrium with its decomposition products. It was shown that only a very slight amount of UX_1 (thorium isotope) was adsorbed on platinum immersed in a hot solution of the uranium salt in a normal hydrochloric acid solution. For a short period of time the amount adsorbed was negligible. When similar experiments were tried with the transuranium elements it was noted that the ekarhenium did not separate, whereas the homologs of the platinum group did plate out in considerable amounts. The results with polished platinum foil were the same as with hydrogen-saturated platinum. The amount of adsorption was apparently not affected if weighable amounts of lead, thallium, and bismuth were added to the solution.

In 1935 Fermi, Amaldi, D'Agostino, Pontecorvo, Rasetti, and Segrè (7) presented their results on the slowing down of neutrons by paraffin and other substances rich in hydrogen. Slow neutrons are produced when neutrons are forced to pass through layers of paraffin or water previous to their action on other substances. Generally, with slow neutron bombardment, the neutron is captured by the elements being bombarded, whereas with fast neutrons there may also be the emission of neutrons from the bombarded substance. (These particular reactions will be considered specifically later in this report.)

The Italian investigators observed that of the four periods previously reported (15 sec., 40 sec., 13 min., and 100 min.) the 40-sec. period was not affected by the use of both slow and fast neutrons, whereas there was a very appreciable increase in the intensities of the 15-sec., the 13-min., and the 100-min. activities. The increase of intensity was found to be 1.6. They suggested that the value for the 15-sec. ratio is perhaps not exceptionally accurate. Since the three periods mentioned were all about equally water-sensitive, the conclusion was that they were all probably due to the same primary process, whereas the 40-sec. activity was due to an independent primary process.

The fact that the intensity of the activity due to the three periods was changed about equally for all, suggested that the three activities were perhaps due to a chain process of disintegration or to a complicated branching phenomenon. A certain amount of evidence was obtained by Fermi (7) and his coworkers for the existence of the chain process from the decay curves obtained when a thick layer of irradiated uranium was measured on the electrometer. These exponential curves showed that the initial activities of the 13-min. and the 100-min. periods were about in the ratio of 100:45, the half-value thickness for the beta rays of the 13-min. activity is 0.14 g. per $cm.^2$ of aluminum; for the longer period the ratio was not definitely determined with any accuracy, but it was decidedly less than the former value and probably about one-half. This indicated that the number of disintegrations for the 13-min. and the 100-min. substances are about equal, which is consistent with the assumption that they are connected by a chain process.

At this same time, the Italian investigators (7) reported their experiments relative to the criticisms made by von Grosse and Agruss. Precipitation of the active materials with various sulfides was made. Silver, copper, lead, and mercury salts were used. The acidity of the different solutions was varied. The amount of activity in the precipitated sulfides was generally good. The sulfide reaction appeared successful, even in the presence of a hydrofluoric acid solution of tantalum. They also proved that the activity was not due to a protoactinium isotope, by dissolving specially purified and irradiated uranium oxide in a 25 per cent hydrochloric acid solution to which were finally added solutions of zirconium nitrate and phosphoric acid. The precipitated zirconium phosphate was inactive, whereas the sulfide precipitate obtained by treatment of the filtrate with hydrogen sulfide was active. This reaction proved the nonidentity of the active material with a protoactinium isotope. This conclusion was also reached by Hahn and Meitner, as indicated above.

Fermi (7) likewise reported in this same paper the first evidence which he and his assistants obtained for some separation of the 13-min. and the 100-min. active materials. This was done by dissolving the carefully purified irradiated uranium oxide in hydrochloric acid, and pouring the resulting mixture into ammonium carbonate solution until the uranium precipitate was again completely dissolved. Lead nitrate or manganese nitrate was added to precipitate the carbonates, which were slightly active. The clear filtrate was treated with hydrogen sulfide in the presence of copper salts. The copper sulfide likewise carried a fraction of the activity. The ratio of the two activities, however, was different in the two precipitates, the 13-min. substance apparently being present in larger quantity in the sulfide precipitate, thereby indicating a partial separation. This same evidence suggests that the active elements must be transuranic in nature. At this time, Fermi assumed that the 15-sec., the 13-min., and the 100-min. activities are chain products, due probably to the elements with atomic numbers 92, 93, and 94, respectively.

Hahn, Meitner, and Strassmann (47) in repeating Fermi's experiments on the effect of slow and fast neutron bombardment did not obtain the same results. Accordingly they began a series of experiments to determine the effect of various periods of bombardment, using both slow and fast neutrons. The decay curves obtained showed systematic variations from one another, depending upon whether the time of bombardment was short (5 to 10 min.) or long (30 to 120 min.).

A short period of bombardment with fast neutrons gave in addition to the rapid decay during the first few minutes (due to the 10-sec. and the 40-sec. substances) an activity which corresponded closely to Fermi's 13-min. material. With slow neutrons the period of decay was decidedly longer. This indicated a relatively larger amount of the so-called 100-min. substance present in this instance. The curves for the irradiation with slow neutrons showed definitely that there were more transformation products than when bombardment was made with fast neutrons. After long periods of bombardment, the amounts of the longer lived substances naturally were greater. For these periods of long irradiation, the formation of UX from the uranium caused difficulty.

In their report Hahn, Meitner, and Strassmann (48) pointed out one very important factor relative to the chemistry involved in the separations of the transuranic elements. They were able to show conclusively, after precipitation with hydrogen sulfide from hot, strongly acid, hydrochloric acid solutions, that the naturally occurring radioelements down to 85, bismuth, remained entirely in the filtrate, whereas the transuranium elements precipitated under these conditions.

These investigators also made chemical separations on the carefully purified uranium which had been subjected to both rapid and slow neutrons for varying periods of irradiation. In these experiments it was likewise shown that systematic variations occurred in the decay curves, demonstrating that bombardment with slow neutrons intensified certain of the processes, and not others. Their conclusion at this time, contrary to the work of Fermi and his assistants, was that the 13-min. substance was formed only after bombardment with fast neutrons, whereas the 100-min. substance could be produced with both fast and slow neutrons, the yield of the latter being greater in the case of slow neutron bombardment.

Another observation made at this time was that apparently transformation products other than the transuranic elements were being created, suggesting therefore that elements 90 to 92 were the active constituents. Hence they proceeded to consider the nature of the nuclear reactions involved in the formation of the transuranium elements and the subsequent disintegration series.

This possibility of thorium, protoactinium, and uranium being present caused Hahn and Meitner (62) to assume that the following reactions were occurring:

$$\mathrm{U}^{238} + n \rightarrow \mathrm{Th}^{235} + \alpha$$

$$\mathrm{Th}^{235} \rightarrow \mathrm{Pa}^{235} \xrightarrow{\beta} \mathrm{U}^{235} \xrightarrow{\beta} \text{Eka-Re} \rightarrow ?$$

The above reaction is one which might be expected upon neutron bombardment of uranium, provided the proper energy relationships existed at the time of bombardment.

The evidence cited by Hahn, Meitner, and Strassmann (47) for the existence of the thorium isotope was based on certain chemical separations. Several grams of UX-free uranium were bombarded for 10 to 15 min. with slow neutrons. The uranium was dissolved as rapidly as possible in 2 normal hydrochloric acid; two precipitations as the sulfide were made after the addition of 10 mg. of platinum, in order to eliminate the transuranium elements. After addition of ferric chloride to the filtrate, the uranium was precipitated with ammonia and dissolved in an excess of ammonium carbonate. The iron hydroxide contained the UX, the thorium isotope, and the protoactinium isotope. Fifteen minutes after cessation of bombardment, the measurement of the activity was begun. The decay curves showed a rapid decrease in intensity of radiation at first, but gradually changed to the characteristic curve for UX. The half-life of the substance causing the initial change in the curve was found to be 4 min. after subtraction of the value for the UX. The authors suggested that the methods of chemical separation used did not permit distinguishing between Th^{235} and Pa^{235}. They believed that this new activity must be associated with Th^{235} on the basis of the radioactive disintegration laws.

This value of 4 min. for a thorium-235 did not agree with other values for the disintegration of either thorium or protoactinium. There is an active thorium isotope (233) of a half-life of 25 min. and a protoactinium of 233 with a short half-life of about 2.5 min., as reported by I. Curie *et al.*

In a later report Hahn, Meitner, and Strassmann (48) clarify the question about this thorium isotope, which they unfortunately postulated. They noted, in some of their experiments, that the substance with the 23-min. activity was scattered through precipitates other than those containing the uranium. For example, in the zirconium phosphate precipitation used for proving the presence of thorium isotopes, some of this active substance was found to be present. Subsequently these observations were explained as follows: Apparently during the bombardment of the uranium material a sort of Szilard–Chalmers (73) separation occurred, in that a portion of the uranium isotope was present in other than the hexavalent condition. By heating the bombarded uranium with nitric acid, the tetravalent uranium was reconverted to the hexavalent state. The zirconium phosphate precipitate, under these conditions, did not contain any of the short-lived material. Thus it was possible for the German investigators to eliminate the above nuclear reactions from consideration as one series in the disintegration of the transuranium elements.

