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ELECTRONIC STRUCTURE OF THE HEAVIEST ELEMENTS

By G. T. Seaborg

July 10, 1948

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ELECTRONIC STRUCTURE OF THE HEAVIEST ELEMENTS

By G. T. Seaborg

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ELECTRONIC STRUCTURE OF THE HEAVIEST ELEMENTS

By Glenn T. Seaborg

Abstract

All of the available evidence leads to the view that the 5f electron shell is being filled in the heaviest elements giving rise to a transition series which begins with actinium in the same sense that the rare earth or "lanthanide" series begins with lanthanum. Such an "actinide" series is suggested on the basis of evidence in the following lines: (1) chemical properties, (2) absorption spectra in aqueous solution and crystals, (3) crystallographic structure data, (4) magnetic susceptibility data and (5) spectroscopic data. The salient point is that the characteristic oxidation state (i.e., the oxidation state exhibited by the member containing seven 5f and presumably also by the member containing fourteen 5f electrons, curium and element 103) is the III state, and the group is placed in the periodic table on this basis. The data also make it possible to give a suggested table of electronic configurations of the ground state of the gaseous atom for each of the elements from actinium to curium inclusive.

ELECTRONIC STRUCTURE OF THE HEAVIEST ELEMENTS

By Glenn T. Seaborg

I. Historical Background

A. Before the Discovery of the Transuranium Elements. The intensive study of the heavy elements during the last few years has given information and data which now enable us to make some definite statements as to their electronic structure. The information obtained about the recently discovered synthetic transuranium elements has been particularly useful in this connection, and it is largely on the basis of these new elements that this question is now well understood.

The heaviest natural elements, thorium, protactinium and uranium, of atomic numbers 90, 91 and 92, respectively, lie in corresponding positions just below the 6th period "transition" elements, hafnium, tantalum and tungsten, in which the 5d electron shell is being filled. The elements, hafnium, tantalum and tungsten are similar in their chemical properties to the corresponding transition elements in the 5th period, zirconium, columbium, and molybdenum, in which the 4d shell is being filled.

It has long been known that the chemical properties of thorium, protactinium and uranium resemble those of these 4d and 5d elements and for this reason most of the textbooks and standard works on chemistry and physics in which the electron structure is discussed have accepted the view that it is the 6d shell which is being filled. Thus the structure of the elements above radon (element 86) through uranium is written to show the addition of the next two electrons in the 7s shell for element 87 (francium) and element

88 (radium) and addition in the 6d shell for the following four elements, actinium, thorium, protactinium and uranium(1).

Many of the early papers which appeared after N. Bohr's classical work(2) on the quantized nuclear atom discuss the electronic structure of the heaviest elements. There has been general agreement that some type of transition group should begin in the neighborhood of these elements, although there has been difference of opinion as to where it begins and as to which electron shells are involved. A number of the earliest publications even have suggested that this transition series involves the filling of the 5f shell, thus possibly giving rise to a "rare earth" group in a manner analogous to the filling of the 4f shell. This filling of the 4f shell results in the well known group of 14 rare earth elements of atomic numbers 58-71 inclusive, following lanthanum. It is of interest here to note a few of these early and also later suggestions in order to review the general previous status of this question. Most of these early investigators were of the opinion that the filling of the 5f shell should begin at some point beyond uranium, that is, beyond the then known elements.

In an early paper Bohr(3) suggested that the addition of the 5f electrons might begin in this region, and in a Bohr-Thomsen type of periodic table he pictured the first entry at the element with atomic number 94. Y. Suguira and H. C. Urey(4) gave calculations indicating that the first entry of the electron into the 5f shell should occur at element 95. J. C. McLennan, A. B. McLay and H. G. Smith(5) suggested as an alternative to the filling of the 6d shell the possibility that the 5f shell begins to be occupied in thorium. In a review article, S. Dushman(6) stated it is doubtful that the added electrons enter the 6d level (thus implying an analogy with cerium, etc.). V. Karapetoff(7) and Ta-You-Wu and S. Goudsmit(8) suggested that the element with atomic number 93 might be the first in which the 5f shell begins to be filled, while A. von Grosse(9) suggested, as a possible alternative to filling of the 6d shell, the entry of the first electron in the 5f shell with uranium. More recently L. L. Quill(10), largely for the purpose of illustration, presented periodic table arrangements in which the first 5f electron appears in element number 95 in one case and in element number 99 in another. The later calculations of M. Goeppert Mayer(11) indicate that the filling of the 5f shell might begin at protactinium or uranium. J. Perrin(12) and R. Rudy(12a) /on general considerations, proposed as a possibility the theory that the first 5f electron appears in thorium and G. E. Villar(13) suggested that some of the chemical evidence supports this viewpoint.

On the basis of his crystallographic work, V. M. Goldschmidt(14) favors the view that the first 5f electron enters at protactinium, the first element beyond thorium, although he points out the possibility that this may occur either earlier, in thorium, or later, in uranium or in the (at the time unknown) transuranium elements. By analogy with the name "lanthanide" series which he had already proposed for the rare earth elements because these 14 elements following lanthanum have lanthanum as their prototype, he proposed the name "thoride" series for the 14 elements following thorium. On the basis of his much more complete crystallographic evidence, including especially observations on the transuranium elements, W. H. Zachariasen(15) has come to the same conclusion.