C. The active transformation processes

Hahn, Meitner, and Strassman (48, 60, 62, 63) have suggested the following three series of reactions to show the genetic relationship of the different substances obtained when uranium is bombarded with neutrons. No suggestion of atomic mass values was considered.

$$\mathrm{U} + n \rightarrow {}^{10\,\mathrm{sec.}}_{92}\mathrm{U} \xrightarrow{\beta} {}^{2.2\,\mathrm{min.}}_{93}\text{Eka-Re} \xrightarrow{\beta} {}^{59\,\mathrm{min.}}_{94}\text{Eka-Os} \xrightarrow{\beta} {}^{66\,\mathrm{hr.}}_{95}\text{Eka-Ir} \xrightarrow{\beta} {}^{2.5\,\mathrm{hr.}}_{96}\text{Eka-Pt} \xrightarrow{\beta} {}_{97}\text{Eka-Au} \rightarrow \,? \qquad \text{(I)}$$

$$\mathrm{U} + n \rightarrow {}^{40\,\mathrm{sec.}}_{92}\mathrm{U} \xrightarrow{\beta} {}^{16\,\mathrm{min.}}_{93}\text{Eka-Re} \xrightarrow{\beta} {}^{5.7\,\mathrm{hr.}}_{94}\text{Eka-Os} \xrightarrow{\beta} {}_{95}\text{Eka-Ir} \rightarrow \,? \qquad \text{(II)}$$

$$\mathrm{U} + n \rightarrow {}^{23\,\mathrm{min.}}_{92}\mathrm{U} \xrightarrow{\beta} {}_{93}\text{Eka-Re} \rightarrow \,? \qquad \text{(III)}$$

1. The chemical identification of uranium isotopes

From the above three transformation series it is seen that active uranium isotopes were formed by irradiation with neutrons.

The evidence for these isotopes may be shown by following one of these materials (the 23-min. substance) through the chemical separations. (The exact conditions for exciting uranium to form this active isotope will be mentioned later.) A UX-free preparation, bombarded with slow neutrons under the proper conditions and then dissolved in a 2 *N* hydrochloric acid solution, was saturated with hydrogen sulfide. The filtrate, containing the uranium, was then treated with a large excess of sodium acetate and the sodium uranyl acetate precipitated with concentrated acetic acid. The activity of the uranyl acetate precipitate was found to decrease during the first 1.5 to 2 hr., and then to increase on account of the gradual formation of UX from the uranium. The decrease showed the presence of a homogeneous substance with a half-life period of 23 min. That the activity of the uranyl acetate might be due to the possible adsorption of the 23-min. substance was guarded against by making fractional precipitations of the acetate on a large amount of irradiated uranium. Three precipitations were made, stepwise, from the uranium solution. Equal quantities of the first and third fractions were tested for their content of the 23-min. substance. The amount of activity was exactly the same for every unit weight of uranium. The possibility of adsorption was eliminated by this procedure, which also showed that this material was chemically identical with uranium. Such a treatment naturally implies that the bombarded uranium had been treated with nitric acid to eliminate any possibility of a Szilard–Chalmers (73) separation.

Since the 23-min. material emits beta particles, it must form an ekarhenium. No definite evidence has been obtained, up to the present, for this particular ekarhenium. Should it be a stable ekarhenium its existence would be very difficult to prove. Also, on the other hand, should this isotope of ekarhenium possess a very long half-life, its presence would not be easily determined.

The existence of the 40-sec. uranium isotope may be proven in similar fashion, according to the German investigators. In this instance fast neutrons are used to produce the active element. The 23-min. material is not formed under these conditions. The purified uranium was bombarded for a very short period, and the uranyl acetate precipitate produced as rapidly as possible. The investigators reported that they were able to begin the radioactive measurements in about 2.4 min. after cessation of bombardment. The initial decline of the decay curves was much more rapid than in the case of the 23-min. material.

Thus far, no chemical test has been successfully made of the 10-sec. substance formed during a 15-sec. bombardment of uranium.

2. The distinction of ekarhenium from homologs of the platinum metals

One difference between ekarhenium and the homologs of the platinum metals is based on the behavior of these elements with respect to the noble metals. The ekarhenium isotopes will not precipitate on platinum foil during electrochemical treatment, whereas the platinum homologs will. This distinction is very characteristic. A typical experiment, as carried out by Hahn, Meitner, and Strassmann (48), begins by bombarding some purified uranium for about 36 min. After this length of irradiation approximately three-fourths of the equilibrium amount of ekarhenium (16 min.) was formed in contrast to about one-fourth that of ekaosmium (59 min.). The uranate was dissolved in 2 *N* hydrochloric acid, which was then diluted to 1 normal acid concentration. A piece of platinum foil, previously washed with hydrogen, was then immersed in the nearly boiling solution for about 10 min. After cleaning the foil with acid and water, the activity of the sheet was determined and found to be due only to precipitated ekaosmium. Control experiments were made, in which both the ekarhenium and ekaosmium were precipitated as the sulfide, the decay curves for these samples being much steeper at the outset, indicating that the percentage of ekarhenium was much greater than that of the ekaosmium.

The electrochemical separation of ekarhenium and the artificially produced noble metals is even more convincing when it is remembered that, on account of the very short life of the ekarhenium, it is necessary for the ekarhenium activity to be at least four times as intense in order to equal that of the ekaosmium.

These particular experiments are likewise excellent evidence of the very noble character of the homologs of platinum metals.

3. The precipitation of the homologs of the platinum metals and the non-precipitation of ekarhenium with metallic bismuth

In another series of experiments the German investigators found that it was possible to precipitate the artificially produced noble metals along with metallic bismuth from a bismuth salt solution, using alkaline stannous chloride as the reducing agent.

UX-free uranium was irradiated in the form of ammonium pyrouranate with neutrons for about 60 min. From a warm solution of the uranate in approximately 5 per cent hydrochloric acid, about 30 mg. of bismuth was precipitated with hydrogen sulfide. The bismuth sulfide which contained the precipitated transuranic elements—mainly ekarhenium (16 min.) and ekaosmium (59 min.)—was dissolved in 25 per cent hydrochloric acid with the addition of a few drops of nitric acid. The nitric acid was used to guarantee complete solution of the platinum homologs. The clear filtrate, after dilution with water, was almost neutralized with sodium hydroxide, mixed with alkaline stannous chloride, and warmed. Metallic bismuth precipitated carrying with it the platinum homologs, of which essentially only ekaosmium was present. The filtrate from the bismuth precipitation was poured into 25 per cent hydrochloric acid solution which contained some rhenium, and an equal volume of 35 per cent hydrochloric acid was added. The rhenium was then precipitated as the sulfide. The tin remained in solution, the ekarhenium going with the rhenium sulfide precipitate.

The decay curves (figure 2) show very clearly this particular separation. Curve A shows the decrease of activity of the metallic bismuth and the included platinum homologs. Curve B is much steeper, at the outset showing only the characteristic decay of ekarhenium (16 min.). Curve A is flatter, and shows the decomposition of the ekaosmium (59 min.). The bending of the curve for the ekarhenium (curve B) is explained as due to the formation of the second ekaosmium isotope (5.7 hr.) from the 16-min. ekarhenium. (This point will be mentioned again later.)

4. The precipitation of ekarhenium with nitron

One of the most typical precipitation reactions of rhenium is the nitron test (38). Rhenium, like nitrates, forms a very insoluble precipitate in a weak acetic acid solution with nitron. It was natural to think that perhaps ekarhenium would do likewise. A uranium preparation was bombarded for a short period in order to give a maximum amount of the 16-min. ekarhenium. This was dissolved in 20 per cent hydrochloric acid, mixed with some rhenium in the heptavalent form, and treated with hydrogen sulfide in the cold. Re_2S_7 separated. This precipitate was then dissolved in a warm solution containing sodium hydroxide and hydrogen

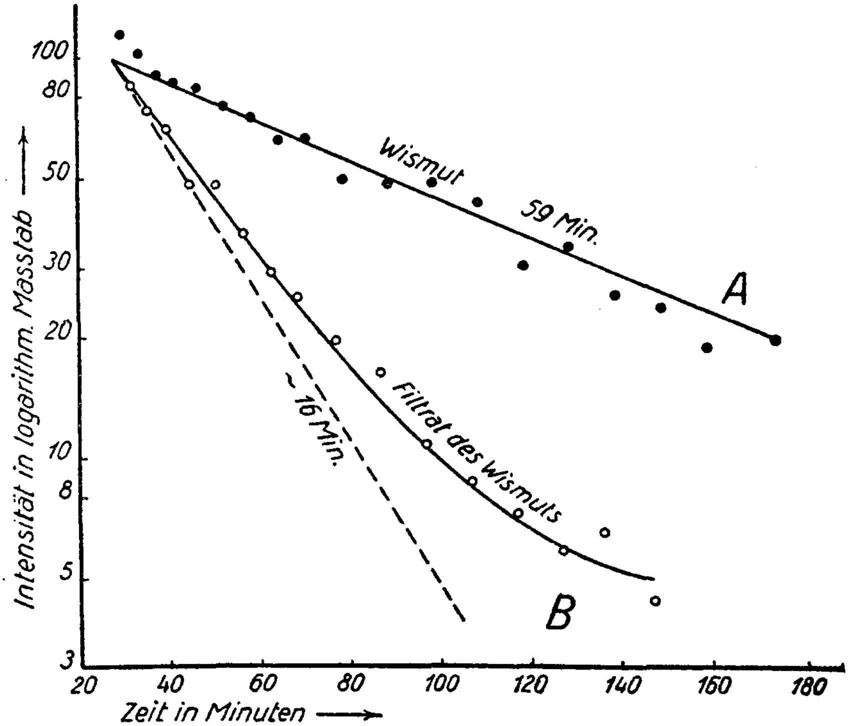


FIG. 2. The separation of ekarhenium from ekaosmium using metallic bismuth as the carrier for the ekaosmium (according to Hahn, Meitner, and Strassmann (48)).

peroxide. The resulting solution was acidified with acetic acid, cooled, and treated with nitron acetate. A voluminous precipitate of nitron perrhenate formed, which was found to contain a large quantity of the substance with the 16-min. half-life, along with a small amount of the 59-min. substance. Dissolving the precipitate in warm hydrochloric acid, almost neutralizing with sodium hydroxide, and adding a further quantity of nitron acetate gave a purer nitron perrhenate containing the 16-min. material in smaller amount but greater purity.

5. The use of sodium hydroxide to separate ekarhenium from the platinum homologs

Hahn, Meitner, and Strassmann (48) also observed the effects of sodium hydroxide upon hot solutions containing weighable amounts of sodium perrhenate, platinum chloride, and osmium ammonium chloride in the irradiated uranium preparations. Rhenium was found to remain quantitatively in solution, platinum was partially adsorbed, and the osmium was completely precipitated. The natural transformation products of uranium were quantitatively precipitated.

Hence an irradiated uranium, bombarded for a short period to give a large amount of the 16-min. isotope of ekarhenium and a small amount of ekaosmium, was treated with a small amount of sodium perrhenate and sodium hydroxide. Part of the ekarhenium went into the alkaline filtrate. The ekaosmium remained completely in the precipitate. Unfortunately part of the ekarhenium was adsorbed by the large quantity of uranium present. The decay curves of the filtrate and the precipitate showed the separation of the ekarhenium and the ekaosmium, as well as the formation of ekaosmium from ekarhenium.

The investigators succeeded in showing, with uranium that had been strongly bombarded and rapidly separated, that some of the 2.2-min. ekarhenium was separated with the 16-min. isotope. It was again noted that the 16-min. material disintegrated into the ekaosmium of 5.7-hr. half-life. Using uranium preparations bombarded for very long periods of time (those in which there was formed a larger concentration of elements 95 and 96), only a fraction of the ekairidium (No. 95) and the ekaplatinum (No. 96) was found in the filtrate.

6. The difference in volatility of the ekarhenium and the ekaosmium isotopes

As mentioned above (page 108), one would expect that ekaosmium should be volatile in the form of certain of its compounds, and that the chloride of ekarhenium should be more volatile than the chloride of ekaosmium.

The experiments of Hahn, Meitner, and Strassmann have shown this to be true.

Irradiated uranium was treated to precipitate the sulfides of ekarhenium and ekaosmium, with platinum as a carrier. Then a series of differentiated ignitions was carried out. For those experiments in which the ekaosmium would be expected to form an oxide, there was a decrease in the activity of the residue remaining after heating. The decay curves for this residue indicated an excess of ekarhenium. In other experiments in which the precipitated sulfides were treated with aqua regia, so that chlorine was present, evaporation to dryness and subsequent ignition showed a change in activity of the residue such that the residue was shown to consist of ekaosmium.

One such experiment consisted in collecting the sublimate and then comparing the activities of the volatile and non-volatile portions. The technique consisted in placing the crucible containing the platinum–osmium precipitate (which had been evaporated down with aqua regia) in a thick asbestos disk so that the bottom of the crucible could be strongly heated without the upper portion of the crucible walls becoming too hot. The crucible was covered with aluminum foil, which was cooled by placing a water-filled aluminum dish on top. The bottom of the crucible was gently heated, and the sublimate collected on the aluminum foil. A second piece of foil was then placed on the crucible, and the crucible was heated very strongly for a considerable period of time. The residue remaining after the ignitions was also measured for its activity. The results obtained were as follows: In the first sublimate the activity showed an excess of ekarhenium in the mixture. In the second sublimate the activity was weak, and was essentially only ekaosmium. (The activity half-life was about 60 min.) In the residue the activity was about ten times that of the second sublimate and was apparently due only to ekaosmium (T (observed) = 60 min.)

In other words, when the substances formed are the oxides or oxygen salts, the ekaosmium compound is the more volatile; if they are in the form of the chlorides, the ekarhenium compound is the more volatile, which is exactly as expected for the properties of these elements.

7. The differences in solubility of the sulfides of ekarhenium and ekaosmium in different concentrations of hydrochloric acid

According to the German investigators most of the transuranic elements are precipitated as the sulfides from a 2 N hydrochloric acid solution. With increasing acid concentration, the solubility of the ekarhenium sulfide increases. For example, from a 10 N hydrochloric acid solution a considerable portion of the ekaosmium precipitates, the ekarhenium being obtained by diluting the filtrate and saturating further with hydrogen sulfide.

8. Distinguishing the platinum homologs from one another

The experimental difficulties arising in the separations of the platinum group homologs are complicated, as one would expect. For example, a mixture might contain the two isotopes of element 94 (59 min. and 5.7 hr.), one of element 95 (66 hr.), and one of element 96 (2.5 hr.). The longer lived ekaosmium (No. 94) and the ekairidium (No. 95) are very weakly active. Hence for one set of separations, three different elements, as well as two isotopes of one of them, must be accounted for.

The differences between ekaosmium and ekairidium are such that certain of the reactions used to distinguish between ekarhenium and ekaosmium are applicable. For example, for these latter two elements (Nos. 93 and 94) several methods give indisputable evidence for the separation of the elements, according to Hahn, Meitner, and Strassmann. These are the precipitation of the osmium with metallic bismuth by the action of alkaline stannous chloride, the greater solubility of ekarhenium sulfide in concentrated hydrochloric solutions, and a partial precipitation of the ekaosmium with sodium hydroxide. These same reactions may be used to distinguish ekaosmium (No. 94) and ekairidium (No. 95). The chief factor in these separations is to continue bombardment for a sufficiently long period in order to build up the concentration of these elements of highest atomic number.

(A). The separation of ekaosmium and ekairidium

(1) A partial separation using metallic bismuth as a carrier. A partial separation of these two elements was found to occur when the bismuth salt solution containing the active material was treated with alkaline stannous chloride. (This treatment has been mentioned on page 128.) However, in this case, the uranium samples were irradiated for at least 10 hr. previous to the chemical separations.

The metallic bismuth precipitate was extremely active. The slope of curve A (figure 3) is steep at first, which is evidence for the short-lived ekaosmium (59 min.). The gradual bending of the curve shows the presence of an appreciable quantity of the 5.7-hr. ekaosmium. Ultimately curve A approaches the value of 66 hr., which is due to the ekairidium forming from the ekaosmium.

The filtrate from the separation of the bismuth was treated with hydrogen sulfide. The resulting sulfide precipitate was also very active, owing to the ekarhenium present. After about 3 to 4 hr. the amount of ekarhenium (No. 93) remaining was very low, and the decay curve (curve B) flattened rapidly, gradually approaching the value of 66 hr. for ekairidium. This curve likewise shows some evidence for the 5.7-hr. ekaosmium. This mixture contained about 2.7 times as much ekairidium as the bismuth precipitate.

The ekaosmium present at the time the metallic bismuth was reduced apparently was quantitatively precipitated, whereas only about one-third of the ekairidium precipitated.

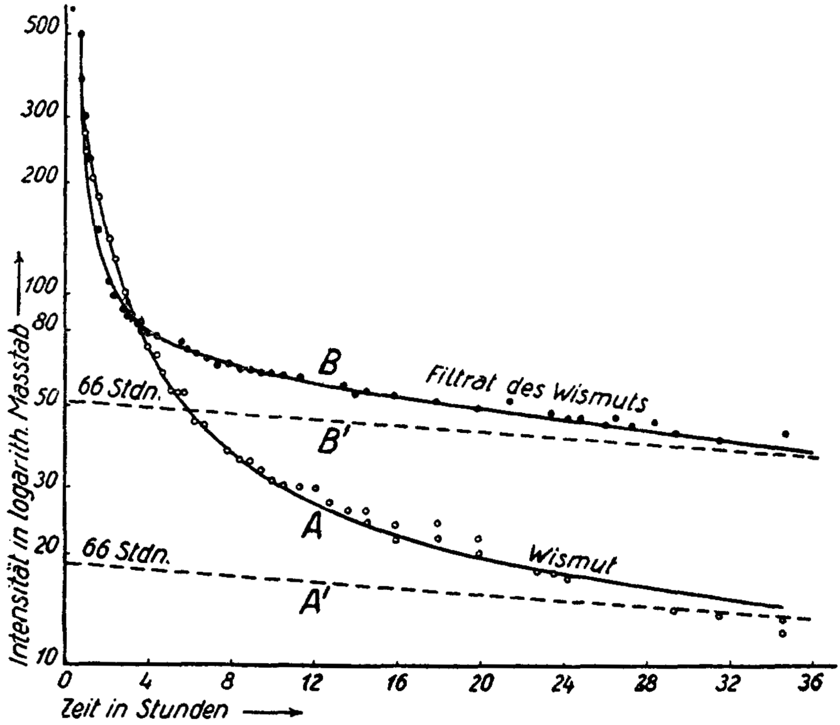


FIG. 3. The partial separation of ekairidium from ekaosmium, using the alkaline stannous chloride-bismuth salt reaction (according to Hahn, Meitner, and Strassmann (48)).