B. After the Discovery of the Transuranium Elements. The recent discovery of the transuranium elements and the study of their properties, especially the chemical properties, have given us a tremendous amount of additional evidence of just the type needed to clarify this problem. As it turns out, it is in the transuranium elements that the really definitive chemical properties, from the standpoint of placing the heaviest elements in the periodic table, first appear. The first best definite evidence that the 5f shell undergoes filling in this heavy region came from the tracer chemical observations of E. M. McMillan and P. H. Abelson(16) on element 93 (neptunium); upon their discovery of this, the first transuranium element, they were able to show definitely that it resembles uranium in its chemical properties and bears no resemblance to rhenium, the element immediately above it in the periodic table. This excellent experimental evidence was interpreted by them to indicate that this new "rare earth" group of similar elements starts with uranium. K. Starke(17) and C. G. Bedreag(18) also interpreted the tracer experiments with element 93 as indicating that the first 5f electron comes at element 93, while F. Strassmann and O. Hahn(19) felt on the basis of their tracer experiments with this element, that it was difficult to make any deduction. As a result of their first tracer experiments with element 94 together with their consideration of the tracer investigations with element 93, G. T. Seaborg and A. C. Wahl(20) in 1942 made the suggestion that this transition group might begin as early as with thorium or actinium.

Since this first tracer work on the transuranium elements, the elements neptunium (atomic number 93) and plutonium (atomic number 94) now have been extensively investigated with substantial, weighable quantities. Americium (atomic number 95) and curium (atomic number 96) also have been available for investigation, on a more limited scale, in weighable quantities. The recent extensive investigations of thorium, protactinium and uranium also have contributed to the evidence which is now useful to interpret this question.

II. Actinide Concept

A. General. The evidence now available leads to the definite view that it is the 5f electron shell which is being filled in these heaviest elements. The evidence seems sufficient to go further than this and to suggest(21) that this rare-earth-like series begins with actinium in the same sense that the rare earth or "lanthanide" series begins with lanthanum. On this basis it might be termed the "actinide" series(21) and the first 5f electron might appear, although not necessarily, in thorium. The salient point is that the characteristic oxidation state (i.e., the oxidation state exhibited by the member containing seven 5f and presumably also by the member containing fourteen 5f electrons, curium and element 103) is the III state.

There is much evidence, all pointing toward this view. We will discuss this evidence which is in the following lines: (1) chemical properties, (2) absorption spectra in aqueous solution and crystals, (3) crystallographic structure data, (4) magnetic susceptibility data and (5) spectroscopic data.

It should be emphasized that the discussion so far has been carried on in a somewhat oversimplified manner, because the details concerning the possible physical or chemical forms in which these elements might exist have a bearing on the electronic structure, as is the case for all the other transition groups including the rare earth elements. Thus the number of 5f electrons in the atom in the gaseous state might differ from that in the metallic state (which in turn can differ from one phase to another), and in turn neither one of these structures might correspond directly to the chemical properties. It is the chemical properties (including absorption spectra, crystallographic data, etc. on the compounds) which should be determinative in placing these elements in the periodic table, just as was the case for the rare earth elements which would be placed differently considering their electronic structures alone.

B. Chemical Evidence. On the basis of an actinide series the characteristic oxidation number for the series is III, and this shows up strikingly in the stabilization of the lower oxidation states with increasing atomic number. In going from uranium to plutonium it becomes increasingly difficult to effect the oxidation from the IV to the VI state and, in fact, with americium it is impossible in aqueous solution to effect an oxidation to the VI state. Similarly, it becomes increasingly difficult to effect oxidation from the III to the IV state in going from uranium to plutonium, and with americium the evidence indicates that it probably is not possible to effect this oxidation in acid solution at all. If this oxidation should be proved possible the indications are that the potential is so great that the higher oxidation state of americium is reduced rapidly by water and cannot be maintained in aqueous solution for any great length of time. These considerations are illustrated by the following table (Table I) in which the standard oxidation-reduction potentials, referred to the hydrogen-hydrogen ion couple as zero (see, for example, W. M. Latimer, "Oxidation Potentials"(22)), are listed for the III-IV and IV-VI oxidations for these elements(23).

TABLE I

Some Oxidation Potentials of the Actinides

(Aqueous Solution, 1 Molal)

Element	Potential in Volts	
	III-IV	IV-VI
92-U	+0.63	-0.60
93-Np	-0.14	-0.94
94-Pu	-0.95	-1.11
95-Am	<-2	<-2

Much of the work done with curium has of necessity been limited to the tracer scale and therefore it has been impossible to make corresponding quantitative deductions. This work, however, has led to the definite qualitative conclusion that it is impossible in aqueous-acid solution to oxidize curium to the VI state and that it is also impossible to oxidize it from the III to the IV state. In fact, the experiments of S. G. Thompson, L. O. Morgan, R. A. James and I. Perlman(24), in which tracer amounts of curium and americium were subjected to strong oxidation under alkaline fusion, indicate that it is more difficult to oxidize curium from the III to an upper state than is the case for americium, and it may be impossible to effect this oxidation at all. These experiments indicate that americium may be oxidized in alkaline media, and can in this manner be separated from curium. In fact, L. B. Werner and I. Perlman(25) were able to oxidize americium(III) in 40% potassium carbonate solution by the use of the strong oxidizing agent hypochlorite to an insoluble compound, probably of americium(V), and hence effect an efficient separation from curium(III) which apparently is not oxidized under these severe conditions. The microchemical experiments of Werner and Perlman(26) with macroscopic concentrations of curium also point toward the existence of curium solely in the III oxidation state.

This tendency toward increasing stabilization of the lower oxidation states, especially the III state, with increasing atomic number also manifests itself notably in the stability of the solid compounds of the various oxidation states of these elements. The best illustration arises from a consideration of the solid non-oxygenated halides of these elements. The first possibility of the production of a trifluoride appears with uranium trifluoride which can be prepared only under drastic reducing conditions and the stability and ease of reduction to the trifluoride increases in going to neptunium and then to plutonium. Moreover, while it is relatively easy to produce uranium hexafluoride, it is more difficult to produce neptunium hexafluoride and very difficult or impossible to produce and to keep the plutonium hexafluoride. In the case of americium it has not been possible to produce any higher fluoride than the trifluoride.

With respect to the other halides, it has, in fact, been impossible to prepare any plutonium or americium chloride, bromide or iodide of oxidation state higher than III, and it has been possible to prepare only the chloride and bromide of neptunium of oxidation state IV (in addition to the chloride, bromide and iodide of oxidation state III). In the case of uranium it has been known for some time that there are chlorides of oxidation state higher than IV and a chloride, bromide and iodide of oxidation state IV. These considerations are well illustrated in the following table (Table II) which lists all of the halides of uranium, neptunium, plutonium, and americium which have been prepared and maintained as stable in the solid state. So far the chemical evidence indicates that it will be difficult, probably impossible, to prepare any of the non-oxygenated halides of americium (and curium) of oxidation state higher than III. In fact, S. Fried and A. E. Florin(27) have treated AmF_3 with fluorine at elevated temperatures and have obtained no evidence for the formation of a higher fluoride.

This chemical evidence indicates that the 5f electrons are less tightly bound than the 4f electrons and therefore can be more easily removed by oxidation, as should be expected on the basis of the predicted lower binding energy of 5f as compared to 4f electrons. Thus the III state of thorium cannot exist in aqueous solution, and until the present the IV and III states of protactinium have not been observed in aqueous solution, although it seems reasonable that with further study one or both of these states will be observed. In the case of solid compounds it seems not impossible that tripositive thorium compounds with one or more of the heavier halogens will be prepared under rather severe reducing conditions, and that tetra- and tripositive compounds of protactinium will be prepared as soon as efforts in this direction are made. (In fact, Zachariasen(28) and J. D. McCullough(29) already have some crystallographic evidence for a dioxide of

TABLE II

Halides of Some of the Heaviest Elements

	Fluorides				Chlorides				Bromides		Iodides	
92-U	UF_6,	UF_5	UF_4,	UF_3	UCl_6,	UCl_5,	UCl_4,	UCl_3	UBr_4,	UBr_3	UI_4,	UI_3
93-Np	NpF_6,	NpF_5(?),	NpF_4,	NpF_3			$NpCl_4$,	$NpCl_3$	$NpBr_4$,	$NpBr_3$		NpI_3
94-Pu	PuF_6(?),		PuF_4,	PuF_3				$PuCl_3$		$PuBr_3$		PuI_3
95-Am				AmF_3				$AmCl_3$		$AmBr_3$		AmI_3

protactinium with the fluorite structure although this might be a case of solid solution of two oxidation states similar to the solutions studied by J. K. Marsh(30, 45) in the PrO_2-Gd_2O_3, TbO_2-Nd_2O_3 and Pr_2O_3-PrO_2 systems and similar to the well known solid solutions in the iron oxide systems.) The characteristic IV oxidation state demonstrated by thorium is then analogous to the IV oxidation state of cerium and the V state of protactinium is analogous to the somewhat uncertain V state of praseodymium(30,31). From the behavior of uranium, neptunium and plutonium, it must be deduced that as many as three of the 5f electrons are given up fairly readily. In this connection it is interesting to note that in the case of the lanthanide elements there are in general only two electrons (beyond the xenon structure) outside of the 4f shell (see Table VI below), although the persistent oxidation state is certainly the III state.

Americium should possess an oxidation state of II which it would attain through the presence of seven electrons in the 5f shell in a manner quite analogous to the II state of europium, the element immediately preceding gadolinium, with its seven 4f electrons. Because of the greater ease in the removal of the 5f electrons it should require a considerably stronger reducing agent to reduce americium from the III to the II state than is the case for europium. It is not impossible, however, but in fact probable, that it will be found that americium can be reduced and maintained in the II state in aqueous solution. Thompson and co-workers(24) have made partial separations of americium (presumably as Am(II)) from curium in tracer amounts in aqueous solution by using sodium amalgam as the reducing agent and carrying americium selectively with samarium(II) sulfate, and in addition by using barium as reducing agent and carrying americium selectively with barium chloride from concentrated hydrochloric acid solution. It is interesting to note as well that AmO has been prepared(32) by treating americium oxide with hydrogen at 800°C (although some impurity may have been responsible for the reduction here). Curium with its seven 5f electrons should exhibit the III state exclusively and all of the evidence, obtained both from tracer and macroscopic quantities, indicates that this is the case.

The following table (Table III) summarizes the known oxidation states of the lanthanide and actinide elements in such a way as to bring out the analogy between the two groups and to show the greater ease of oxidation for the members of the latter group. The uncertain states are designated with parentheses.

TABLE III

Oxidation States of Lanthanide and Actinide Elements

At. No.	57	58	59	60	61	62	63	64	65
Element	La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb
Oxid. States						2	2		
	3	3	3	3	3	3	3	3	3
		4	4	(4)					4
			5						
At. No.	89	90	91	92	93	94	95	96	
Element	Ac	Th	Pa	U	Np	Pu	Am	Cm	
Oxid. States							2		
	3			3	3	3	3	3	
		4	(4)	4	4	4			
			5	5	5	5	(5)		
				6	6	6			

The metals of the elements thorium to americium inclusive have been prepared and their properties studied and these bear a striking resemblance to the metals of the rare earth elements. All of them are highly electropositive, and to about the same degree, which is a similarity to the rare earth metals and a difference from the corresponding 5d elements, hafnium (element 72) to iridium (element 77) inclusive, in which the electropositive character of hafnium is lost as 5d electrons are added in going toward iridium. Another remarkable resemblance to the rare earth elements lies in the densities of the metals. Both americium(33) and the analogous 4f element, europium(34), have densities much lower than their neighboring elements. Thus these metals seem to have radium-like or barium-like structures, with abnormally high radii and analogous electronic structures. This analogy in the densities is shown in Table IV where the densities (of the forms of the metals which are stable at room temperature) are listed(34,35).