(*2*) *Partial separation of ekairidium and ekaosmium with sodium hydroxide.* Samples of uranium which had been bombarded for long periods of time in order to build up the concentration of ekairidium were treated with sodium hydroxide. It was observed that a considerable portion of the 66-hr. substance passed into the filtrate along with the ekarhenium, and that the precipitate contained an appreciable amount of the 59-min. ekaosmium. This compares well with the experiments on samples bombarded for short periods, which showed likewise that the ekaosmium precipitated. However, if large quantities of uranium were used in these experiments, the German investigators noted that the quantity of the ekairidium adsorbed by the uranium precipitate increased. No quantitative results were obtained, the suggestion being that the presence of the ekaplatinum of 2.5-hr. half-life interfered.

(3) *The solubility of ekairidium sulfide in 10 N hydrochloric acid.* Hahn, Meitner, and Strassmann (48) also studied the effect of hydrogen sulfide on ekairidium in solutions of varying hydrochloric acid concentration. A quantity (about 10 g.) of uranium was bombarded for 60 min. The usual sulfide precipitation from a 2 *N* hydrochloric acid solution, using platinum as a carrier, was made. The platinum sulfide was dissolved and allowed to stand for 2.5 hr. to permit the disintegration of the ekarhenium (16 min.). This solution, which was 10 *N* in hydrochloric acid and to which some perrhenate solution had been added, was again saturated with hydrogen sulfide in the cold. Part of the platinum and all of the rhenium precipitated. The filtrate, after dilution to 2 *N* concentration, was heated to boiling, and the remainder of the platinum precipitated as the sulfide. The decay curves showed that the ekaosmium (59 min.) which remained in the mixtures was almost quantitatively in the precipitated sulfide from the strongly acid solution, whereas none of the 59-min. material seemed to be in the precipitate obtained from the dilute acid solution. The activity curves were very complicated, since apparently four different kinds of atoms were present. The decay curves for each of the precipitates gradually changed over to the characteristic 66-hr. slope, indicating also about equal distribution of the ekairidium in each. The conclusion reached by the investigators was that ekaosmium can precipitate more readily in strongly acid solutions than can ekairidium.

(B). The separation of ekaplatinum (No. 96) from the other platinum group homologs

Hahn, Meitner, and Strassman conducted experiments to prove the existence of ekaplatinum. They bombarded uranium for days or weeks to permit a building-up of the elements of greatest atomic number. The transuranic elements were then separated by means of hydrogen sulfide, using platinum as a carrier. The precipitates were allowed to stand from 1 day to several days. The decay curves of these precipitates indicated that the ekarhenium (16 min.) and the ekaosmium (59 min.) had disintegrated, leaving only the 66-hr. element (No. 95) and its decomposition product, the 2.5-hr. ekaplatinum (No. 96). The 66-hr. substance seemed to be almost inactive. The formation of the ekaplatinum caused a very definite increase in the activity of the precipitate.

The German investigators suggested six different methods for separating ekaplatinum and ekairidium from mixtures which had been standing long enough to permit the complete disintegration of the ekarhenium and ekaosmium.

Method 1. Ekaplatinum was found to precipitate in very slight amount when a 10 *N* hydrochloric acid solution was saturated with hydrogen sulfide. In 2 *N* hydrochloric acid ekaplatinum was also not entirely precipitated. The increase in activity of a rhenium sulfide precipitate prepared from a 10 *N* hydrochloric acid solution suggested that about 75 per cent of the ekaplatinum (No. 96) remained unprecipitated.

The order of precipitation by hydrogen sulfide in a strongly acid solution is: eka-Os > eka-Re > eka-Ir > eka-Pt.

Method 2. Apparently only a small amount of ekaplatinum precipitated with bismuth when alkaline stannous chloride was used as the reducing agent. This separation of ekaplatinum from ekairidium seems to be more efficient than the precipitation with hydrogen sulfide.

At this point it should be mentioned that one would expect, from the periodic table relationships, that all of the platinum group homologs would be easily and completely precipitated along with bismuth using alkaline stannous chloride, even though the results of the German investigators seem to indicate the opposite. This again raises the question as to the exact structures and chemical properties of these elements of high atomic number. Are they members of a transition group or not? Could it also be that, in contrast to the structures proposed above, either ekaosmium or ekairidium is the first member of a new "inner transition" group and that ekaplatinum is exhibiting properties which might be expected of the second or third member of such a group?

Method 3. Ekaplatinum is apparently more volatile than ekairidium, according to Hahn, Meitner, and Strassmann. This information resulted from several different tests, as follows: (*a*) A precipitated mixture of platinum and rhenium sulfides containing an excess of ekairidium was carefully ignited in an open crucible. The activity of the residue in the crucible showed it to be almost pure ekairidium, the decay curve showing the eventual formation of ekaplatinum. (*b*) The transuranium elements were removed from uranium which had been subjected to a long period of bombardment by precipitation with hydrogen sulfide, with platinum as a carrier. The platinum sulfide was dissolved in aqua regia, and reprecipitated as the sulfide in order to eliminate traces of UX formed during the bombardment period. This preparation was then carefully ignited, the resulting mixture then being allowed to stand until the next day. The activity then showed an equilibrium between the 66-hr. ekairidium and the 2.5-hr. ekaplatinum.

A series of sublimations similar to those mentioned for the separation of ekaosmium and ekairidium was made on the above preparation. The first sublimate resulted from a 2.5-min. heating with a Bunsen flame. The second sublimate resulted from a 2.5-min. ignition with a blast lamp. The decay curve of the first sublimate showed a very definite excess of ekaplatinum (2.5 hr.). The activity curve gradually changed to show the formation of ekaplatinum from ekairidium.

A question which should be asked in connection with these fractional sublimation experiments is whether the elements themselves, or their oxides or sulfides, were volatilized? If it were the elements themselves, it indicates that these elements resemble metallic mercury in their tendency to volatilize somewhat readily. Such a tendency would also indicate that the physical properties of these elements are not the same as those predicted previously. Since the sublimations were made in air, it was probably the oxides that evaporated, especially for the low temperature ignitions. It could be possible to sublime these metals slowly in a blast lamp.

Method 4. The adsorption of ekaplatinum on precipitated ammonium pyrouranate. As mentioned previously (page 130), ekaosmium precipitates completely, whereas ekairidium (No. 95) is only partially precipitated upon treatment of the irradiated uranium solutions with sodium hydroxide. Experiments conducted on samples rich in the 66-hr. ekairidium and the 2.5-hr. ekaplatinum (No. 96) showed that the latter was even more soluble than ekairidium (66-hr.) in the basic solutions. The German investigators also noted that the tendency of the platinum homologs to coprecipitate or to be adsorbed with ammonium pyrouranate varied in the following order: eka-Os > eka-Ir > eka-Re > eka-Pt.

As in the precipitation of the sulfides from strong hydrochloric acid solutions, ekaosmium is the most easily separated or least soluble and ekaplatinum the least easily precipitated or adsorbed.

Method 5. The separation of ekaplatinum with platinum ammonium chloride. Seeking to learn if the platinum homologs formed were analogous to platinum ammonium chloride and iridium ammonium chlorides, Hahn *et al.* found that mixed crystals could be formed and that, in addition, a separation could be effected. A mixture of the 66-hr. ekairidium and the 2.5-hr. ekaplatinum was added to a dilute hydrochloric acid–platinum chloride solution, to which ammonium chloride was then added. Activity measurements showed that the 2.5-hr. material precipitated in the crystals formed, even though present in very slight amount. In another study ekairidium also crystallized, with ekaplatinum in definite excess. Thus it was shown that these elements are similar to the platinum group in forming the same typical salts, and that a separation of elements 95 and 96 could be made.

Method 6. The use of platinum foil for separating ekairidium. As mentioned earlier (page 127), ekarhenium could be separated from eka-osmium because the more noble character of the latter permitted its precipitation on a platinum foil. When a piece of platinum was placed in a solution containing a mixture of ekairidium and ekaplatinum, both metals were found in the plated deposit. Ekaplatinum was present in excess. Curve A in figure 4 indicates the activity of the materials plated on the platinum during a 10-min. immersion in a hot solution containing the transuranium elements (in this case, essentially only elements 95 and 96) in the presence of small amounts of lead, titanium, bismuth, and zirconium. Curve B records the activity of the sulfide precipitate obtained by treating the solution, remaining after the platinum foil was removed, with hydrogen sulfide. The trend of the curve shows the building-up of the 2.5-hr. ekaplatinum. (It must be remembered that, for equal numbers of atoms, the shorter lived ekaplatinum emits a greater number of particles per unit time than the more stable ekairidium.)

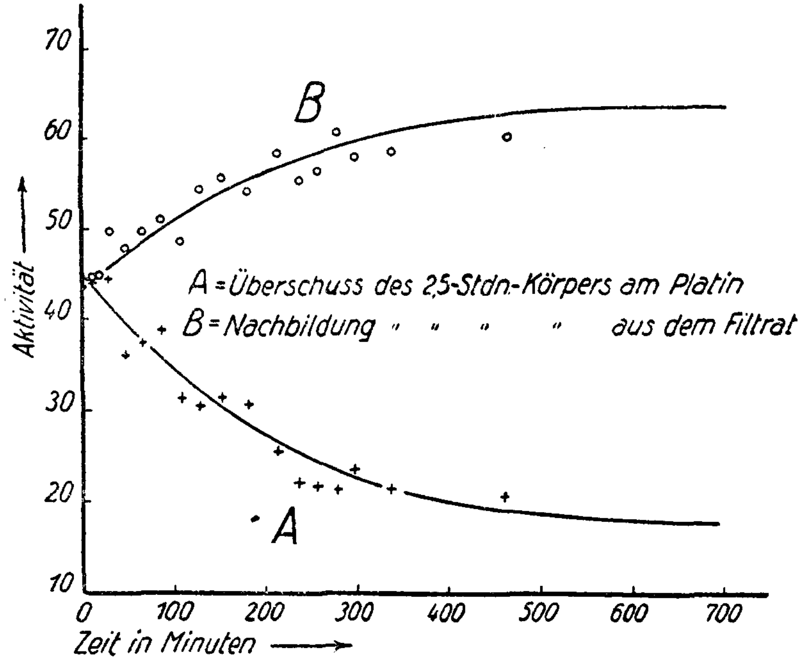


FIG. 4. The activity curves explaining the electrolytic separation of ekaplatinum from ekairidium (according to Hahn, Meitner, and Strassmann (48)).

Obviously these experiments of Hahn, Meitner, and Strassmann are not complete. Their excellent work, however, has paved the way for the establishment of the three different series formed and of the chemical nature of these various elements.

I. Curie and Savitch (18) have verified the greater portion of the results of the German group, although they stress that certain separations as outlined by Hahn *et al.* are difficult to carry out, that UX is very troublesome, and that all of the transuranium elements are produced under almost identical conditions. They observed and separated chemically the substances exhibiting half-lives of 16 min., 23 min., 59 min., and 2.7 days, their results being in accord with those of Hahn, Meitner, and Strassmann.

D. The existence of another transformation process

It will be recalled from the results of the German workers that difficulty was experienced, owing to an activity apparently associated with an element of atomic number less than 92. This behavior was finally explained as being due to a Szilard–Chalmers reaction.

The problem of counteracting the effect of UX was undertaken by I. Curie and Savitch (18), who used filters around their counter to absorb the β-rays of this element. A copper filter of 0.1 g. per $cm.^2$ cross section was found to absorb one-half of the β-rays of UX, a 0.5 g. per $cm.^2$ filter absorbed down to 5 per cent, and a 0.75 g. per $cm.^2$ filter absorbed to about 2 per cent, this last residue being attributed to gamma rays. Accordingly they measured the radiations from bombarded uranium penetrating the copper filters of 0.5 g. per $cm.^2$ The uranium preparations had been subjected to long periods of bombardment, the samples being in thick layers and large quantities (4 g. per 10 $cm.^2$).

For their activity measurements the French scientists used a compensating Hoffmann electrometer in measuring the ionization. Two thick uranium preparations, only one of which had been irradiated, were placed in two identical chambers connected with the electrometer. Hence the difference in the ionization currents produced was determined, the current due to the formation of UX being automatically compensated for during the experiment. For preparations bombarded over long periods of time, it was observed that the 3.5-hr. activity seemed to be dominant after the disappearance of the 16-min. activity. Their estimate of the number of β-particles emitted by the 3.5-hr. isotope, for thin preparations bombarded to saturation, was about one-fourth the initial number of β-rays from the 16-min. ekarhenium.

The periods observed were (*a*) two short periods (2 min. and probably the 40-sec. period); (*b*) the 16-min. period; and (*c*) a period of 3.5 hr. They have presented two explanations for this 3.5-hr. material, the earlier suggestion being that it was isotopic with thorium, and the more recent idea being that it is an actinium isotope.

Reviewing the first suggestion, the French investigators reported that the 3.5-hr. substance did not precipitate with hydrogen sulfide during the separation of uranium and the transuranium elements. The substance could be separated from uranium, however, apparently going with UX during the separation of this element from uranium. Since it seemed somewhat similar to UX in chemical behavior, and since UX presents difficulties in activity measurements, accurate determinations were extremely bothersome to make. The quantities of uranium used were large (15 to 20 g.); the presence of residual beta and gamma rays of UX affected the physical measurements.

I. Curie and Savitch (18) postulated, at first, that the 3.5-hr. substance was isotopic with thorium, and was formed by neutron bombardment with the emission of an alpha particle as follows:

$$^{238}_{92}\mathrm{U} + ^{1}_{0}n \rightarrow ^{235}_{90}\mathrm{R}_{3.5\,\mathrm{hr.}} + ^{4}_{2}\mathrm{He}$$

According to this scheme, the product of the disintegrating 3.5-hr. substance would be an isotope of protoactinium.

I. Curie and Savitch (18, 19) reported in both articles that this radioelement formed with both fast and slow neutron bombardment under almost the identical conditions for preparing the transuranium elements. Originally their thought was that the 3.5-hr. substance was a derivative of ekarhenium, because of the similarity in the conditions for producing these two active substances. The chemical separations indicated that they were not genetically related, and as a result the formation of the thorium isotope was suggested.

Since Hahn and Meitner could not find an isotope of thorium in the activated uranium, Curie and Savitch (19) investigated the 3.5-hr. material with still greater care. In this latter investigation they dissolved the irradiated uranium in hydrochloric acid, added some platinum and lanthanum, and finally ammonium carbonate. The lanthanum precipitate thus formed was redissolved in acid. A small amount of thorium (free from thorium X and the active deposit) was added to the solution, which was again treated with ammonium carbonate. This procedure was repeated twice. Uranium and those of the transuranium elements that are slightly soluble under these conditions were thus removed. Finally the precipitated lanthanum was redissolved in 2 N hydrochloric acid, some platinum added, and the remaining transuranic elements precipitated by hydrogen sulfide. The lanthanum fraction contained the 3.5-hr. substance and a trace of UX.

In their later report Curie and Savitch (19) stated that this active body (*a*) is not precipitated by hydrogen sulfide from 2 N hydrochloric acid, (*b*) is precipitated by ammonium carbonate, (*c*) is not precipitated by potassium iodate in concentrated nitric acid solution (difference between Th and UX), (*d*) is not precipitated by phosphoric acid from 2 *N* hydrochloric acid in the presence of zirconium (difference from protoactinium), and (*e*) is precipitated by oxalic acid from a weakly nitric acid solution in the presence of lanthanum (analogous to the rare earths and actinium).

These chemical properties caused the French workers to suggest that the material is either an actinium isotope or a new transuranium element possessing chemical properties entirely different from those of the higher homologs of rhenium and the platinum metals. Both hypotheses are difficult to visualize, and the investigators suggest that considerable more research will be necessary to clarify the problem.

Should the chemistry of this active substance as reported by I. Curie and Savitch prove to be correct, one might conjecture as to the possibility of *K*-electron capture in addition to the emission of an alpha particle to explain the transition from uranium to actinium as a result of neutron bombardment. To change from uranium to actinium, the atomic number must be decreased by three units. This may be effected by having either a proton or a positron and an alpha particle emitted; or a *K*-electron capture in conjunction with alpha emission must occur. A possible reaction might be

$${}_{92}\mathrm{U} + {}_{0}^{1}\mathrm{N} + (K\text{-electron}) \rightarrow {}_{89}\mathrm{Ac} + {}_{2}^{4}\mathrm{He}$$

Naturally it is impossible to consider the masses involved.

The evidence favoring *K*-electron capture is very meager. To begin with, no elements beyond erbium (No. 65) have been found to exhibit positron emission, this latter effect often being associated with the electron capture. Furthermore, the fairly conclusive evidence for the 10 sec.-, 40 sec.-, and 23 min.-isotopes of uranium is indicative that the *K*-electron capture probably would be very difficult to bring about. These short periods indicate that it is relatively easy to activate the uranium for the formation of the three transformation series suggested by Hahn. Also, there is no evidence of *K*-electron capture in the natural radioactive series, which includes these same elements. This conjecture is worthy of consideration only if no definite evidence of a simpler type of change is observed.

Hahn and his coworkers have not reported any evidence of the 3.5-hr. material. It is entirely possible that these investigators might have had the correct mixture of the 3.5-hr. substance along with certain of the naturally occurring isotopes (including the 6.7-hr. UX) to give decay curves which might not, on first analysis, have given any indication of the 3.5-hr. element. The question is still open.

A meager amount of additional evidence for the existence of the 3.5-hr.

substance is indicated in some unpublished work of Pool (68). Metallic uranium was bombarded for 6.5 hr. with fast neutrons produced by the lithium–deuteron reaction. The deuteron source was the Michigan cyclotron. Both the beta and the gamma activities of the uranium were determined. The beta decay curves showed an intense 3.7-hr. activity. The gamma decay curves showed a 3.3-hr. activity for the same sample. The changing slope of the curves would easily lead one to believe that the period could be 3.5 hr., which would agree with the results of Curie and Savitch. No chemical separations were made, all results being strictly dependent on the activity measurements. The intensity of the neutrons developed with the cyclotron is tremendously greater than that of the radon–beryllium sources, hence it is conceivable that this method might intensify processes difficult to bring out by the lower energy sources. I. Curie and Savitch report that their sources gave about 300 to 1000 millicuries with 13 M.e.v. as the upper limit for neutrons. The intensity of neutrons from the lithium–deuteron reaction (with the cyclotron) is several Curies with about 20 M.e.v. as the upper limit for neutrons.