TABLE IV

Densities of Room Temperature Forms of Some Lanthanide and Actinide Metals

At. No.	60	61	62	63	64
Element	Nd	Pm	Sm	Eu	Gd
Density	7.00		6.93	5.24	7.95
At. No.	92	93	94	95	96
Element	U	Np	Pu	Am	Cm
Density	19.0	19.7	19.8	~11	

A comparison with tungsten, rhenium, osmium and iridium brings forth no such analogy.

C. Absorption Spectra in Aqueous Solution and Crystals. One of the characteristic properties of the elements of the lanthanide series, a property which depends upon the 4f electrons, is their sharp absorption bands, to a large extent in the visible spectrum. This absorption is due to transitions involving the 4f electrons and the sharpness is a consequence of the shielding of these, both in the ground and excited states, by electrons in the outer shells. The investigations of this type with the elements uranium, neptunium, plutonium, americium and curium have shown a striking similarity in this property to the rare earths, which is further evidence that we are dealing with 5f electrons. The analogy between the rare earths and uranium(IV) in this property was noticed by Goldschmidt(36) and between the rare earths and uranium(III) and (IV) by F. Ephraim and M. Mezener(37) many years ago. This similarity between the actinide and the lanthanide elements is more than qualitative in that the general complexity of the absorption picture undergoes analogous simplification as we approach the middle of the two series, that is, as we approach the elements gadolinium and curium, with their seven 4f or 5f electrons. Unfortunately, up to the present the most extensive work on the absorption of uranium and the transuranium elements has been done in solution, where much of the sharpness is lost, and with instruments of not very high resolving power. The aqueous solution absorption spectra of the tripositive actinides, taken from various sources on the Plutonium Project(38) are shown in Fig. 1, where the corresponding spectra of the rare earth elements also are shown for purposes of comparison. Although the absorption curves of W. Prandtl and K. Scheiner(39) are available for comparison, this work was not done under conditions comparable to those under which the work on the actinides was done. Therefore, D. C. Stewart(40) has measured the absorption spectra of the rare earth elements under comparable conditions and his results are also given in Fig. 1. The preliminary results of P. M. Lantz and G. W. Parker(41) on the absorption spectrum of promethium (element 61) are also included. It appears that as we approach the middle of the two series (i.e., as we approach the elements gadolinium and curium) the ground states involving the seven f electrons lie very definitely below the next higher states, leading to energy differences of a sufficient magnitude to cause the main absorption to fall outside of the visible in the ultraviolet region.

The best method for the comparison of the absorption spectra of the two groups of elements is to compare the spectra obtained with crystals, where the absorption lines are known to be very sharp for the rare earth elements on the basis of a large number of measurements with many of these elements. Such measurements are underway for a number of transuranium elements and the results so far indicate striking analogies. S. Freed and F. Leitz(42) have measured the absorption spectrum of solid americium trichloride and they find sharp lines, of the order of one to five Angstroms wide, which is a width comparable with the sharpest rare earth spectra. In fact the sharpness in the spectra(42) of americium chloride and americium bromide is so extreme, at room temperature and at 77°K., that only the tripositive europium ion is comparable; since the absorption spectrum of the latter originates from a ground state involving six 4f electrons, it seems very likely that the basic state of tripositive americium contains six 5f electrons. They(42) also measured the absorption spectra at room temperature and at 77°K of uranium tetrachloride, neptunium tetrachloride and plutonium trichloride and the sharpness of the lines indicates that the least stable electrons of these ions are in the inner 5f shell in the activated as well as in the basic electronic states.

D. Crystallographic Data. As mentioned above, crystallographic evidence in addition points to the filling of the 5f shell in this neighborhood of heavy elements. Some years ago, Goldschmidt(14, 43) had already noticed the isomorphism of ThO_2 and UO_2, and the decrease in size in going to UO_2, and had interpreted this to indicate the presence of 5f electrons in uranium. The observation of Zachariasen(28) of the isomorphism of the compounds ThO_2, PaO_2, UO_2, NpO_2, PuO_2 and AmO_2(44), together with his observation of the regular decrease in radius of the metallic ion in these oxides, has been interpreted by him to be excellent evidence that the electrons are going into the 5f shell. (In this series, however, the lattice constants of the PaO_2(28,29) and AmO_2(45) do not fit perfectly into the regular pattern for decrease in atomic radius of the metallic ions; this might be due to mixed oxidation states similar to the situation found for the higher praseodymium oxide by Marsh(46).) Both of these investigators have advanced the hypothesis that it is a "thoride" series, i.e., that the first 5f electron appears in the first element beyond thorium, namely protactinium.

Zachariasen has used the x-ray diffraction method to determine the molecular structures of a great number of compounds of thorium, uranium and the transuranium elements. All of these measurements point toward the filling of the 5f shell since analogous compounds are found to be isomorphous, indicating that the successive electrons are added in such a way (i.e., to an inner shell) as to allow the analogous compounds of successive elements to have identical molecular structures. Thus, Zachariasen(47) finds that practically all of the various halide types shown in Table II above have isomorphous structures: for example, all members of the group ThF_4-UF_4-NpF_4-PuF_4 are of identical structure types, all members of the group UF_3-NpF_3-PuF_3-AmF_3 are also isomorphous with each other, and the same is true for the group UCl_3-$NpCl_3$-$PuCl_3$-$AmCl_3$ etc. To be sure, in some cases (for example, UBr_3-$NpBr_3$-$PuBr_3$-$AmBr_3$), there is a change in structure type in proceeding up the group, but this is to be expected on the basis of the contraction which takes place, and is entirely consistent with the addition of the successive electrons to the 5f shell. Zachariasen(47) has used these structure data to calculate ionic radii and these radii show a progressive decrease in size with increasing atomic number in a manner quite analogous to the well known "lanthanide contraction" observed with the rare earth elements, the compounds of which are in turn isomorphous with the corresponding compounds of the actinide elements. In order to further illustrate these considerations the following table (Table V) gives the ionic radii of a number of the actinide and lanthanide elements(47).

TABLE V

Ionic Radii of Actinide and Lanthanide Elements

No. of 4f or 5f Electrons	Actinide Series				Lanthanide Series	
	III State		IV State			
0	Ac^{+3}	1.11A	Th^{+4}	0.95 A	La^{+3}	1.04 A
1	(Th^{+3})	(1.08)	Pa^{+4}	(0.91)	Ce^{+3}	1.02
2	(Pa^{+3})	(1.06)	U^{+4}	0.89	Pr^{+3}	1.00
3	U^{+3}	1.04	Np^{+4}	0.88	Nd^{+3}	0.99
4	Np^{+3}	1.02	Pu^{+4}	0.86	61^{+3}	(0.98)
5	Pu^{+3}	1.01	Am^{+4}	(0.85)	Sm^{+3}	0.97
6	Am^{+3}	1.01			Eu^{+3}	0.97

E. Magnetic Susceptibility Data. Magnetic susceptibility measurements on compounds of the heaviest elements ideally should lead to the resultant magnetic moments in fundamental units and in this way give information as to the quantum states of the responsible electrons. Actually, as evidenced by the rare earth elements, the situation is rather complex and the exact behavior expected for the heaviest elements, on the basis of the presence of either 5f or 6d electrons, cannot be, or at least has not been, predicted. Nevertheless, such measurements should give, and indeed have given, information on this point.

The earliest magnetic susceptibility measurements that were made on compounds of uranium(48, 49, 50, 51) and plutonium(52) showed that these are paramagnetic, yet the results are difficult to interpret quantitatively. A simple qualitative explanation of the magnetic susceptibilities of Pu(III), Pu(IV) and Pu(VI) lies in the assumption that there are five 5f electrons in Pu(III) which are successively removed as one goes to the higher oxidation states. These measurements, however, do not lead to this interpretation as the sole and unambiguous one, and as a result must be regarded only as being consistent with and lending weight to this view, but not giving proof of it.

Later C. A. Hutchison and N. Elliott made magnetic susceptibility measurements over a wide range of temperatures on a number of compounds of the heaviest elements. In the case of uranium(IV) compounds(53,54) they found that a number behave in a manner similar to praseodymium(III) compounds, indicating that these two groups of compounds have isoelectronic structures with respect to f electrons and thus have two such electrons. They disclosed in addition that the temperature dependence of the magnetic susceptibility of these uranium(IV) compounds obeys the Curie-Weiss law over a range of temperatures, and through extrapolation with the use of this law they deduced a resultant magnetic moment very close to that expected for two f electrons. They also concluded that the crystal fields produce more pronounced perturbing effects in this case than in the corresponding case involving 4f electrons. Their measurements(54) on neptunium(V), which is isoelectronic with uranium(IV), also indicated the presence of two f electrons here.

J. J. Howland and M. Calvin(55) have measured the magnetic susceptibilities of the cations of uranium, neptunium, plutonium, and americium in most of their stable oxidation states. In order to account consistently for the observed values of the magnetic susceptibility, the central atom must be assumed to have electronic configurations (beyond the radon structure) of the type

$(5f)^{1-6}$: for example, Np(VI) corresponds to the structure 5f, U(IV), Np(V) and Pu(VI) to $5f^2$, Np(IV) to $5f^3$, Pu(IV) to $5f^4$, Pu(III) to $5f^5$ and Am(III) to $5f^6$. The experimental effective magnetic moments are in the examples of two, three, and four electrons lower than would be expected for the lowest electronic states predicted by Hund's rules for L-S coupling; because the spin-orbital interaction is very strong in heavy atoms, other states which belong to the lowest group in j-j coupling can lie deep enough to be significantly populated at room temperature. For the configurations $5f^1$, $5f^5$, $5f^6$, and $5f^7$, however, no such complication exists; either atomic coupling scheme leads to only one, the same, low lying state.

F. Spectroscopic Data. Spectroscopic evidence also lends support to the actinide interpretation. C. C. Kiess, C. J. Humphreys and D. D. Laun(56,57) investigated the spectrum of uranium atoms and they gave the interpretation that the electron configuration of the lowest state of neutral uranium is $5f^3 6d 7s^2$ (beyond radon), a configuration which fits in very well since uranium is the third element in the series. Other work by P. Schuurmans and co-workers(58,59) on the spectrum of gaseous U(II) and U(I) has given results which are consistent with this structure for the neutral uranium atom. Spectroscopic evidence that thorium possesses a ground state analogous to that of cerium also indicates that the characteristic configuration for uranium could involve three 5f electrons. Other observations(60,61,62) made on the gaseous Th(II) spectrum indicate that the 5f is very close to the 6d electron in binding energy in this atom.

H. Russell(63) has made a complete analysis of the x-ray data for radium, thorium and uranium and has concluded that the 5f lies lower than the 6d level and that the 5f shell begins to fill at thorium.