The 3.5- and 3.7-hr. periods found by Pool are dominant over the extremely short-lived periods. For the strongly activated uranium (bombardment for 6.5 hr. with fast neutrons) the intensity of the beta emission was approximately 23 μc, with a saturation value of 33 μc. This intensity was recorded after the short-lived substances had completely disappeared. This observation of the dominant nature of the 3.3- to 3.7-hr. periods by Pool corresponds to the observations of the French investigators.

Abelson (1), using the Berkeley cyclotron, activated uranium with neutrons, and reported that the extensive work of Hahn was verified, for the most part, by his results. The emitted particles were found to be negative in all cases. He verified the contention that at least two separate chains of β-emitting substances result from the activation of uranium by thermal neutrons. In addition to the periods reported by the French, Italian, and German groups, Abelson reported the detection of a 17-hr. period. Activated material was examined for α-particles within 5 min. after cessation of bombardment, using a linear amplifier. It was concluded that α-active isotopes of half-life periods less than 1000 yr. were not present in amounts comparable to the β-active isotopes.

A 16.3-hr. period was noted by Pool (68), which might correspond to the above 17-hr. period. Abelson did not report a 3.5-hr. activity.

E. The energy relationships involved

The German group has made an exhaustive survey of the energy relationships necessary to activate uranium and bring about the different transformation series. In contrast to the suggestion of Curie and Savitch, they have no definite evidence of alpha emission. They report only the chains of β-particles, which is an effect not observed in the natural disintegration series.

Meitner *et al.* (63) used a Geiger-Müller counting tube of aluminum of 0.1 mm. wall thickness and an amplifying system when determining the genetic relationships. Their counters were built into thick lead chambers into which the preparations were placed in small lead boats.

To observe the very short-lived uranium isotopes, UX-free uranium preparations were bombarded for very short periods (15 to 60 sec.) with fast neutrons to avoid formation of the 23-min. isotope. In figure 5 curve A is the activity for a 13-sec. bombardment; curve B for 30 sec.

The existence of several substances of very short periods is suggested. In curve B the slope indicates the presence of a 2.2-min. material, shown to be ekarhenium chemically. Subtracting this value, the resultant is curve B, which likewise is not straight. It indicates that a 40-sec. period is present. Making another subtraction curve B results, which is a straight-line exponential of 10-sec. half-life. This and the 40-sec. value are characteristic of two different isotopes of uranium, as proven chemically. These being the shortest periods and not chemically separable from uranium, they must head the families. The quantity of the 2.2-min. substance is affected by the period of irradiation, hence it must be a disintegration product of the 10-sec. uranium.

By varying the time of bombardment the Germans showed that the 16-min. substance was a disintegration product of the 40-sec. uranium. Since beta emission occurs, according to the radioactive displacement laws, the only possibility for the new element formed would be an element 93, ekarhenium. This verifies the original declaration by Fermi that transuranium elements are formed. The chemical separations made by both the Italian and German groups substantiate the contention.

Using longer irradiation periods, Meitner *et al.* proved that the 59-min. activity was due to an ekaosmium isotope formed from the 2.2-min. ekarhenium, and that the 66-hr. active principle (ekairidium) changed into ekaplatinum of 2.5-hr. half-life. Ekaplatinum was essentially inactive.

The German group reported that materials subjected to extremely long periods of bombardment exhibited a very weak but detectable alpha emission in a Wynn-Williams proportional amplifier. No evidence of α-activity was noted for any material subjected to short periods of bombardment. The suggestion made was that a long-living alpha-emitter, whose genetic relationship has not been determined, results from long bombardment.

Likewise the evidence showed that the 16-min. ekarhenium changed to the 6-hr. ekaosmium. Earlier the value was given as 10 to 12 hr., then as 6, and the newer value as 5.7 hr.

Whether ekairidium is formed from this ekaosmium isotope is still a question. If active, the ekairidium is probably very long-lived. Meitner, Hahn, and Strassmann suggest that some of their residues have exhibited a very long period.

For the third process (see page 126), evidence has been secured for only one transformation product, a beta-emitting uranium isotope of 23-min. period, which goes into ekarhenium. The latter may be long-lived. The 66-hr. substance mentioned above is definitely not related to this ekarhenium because the former substance results from fast neutron bombardment, whereas the 23-min. uranium isotope results only from slow neutron irradiation.

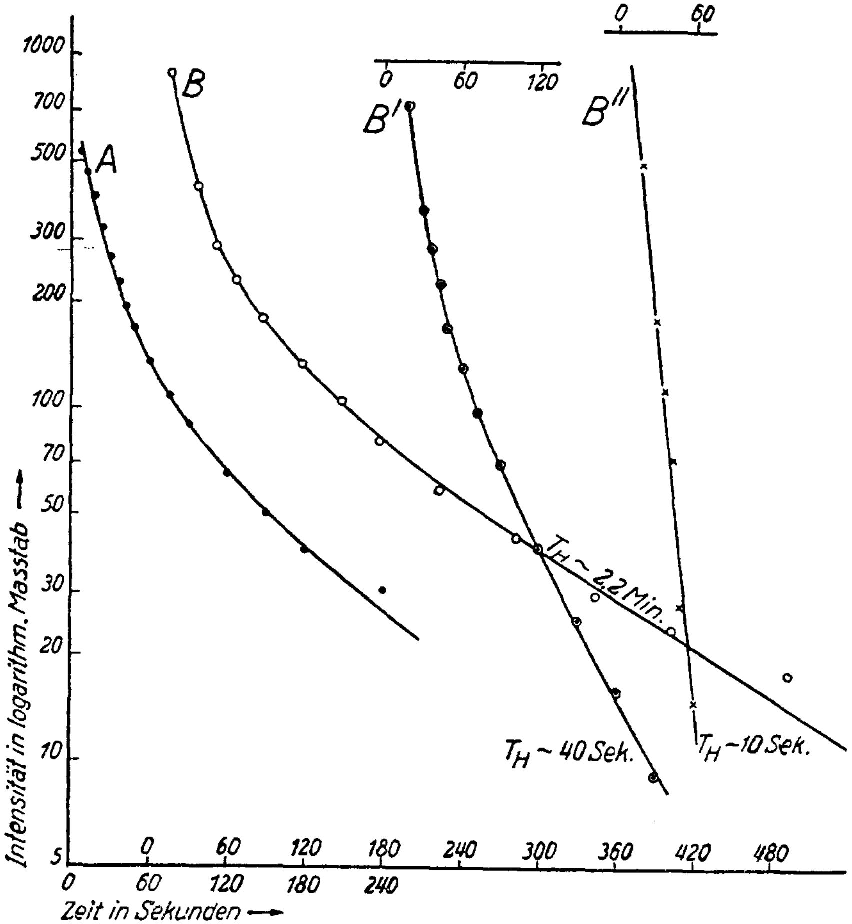


FIG. 5. The activity curves showing the existence of the 10-sec., the 40-sec., and the 2.2-min. active substances (according to Meitner, Hahn, and Strassmann (63)).

1. The effect of fast and slow neutrons

After Fermi and Amaldi (35, 7) reported that different effects were obtained with fast and slow neutrons, Meitner, Hahn, and Strassmann (63) began a systematic study of the effect of slow and fast neutrons on uranium. They wished to observe the energies of the neutrons effecting the three transformation processes, and to determine the effect of filters on processes I and II.

Both processes I and II resulted from irradiation with fast as well as with thermal neutrons. When a cadmium filter was used during activation by fast neutrons the same relative intensities were always obtained. With slow neutrons the yield of active materials was made as follows: A paraffin cylinder of about 20 cm. cross section and 20 cm. length, with the neutron source (several radon-beryllium tubes) placed in the middle, was utilized. The preparations being studied were bombarded through different thicknesses of paraffin. Also, bombardments were made with and without a cadmium filter under 4 cm. of paraffin. The intensity of the activity was found to be reduced to about 87 per cent when a 0.42-cm. thick cadmium filter was placed on both sides of the uranium. Therefore, about 90 per cent of the activation was produced by thermal neutrons. This indicates also that the 66-hr. ekairidium (No. 95) and the 2.5-hr. ekaplatinum (No. 96) belong to process I or II and not to process III. The same result was noted after long bombardment (21 hr.) with and without the use of the cadmium filter. The concentration of the same two substances was decreased, in both cases, in the same proportion as the shorter-living uranium isotopes.

The German investigators reported that they were unable, within the limits of error, to detect any difference in the intensity for processes I and II using either thermal or rapid neutrons.

A series of experiments was performed in which (*a*) the uranium preparation was surrounded by paraffin, (*b*) a layer of paraffin was placed only between the sample and the neutron source, and (*c*) a cadmium filter was used for comparison experiments in studies *a* and *b*. With the uranium imbedded in paraffin, and using a cadmium filter, the activation was decreased to 21 per cent. That is, 80 per cent of the activation resulted from thermal neutrons and 20 per cent from fast neutrons. With paraffin only between the sample and the neutron source, bombardments, with and without the cadmium filter, showed that the induction was only 25 per cent, since thermal neutrons were eliminated by this treatment.

That the intensity of both processes I and II was the same even when the thermal neutrons were filtered makes it very probable that no resonance absorption exists in the energy ranges intermediate between fast and thermal neutrons. Beryllium neutrons possess definite energies between

10^5 to about 14×10^6 volts. Hence a definite statement of the effective energies of the fast neutrons is difficult to make. The measurements with fast neutrons *with* and *without* the cadmium filter always gave similar results. Thus the neutron sources did not give off primarily thermal neutrons.

Comparison experiments were also made by the German investigators, using thicknesses of uranium preparations ranging from 61 mg. of uranium per cm.^{2} to 1.27 g. of uranium per cm.^{2}, and with a cadmium filter 0.46 mm. thick. The intensities with and without the filter were again the same within the limits of experimental error.

The practical significance of these determinations is that one can bombard very thick layers of uranium with rapid neutrons to obtain an evenly activated product.

Filters possessing very definite resonance absorption regions in the range from 1 to several volts and which absorb thermal neutrons to only a slight degree did not cause any decrease in the intensities of the two processes.

2. The effective neutron energy for process III

Relatively large amounts of uranium were used for this determination, since only one uranium isotope is detected in the activated material. Using identical conditions for all of their experiments, as far as the amounts of uranium, etc., were concerned, it was shown that about 65 per cent of the activation was due to other than thermal neutrons when the uranium was imbedded in paraffin with a 4-cm. thickness of paraffin between the neutron source. Process III is therefore caused by slow neutrons.

To ascertain whether the activation by slow neutrons was due to resonance, the German group measured the auto-absorption in uranium. A thin layer of the uranium preparation containing 43 mg. of uranium per cm.^{2} was bombarded with neutrons, the source of which was but a short distance from the sample. The use of a cadmium filter decreased the intensity of activation about 5 per cent, indicating that 95 per cent of the intensity was certainly not due to thermal neutrons. A uranium filter of 61 mg. of uranium per cm.^{2}, used with and without the cadmium, diminished the intensity by neutrons other than thermal neutrons by about 31 to 34 per cent. This gave a mean mass absorption coefficient of $\mu/\rho = 7$ cm.^{2} per gram, due to strong absorption in the uranium. Allowing for the angle at which the neutrons travelled, the actual mass coefficient was calculated as $\mu/\rho = 3$ cm.^{2} per gram. The coefficient for thermal neutrons, as determined by Dunning (26) and his coworkers, was found to be 0.1 cm.^{2} per gram. Thus the slow neutrons causing activation of the uranium show a mass coefficient of absorption at least thirty times greater.

Meitner *et al.* (63) also determined the energy of the resonance neutrons by comparing the absorption of these neutrons in boron with the value for thermal neutrons in boron using the same experimental conditions. For this calculation the relationship

$$E_R = E_{\text{th}} \left(\frac{\mu_{\text{th}}}{\mu_R}\right)^2$$

may be used. μ_{th} is the absorption coefficient of the thermal neutrons in boron, the value of which was determined by Rasetti and Goldsmith (69). μ_R is the resonance absorption coefficient and was determined by Meitner. The resonance energy was found to be 25 ± 10 e.v., which is the resonance level of the slow neutrons activating process III.

$$_{92}\text{U} + n \rightarrow {}_{92}\text{U}; \qquad {}_{92}\text{U} \xrightarrow[\text{23 min.}]{\beta} {}_{93}\text{Eka-Re}$$

3. The energy of the emitted beta rays

Meitner (61) made a careful study of Wilson photographs of the sulfide precipitates obtained from preparations bombarded for different periods in order to determine the β-particle energies. The analysis of over 400 β-emissions showed a range from about 3200 kv. to 600 kv. In noting which products were present at any instant with the corresponding time of β-emissions, it was concluded that those emissions ranging from 1000 kv. upward, with the maximum of the beta-ray spectrum in the neighborhood of 3200 kv., were due to eka-$\text{Re}_{93}^{16\text{ min.}}$. The eka-$\text{Os}_{94}^{59\text{ min.}}$ has fewer penetrating primary β-emissions, the limit for the more rapid betas being about 600 kv. For samples bombarded for several months, it was decided that the maximum of 700 kv. observed was due to the β-particles from eka-$\text{Pt}_{96}^{2.5\text{ hr.}}$, since eka-$\text{Ir}_{95}^{66\text{ hr.}}$ is very feebly β-active.

Curie and Savitch (18) found slightly different results in their β-ray study. For eka-$\text{Re}_{93}^{16\text{ min.}}$ they report two groups of beta particles, one resembling those of UX, and in the range of 2×10^6 e.v., representing 90 per cent of the intensity; the other is very penetrating and mixed with gamma radiation. They suggest that Meitner's value of 3×10^6 e.v. may probably correspond, not to the upper limit of the spectrum, but to the lower region of the penetration group. They also comment that the energies of the β-rays of the 2-min. and the 3.5-hr. substances should be at least of the order of 6×10^6 e.v.

F. The processes of formation of the artificial radioelements

Since the experimental evidence indicates three and perhaps four separate disintegration series resulting from irradiated uranium, it does not seem possible to interpret the numerous radioelements without calling upon the hypothesis of isomerism. Accordingly, for the moment, three processes will be postulated as follows:

$$ {}^{238}_{92}\mathrm{U} + n\left\{\begin{matrix}\text{fast}\\ \text{slow}\end{matrix}\right\} \rightarrow {}^{239}_{92}\mathrm{U} \xrightarrow[\text{10 sec.}]{\beta} {}^{239}93 \xrightarrow[\text{2.2 min.}]{\beta} {}^{239}94 \xrightarrow[\text{59 min.}]{\beta} $$

$$ {}^{239}95 \xrightarrow[\text{66 hr.}]{\beta} {}^{239}96 \xrightarrow[\text{2.5 hr.}]{\beta} {}^{239}97 \qquad \text{(I)} $$

$$ {}^{238}_{92}\mathrm{U} + n\left\{\begin{matrix}\text{fast}\\ \text{slow}\end{matrix}\right\} \rightarrow {}^{239}_{92}\mathrm{U} \xrightarrow[\text{40 sec.}]{\beta} {}^{239}93 \xrightarrow[\text{16 min.}]{\beta} {}^{239}94 \xrightarrow[\text{5.7 hr.}]{\beta} {}^{239}95 \rightarrow\ ? \qquad \text{(II)} $$

$$ {}^{238}_{92}\mathrm{U} + n\,(\text{slow}) \rightarrow {}^{239}_{92}\mathrm{U} \xrightarrow[\text{23 min.}]{\beta} {}^{239}93 \qquad \text{(III)} $$

It is noted that the mass number assigned to the radioelements is 239 in each case. One might ask why an atom of mass 237 is not suggested. This would be in accordance with observed experimental facts that under fast neutron bombardment two neutrons are ejected and the resulting element has a mass one less than the original, represented as follows:

$$ {}^{M}_{Z}\mathrm{R} + {}^{1}_{0}n\,(\text{fast}) \rightarrow {}^{M-1}_{Z}\mathrm{R} + 2\,{}^{1}_{0}n $$

No experimental evidence thus far indicates such a possibility. In most cases bombardment of a given element with fast neutrons and then with slow neutrons will give two different periods, thus enabling the investigator to say with certainty which change is occurring, an increase or a decrease of one unit in atomic weight. Such a procedure is a standard method for identifying those isotopes giving rise to the various observed periods. However, as already mentioned, the intensities of the activities noted for both fast and slow neutron irradiation of uranium are essentially the same. This means, therefore, that processes I and II are very probably isomeric. Thus if the atomic mass is 239 for process I it must be the same for process II.

There can be no doubt about process III, since it is a simple neutron capture of the type

$$ {}^{M}_{Z}\mathrm{R} + {}^{1}_{0}n \rightarrow {}^{M+1}_{Z}\mathrm{R} $$

Thus the first members of these series are isomers (isobaric isotopes).

Since processes I and II represent long isomeric series it is logical to inquire if it is possible for one isomeric nucleus to change to a second isomeric nucleus by liberation of energy. Such a transformation is not at all improbable, since either gamma emission or prohibited beta-particle transition is possible. Gamma rays have been recorded by Meitner, Pool, and others in their studies. It is also very probable that the β-ray transition of ekaplatinum to ekagold is a prohibited transition (due to a change of impulse momentum). The studies by Meitner on the γ-emission showed that the ekaosmium (59 min.) exhibited a much more intense γ-radiation than did ekarhenium (16 min.), these elements being in the different isomeric series. This indicates, if the Weiszacker representation is correct, that process I starts from a metastable state of U^{239}, whose impulse momentum must be considerably greater than that of the U^{239} which heads process II.

Another manner of looking at this concept is to assume that X, Y, and Z are three members of a disintegration series, and that X^1, Y^1, and Z^1 are the corresponding nuclear isomers of lower energies. Transformations such as $X \xrightarrow{\beta} Y$; $X^1 \xrightarrow{\beta} Y^1$, etc., are normal radioactive changes. There is also the possibility of changes such as $X \xrightarrow{\beta} Y^1$ and $Y \xrightarrow{\beta} Z^1$, probably associated with γ-emissions.