F. S. Tompkins and M. Fred(64) have made a qualitative comparison of the emission spectra of the actinide and the lanthanide elements. They found such a strong analogy between the average intensity of the lines in the case of americium and europium that it seems safe to conclude that these have similar electronic structures in the ground states. This then indicates the configuration $5f^7 7s^2$ for the gaseous atom of americium.

III. Correlations and Deductions

A. Electronic Configurations. The following table (Table VI) gives what appears to be the configuration or the best prediction for the configuration (beyond the radon structure) of the ground state of the neutral gaseous atom for each of the elements actinium to curium inclusive. The trend in the chemical properties, with its implication that the 5f becomes progressively of lower energy compared to the 6d level as the atomic number increases, is used as an aid in making the predictions.

The configurations(65,66) (beyond xenon) of the corresponding neutral rare earth elements are given for comparison. The ground states given for cerium and praseodymium are those predicted(65,66) on the basis of the recently determined ground states of the singly ionized atoms(67), and that of element 61 is obtained by interpolation, and as a consequence these are subject to some doubt. The ground states given for neodymium(58), samarium, europium and gadolinium are those spectroscopically determined(67) and should be considered as well established.

TABLE VI

Suggested Electron Configurations (Beyond Radon and Xenon) for Gaseous Atoms of Actinide and Lanthanide Elements

89-Ac	$6d7s^2$	57-La	$5d6s^2$
90-Th	$5f6d7s^2$ or $6d^27s^2$	58-Ce	$4f^26s^2$
91-Pa	$5f^26d7s^2$ or $5f6d7s^2$	59-Pr	$4f^36s^2$
92-U	$5f^36d7s^2$	60-Nd	$4f^46s^2$
93-Np	$5f^46d7s^2$ or $5f^57s^2$	61	$4f^56s^2$
94-Pu	$5f^56d7s^2$ or $5f^67s^2$	62-Sm	$4f^66s^2$
95-Am	$5f^77s^2$	63-Eu	$4f^76s^2$
96-Cm	$5f^76d7s^2$	64-Gd	$4f^75d6s^2$

It should be pointed out that it would be entirely consistent from the point of view that we are dealing here with a series of actinide elements if it should eventually be found that there are no 5f electrons present in thorium (or protactinium). It is quite possible on the basis of present evidence that protactinium, or even uranium, might be the first to have 5f electrons. It seems quite likely, however, that electrons will be placed in the 5f shell earlier in the series than uranium and that protactinium will have at least one. An essential point is that curium definitely seems to have seven 5f electrons and element 103 probably would have fourteen 5f electrons.

In the case of some of the elements in the series it may be difficult to assign electrons to the 5f or 6d shells since the energy necessary for the shift from one shell to the other may be within the range of chemical binding energies. The electron configuration may differ from compound to compound or even with the physical state of a given compound. Moreover one certainly cannot be sure that the configuration of the gaseous atom, for example, will correspond to that of the compounds or of the hydrated ions in solution. In the case of the lanthanides, in fact, the configuration of the gaseous atom has in general only two electrons (beyond the xenon structure) outside of the 4f shell, although the predominant oxidation state is certainly the III state. Since the energy difference between such far outlying levels as the 5f and 6d shells is rather small and since resonance effects should be rather large, these may predominate in determining that a composite energy level lies lowest. Thus some of these elements could possibly constitute what might more properly be called a 5f-6d range in this series, rather than a part of a totally 5f transition group.

The evidence that has accumulated so far seems nevertheless to point to lower energies for the 5f compared to the 6d levels for the compounds of the element, as early as uranium in this series. It is in the case of the elements thorium and protactinium where the relative energy positions of these levels is as yet most uncertain. As in the other transition series, the relative energy level of the shell which is undergoing the filling process becomes lower as the successive electrons are added, and by the time americium and curium, and presumably the subsequent members of the series, are reached the 5f seems clearly to be definitely of lower energy than the 6d shell. It is not possible to place the electrons in neptunium and plutonium with confidence and hence in Table V the alternative structures $5f^5 7s^2$ and $5f^6 7s^2$, respectively, for gaseous neptunium and plutonium are suggested in recognition of the possibility that already with these elements the 6d shell is not occupied.

B. Possible Deductions without Data on Transuranium Elements. Although it is the information on the transuranium elements that has been decisive in enabling us to come to the present view concerning the electronic structure, or more properly speaking, the position in the periodic table of the heaviest elements, it is interesting to conjecture, in retrospect, about the possibility of having arrived at a similar conclusion without this information. Actually there has been much information about actinium, thorium, protactinium and uranium, especially about the latter, which pointed in this direction. As mentioned above, there is the similarity among the metals of these elements with respect to electropositive character. In addition, the melting point of uranium metal seems to relate it more to the immediately preceding elements than to tungsten and molybdenum. The analogy of uranium to neodymium with respect to light absorption by the tripositive ions and the spectroscopic evidence for a ground state of the gaseous uranium atom involving three 5f electrons has already been mentioned.

Uranium differs considerably from tungsten (and molybdenum) in the chemistry of the lower oxidation states and, in fact, uranium(III) has great similarity to the tripositive rare earth elements and actinium, while uranium(IV) resembles thorium and cerium(IV). Thus uranium(III) and uranium(IV) are not acidic in character, do not tend to form strong complex ions in solution, have fluorides which are insoluble and isomorphous with the fluorides of the rare earth elements and have other halides with crystal structures which are in general isomorphous with the corresponding rare earth halides. On the other hand, tungsten(III) and tungsten(IV) exist in aqueous solution predominantly as strong complex ions; for example, tungsten(III) has a strong chloride complex ion and tungsten(IV) forms strong fluoride and cyanide complex ions. In this connection Thompson(68) has pointed out that tungsten(IV) forms the very stable complex ion $W(CN)_8^{\equiv}$, with the stable configuration of eighteen outer electrons, while uranium(IV) possesses no significant tendency to form an analogous complex cyanide ion as would surely be expected if uranium possessed the same outer electronic structure as tungsten.