Other probable changes are transitions such as $X \xrightarrow{\gamma} X^1$, $Y \xrightarrow{\gamma} Y^1$, $Z \xrightarrow{\gamma} Z^1$ which might occur, especially if the energy differences between the metastable and the stable isomeric nuclei are small in comparison with the energies of emission of beta rays. With respect to processes I and II, it could be conjectured that one of these families is more metastable than the other, thus leading to transitions between the two. As Curie and Savitch point out, one might assume that the 66-hr. substance of process I might be formed in part from the 5.7-hr. element 94 of process II, since there is no definite evidence permitting complete verification of the change that Hahn *et al.* have postulated, namely, that the 66-hr. material is a product of the 59-min. ekaosmium.

Naturally active uranium is a mixture of three isotopes, 234 (UII), 235, and 238 (their relative abundance is 0.007:0.00006:1), any of which might be activated. Processes I and II result from fast neutron (also slow) bombardment. Meitner *et al.* have shown the effective cross section (for U^{238}) to be 1.6×10^{-25} cm.^{2} Isotopes 234 and 235 are probably not the parent substances of processes I and II, because the magnitude of the effective cross section would need to be so great that fast neutrons would be entirely ineffective. Both processes must start from U^{238}.

Process III probably can not originate from U^{235}, since from the measured absorption coefficients in uranium the resonance absorption cross section should be about $3{,}000{,}000 \times 10^{-24}$ cm.^{2}, which is considerably greater than $\lambda^2/4\pi$, where λ is the proper de Broglie wave length. It is probably a simple capturing process originating also with U^{238}, thus giving a third isomeric nucleus U^{239}.

That one of these processes may result in a series of mass number 238 can not be disregarded. Such a family could be headed by U^{238}, which is changed to a metastable state from the stable U^{238} merely as a result of absorbing some energy from the neutrons without capturing one of the particles or losing one from its own nucleus. Such a possibility could be proven by subjecting uranium to gamma radiation of high energy.

There is no doubt whatsoever that considerable more thought and research must be done before one can state definitely which isotopes are involved in these transformations, how the series are related, and what the energies involved are.

VI. SUGGESTED FUTURE STUDIES ON THE TRANSURANIUM ELEMENTS

The interesting puzzle of the transuranium elements is only partially solved. The excellent work of the scientists who have been investigating this field has contributed to the solution, but at the same time has brought forth new questions to be answered. A few suggested factors which should be investigated are the following:

1. Now that some of the chemical properties of ekarhenium are known, minerals should be investigated for the presence of this element. The probability of discovery is very slight, but the possible existence of an isotope of 93 of exceptionally long half-life should not be overlooked.
2. An exhaustive study of these elements should be made, using intense sources of neutrons (available at those places having cyclotrons, such as the Universities of California, Michigan, Rochester, etc.) Long and short bombardments should be made so that a very complete range in the activating periods may be utilized.
3. A study using monokinetic sources of neutrons (such as those produced with the deuteron–deuteron reaction) may produce valuable information about certain of the energy changes involved.
4. The investigations on the emission of α-particles, which have been started, should be extended, using the more energetic neutron sources.
5. Careful observations should be made to ascertain whether alpha particles, positrons, etc., are being emitted only during the bombardment period. The evidence on this point up to the present is negative.
6. Special precautions should be taken to observe whether any exceptionally long-lived substances which are very feebly active are formed.
7. The possible formation of isotopes of thorium, protoactinium and other elements of lower atomic number demands further study.
8. The bombardment of uranium with intense gamma radiation should permit, if activation resulted, a definite statement as to whether one of the disintegration series mentioned earlier has a U^{238} isotope as the head of the family. Perhaps gamma activation might give rise to still other series.
9. Deuteron bombardment is known to activate uranium. Owing to

TABLE 13

A summary of the properties of the transuranium elements

(Based essentially on the work of Hahn, Meitner, and Strassmann)

ELEMENTS	SIMILARITIES	DIFFERENCES
Rhenium and ekarhenium.	Neither is precipitated from acid solution on the noble metals (Pt) Neither is precipitated by metallic bismuth in alkaline solution Both are quantitatively precipitated by hydrogen sulfide from hydrochloric acid solutions of moderate concentration, and are not so easily precipitated from highly acid solutions Both are precipitated by nitron Both are volatile as the oxide or the chloride at high temperatures	Ekarhenium is not volatile from a sulfuric acid solution even in the presence of a stream of hydrogen chloride
Osmium and ekaosmium...	Both are precipitated by metallic bismuth in alkaline solution Both are quantitatively precipitated by hydrogen sulfide even from strongly acid solutions Both are quantitatively precipitated by sodium hydroxide Both are volatile as oxides at higher temperatures	Ekaosmium does not distill from a nitric acid solution Ekaosmium is precipitated from acid solutions by platinum
Iridium and ekairidium...	Both separate with metallic bismuth in alkaline solution Both are almost completely precipitated by hydrogen sulfide from weak hydrochloric acid solutions	Ekairidium is appreciably volatile when heated strongly Ekairidium precipitates on platinum from acid solutions Ekairidium does not tend to form mixed crystals with platinum ammonium chloride to any extent Ekairidium is only partially precipitated by sodium hydroxide

TABLE 13—*Concluded*

ELEMENTS	SIMILARITIES	DIFFERENCES
Platinum and eka-platinum	Both precipitate well from hydrochloric acid solutions of not too high a concentration Mixed crystals of ammonium platinum chloride and ammonium eka-platinum chloride may be formed Both are incompletely precipitated by sodium hydroxide	Ekaplatinum as the oxide is appreciably volatile when ignited gently Ekaplatinum is precipitated from acid solution by platinum

TABLE 14

Observed beta-emitting transformation products

ISOTOPE	HALF-LIFE PERIOD	INVESTIGATORS
92 Uranium	10 sec., 40 sec., 23 min.	Hahn *et al.*
	10 sec., 40 sec., 23 min.	Fermi *et al.*
	40 sec., 23 min.	Curie and Savitch
93 Ekarhenium (Ao)	2.2 min., 16 min.	Hahn *et al.*
	2.2 min., 13 min.	Fermi *et al.*
	2 min., 16 min.	Curie and Savitch
94 Ekaosmium (Hs)	59 min., 5.7 hr.	Hahn *et al.*
	59 min., 10 hr.	Fermi *et al.*
95 Ekairidium	66 hr.	Hahn *et al.*
	3 days	Fermi *et al.*
	2.7 days	Curie and Savitch
96 Ekaplatinum	2.5 hr.	Hahn *et al.*
	2.5 hr.	Fermi *et al.*
Actinium, thorium, or protoactinium or ?	3.5 hr.	Curie and Savitch
	3.3–3.7 hr.	Pool
?	17 hr.	Abelson
	16.3 hr.	Pool

the fact that elements of low atomic weight are very easily activated by deuterons, traces of these elements in uranium could cause extreme difficulty. Deuteron bombardment might be of value in helping to establish the various transformation series.

10. Although not highly probable, the capture of *K*-electrons by the nucleus should be studied. The suggestion given in certain theoretical considerations that there must be a point at which the volume of the nucleus and that of the *K*-electronic orbit approach the same value lends strength to this idea. On the other hand, the fact that no *K*-electron capture has been observed in connection with the naturally radioactive series and that there is no definite evidence of positron emission beyond erbium ($Z = 68$) tends to indicate that *K*-electron capture in uranium is not very probable.

11. The chemical and physical properties of these elements require further study so as to effect a better correlation of their properties with those of the known elements, as well as to enable scientists to perfect the methods of separation and identification necessary in these studies of the artificially produced radioelements.

12. The entire field of energy relationships involved requires further study. The results thus far obtained are not as indicative as they might be of the various relationships existing between energies of activation, of disintegration, etc.

VII. A SUMMATION OF THE PROPERTIES OF THE TRANSURANIUM ELEMENTS

The properties of the transuranium elements are summarized in table 13. In table 14 are given the observed beta-emitting transformation products.

VIII. SUMMARY

1. No evidence of the existence in minerals of elements beyond uranium has been established.

2. A number of conjectures as to the total number of possible elements have been published.

3. Predictions of the properties of elements beyond uranium have been postulated.

4. Uranium may be readily activated by neutrons. The contention of Fermi that transuranic elements resulted from the disintegration of the activated uranium has been substantiated by workers in his own laboratory, by Hahn, Meitner, and Strassmann, by Curie *et al.*, by Abelson, and others.

5. Three isomeric nuclear series have been postulated to explain the observed transformations. A fourth process involving alpha radiation

and the formation of isotopes having atomic numbers less than 92 can not be overlooked.

6. Energy studies of the transformation processes have been made, but the results as yet are not sufficiently adequate and conclusive to explain all effects involved.

7. The chemical properties of these radioelements have been established sufficiently accurately and carefully to permit of separation and identifications. The chemistry of these elements is complex and not yet perfected.

8. The study of these elements is complicated by the chemical and the radioactive character of the other elements in this region of the periodic classification.

9. The continuous liberation of beta particles in processes I and II is a very interesting phenomenon. It has no counterpart in the natural radioactive series.

10. The names "ausonium" (Ao) and "hesperium" (Hs) have been mentioned by the Italian investigators for elements 93 and 94, respectively.

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