Although molybdenum dioxide and tungsten dioxide have isomorphous crystal structures, tungsten dioxide and uranium dioxide do not, while uranium dioxide, thorium dioxide and cerium dioxide all do possess isomorphous structures. It is interesting to note that although uranium is not associated with tungsten in minerals, uranium and thorium minerals practically always have the rare earth elements associated with them and the rare earth minerals practically always contain uranium or thorium.

Arguments on the basis of the scanty evidence from the chemical properties of thorium and uranium alone have been given by others, including Villar(13) and more recently D. F. Stedman(69), for a 5f type transition series in the heaviest elements, beginning with thorium.

C. <u>Position in Periodic Table and Nomenclature</u>. A method of presenting the actinide elements in the periodic table is shown in Fig. 2. Here are shown the fourteen elements of atomic numbers 90 to 103 inclusive, with actinium (element 89) as the prototype listed as a series below, and in a manner similar to, the common listing of the fourteen rare earth elements of atomic numbers 58 to 71 inclusive, for which lanthanum (element 57) is the prototype. It is not proposed that this particular form of the periodic table has any more merit than any of a number of others which place these elements in positions homologous to the rare earth elements, since it is obvious that they can be analogously placed in a number of other types of tables or charts. The elements 90 to 96 inclusive could be listed separately below the 5d elements in recognition of the resemblance of the first few of these to 5d elements. This appears to be undesirable however since the last members of this group bear no such resemblance and it is probably impossible to draw a line as to just where the resemblance ends.

As mentioned above, the important point is the presence of seven 5f electrons in stable, tripositive curium (element 96), making this element very actinium-like. A series of " thoride" elements, for example, would imply stable IV oxidation states in elements 95 and 96, with the presence of seven 5f electrons and the IV state exclusively in element 97. A series of this type seems to be ruled out by the now known impossibility of having americium in solution in the IV state and by the apparent non-existence of any oxidation state other than III in curium. Moreover, the III state of uranium would be surprising on this basis since this element would be the <u>second</u> member of a " thoride" or

" IV oxidation state" series.

The group probably could have been just as well described by some other term such as " curide series", rather than " actinide", which is derived from analogy with the term lanthanide. Another possibility would be to use a name analogous to " rare earths". A suggestion here would be " synthetic earths" in view of the synthetic source of all except the first three members. (Even among these, the synthetic production of Pa^{231} according to $Th^{230}(n,\gamma)Th^{231} \longrightarrow Pa^{231}$ and of U^{233} from $Th^{232}(n,\gamma)Th^{233} \longrightarrow Pa^{233} \longrightarrow U^{233}$, accounts for the best sources of important isotopes of protactinium and uranium; the best source of actinium also comes from pile neutrons, by the reactions $Ra^{226}(n,\gamma)Ra^{227} \longrightarrow Ac^{227}$.)

Irrespective of the name which usage will finally assign to this group of elements, however, it seems that the outstanding characteristics of the group, namely the " eka-gadolinium" character of curium (and the presumed " eka-lutecium" character of element 103), together with the regularly increasing trend toward actinium-like character in going from thorium to curium, are best represented by listing these elements in corresponding positions under the rare earth elements. (Some spatial classifications (70,71) of the elements have appeared recently in which these elements, starting with thorium as the homologue of cerium, are listed as the chemical homologues of the rare earth elements, but the reason in these cases appears to be mainly connected with the symmetry of, and the ease of making, such an arrangement.)

D. <u>Predicted Properties of Transcurium Elements</u>. It is interesting to speculate about the chemical properties of the undiscovered elements beyond curium (atomic number 96). The immediately following seven elements, that is, elements 97 to 103 inclusive, should constitute the second half of this rare earth-like transition group. It appears likely that the added electrons in proceeding up this series will be placed in a 5f shell of definitely lower energy than the 6d shell. Element 97 will probably have a IV as well as a III oxidation state and in view of the lower binding energy of the 5f as compared to the 4f electrons it should be somewhat easier to oxidize element 97 (eka-terbium) to this IV state than is the case for terbium. Correspondingly, it might be somewhat easier to oxidize element 98 (eka-dysprosium) to the IV and V oxidation states than is the case for dysprosium, for which, in fact, oxidation above the III state is practically impossible. Toward the end of the series, elements 102 and 101 should be capable of being reduced to the II oxidation state, analogously to ytterbium and thulium, while element 103 should be analogous to lutecium with respect to the complete stability of the III state.

Element 104 should continue with the filling of the 6d shell and be a true eka-hafnium. After the filling of the 6d shell in the following elements there might be addition to the 7p shell with the attainment of the rare gas structure at element 118.

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ABSORPTION SPECTRA of the AQUEOUS TRIPOSITIVE

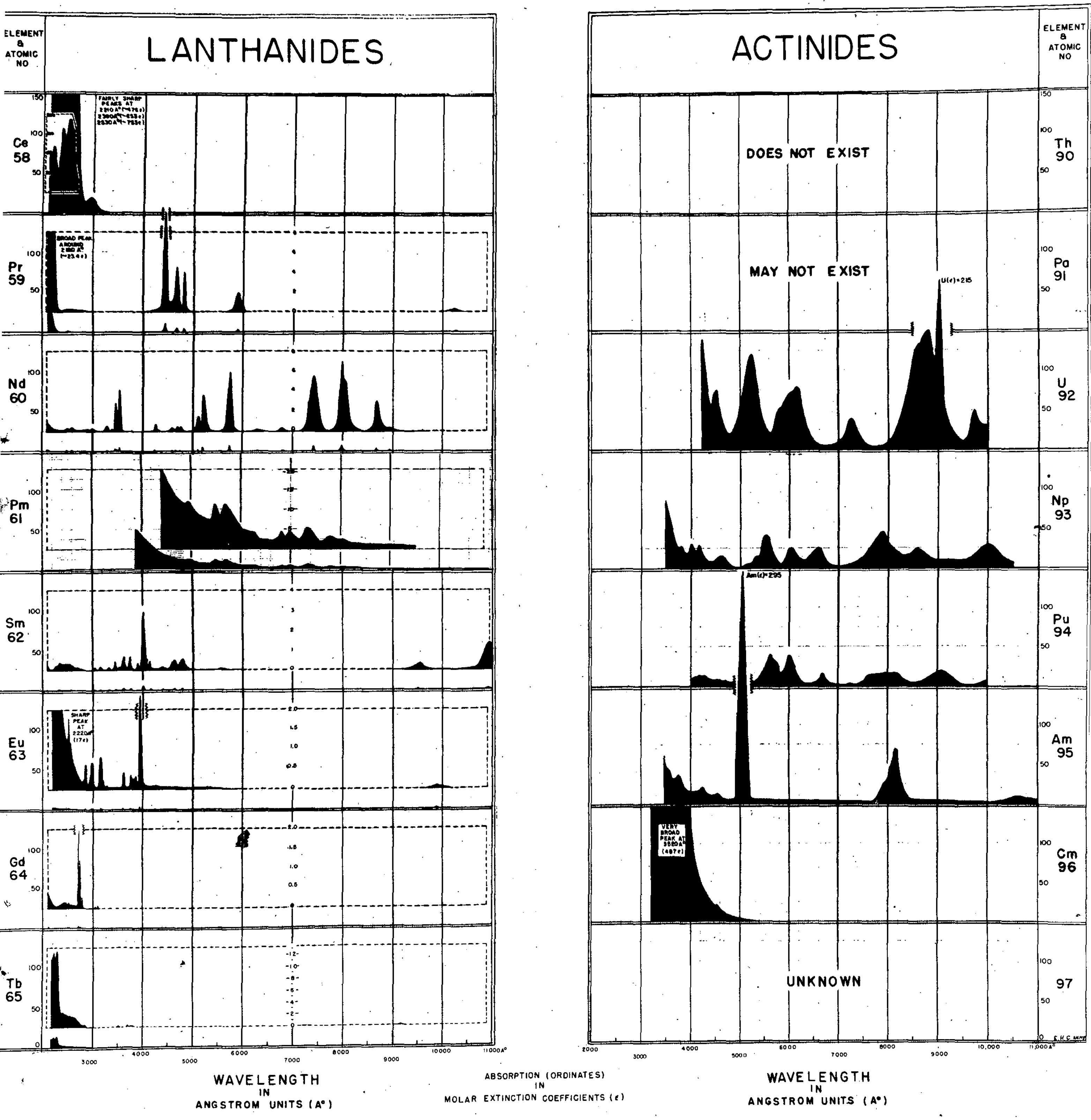

FIG. 1

PERIODIC TABLE SHOWING HEAVY ELEMENTS AS MEMBERS OF AN ACTINIDE SERIES

1 H 1.008																1 H 1.008	2 He 4.003
3 Li 6.940	4 Be 9.02											5 B 10.82	6 C 12.010	7 N 14.008	8 O 16.000	9 F 19.00	10 Ne 20.183
11 Na 22.997	12 Mg 24.32	13 Al 26.97										13 Al 26.97	14 Si 28.06	15 P 30.98	16 S 32.06	17 Cl 35.457	18 A 39.944
19 K 39.096	20 Ca 40.08	21 Sc 45.10	22 Ti 47.90	23 V 50.95	24 Cr 52.01	25 Mn 54.93	26 Fe 55.85	27 Co 58.94	28 Ni 58.69	29 Cu 63.57	30 Zn 65.38	31 Ga 69.72	32 Ge 72.60	33 As 74.91	34 Se 78.96	35 Br 79.916	36 Kr 83.7
37 Rb 85.48	38 Sr 87.63	39 Y 88.92	40 Zr 91.22	41 Cb 92.91	42 Mo 95.95	43 Tc	44 Ru 101.7	45 Rh 102.91	46 Pd 106.7	47 Ag 107.880	48 Cd 112.41	49 In 114.76	50 Sn 118.70	51 Sb 121.76	52 Te 127.61	53 I 126.92	54 Xe 131.3
55 Cs 132.91	56 Ba 137.36	57 La 138.92 58-71 SEE La SERIES	72 Hf 178.6	73 Ta 180.88	74 W 183.92	75 Re 186.31	76 Os 190.2	77 Ir 193.1	78 Pt 195.23	79 Au 197.2	80 Hg 200.61	81 Tl 204.39	82 Pb 207.21	83 Bi 209.00	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 Ac SEE Ac SERIES															

LANTHANIDE SERIES	57 La 138.92	58 Ce 140.13	59 Pr 140.92	60 Nd 144.27	61 Pm	62 Sm 150.43	63 Eu 152.0	64 Gd 156.9	65 Tb 159.2	66 Dy 162.46	67 Ho 163.5	68 Er 167.2	69 Tm 169.4	70 Yb 173.04	71 Lu 174.99
ACTINIDE SERIES	89 Ac	90 Th 232.12	91 Pa 231	92 U 238.07	93 Np 237	94 Pu	95 Am	96 Cm							

FIG. 2