

X-Ray Absorption Coefficients of Thorium, Uranium, and Plutonium— Experimental Determination and Theoretical Interpretation*

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The x-ray absorption coefficients of thorium, uranium, and plutonium have been determined for the $K\alpha$ and $K\beta$ radiations of silver, molybdenum, copper, cobalt, iron, and chromium. Four absorption edges were found in the wavelength region covered. These were identified as the L_I , L_{II} , L_{III} , M_I , edges and a M_I , M_{II} complex for plutonium. There is considerable disagreement between the experimental coefficients and the theoretical coefficients obtained from the *Internationale Tabellen*. Reasons for this discrepancy are discussed briefly. A correlation is obtained between the absorption of x-ray energy and the number of electrons in the 5f shell.

Through a consideration of the atomic number, a screening constant, principal quantum numbers, and the atomic energy levels for thorium, uranium, and plutonium, the x-ray absorption coefficients of these elements have been calculated. Screening constants and quantum conditions in the various equations were adjusted until, on the average, the experimental and calculated values agree within 2%. An increase in the screening constant of plutonium over that for thorium and uranium is interpreted in terms of the number of electrons occurring in the 5f shell. The change in certain quantum conditions for plutonium in comparison to those for thorium and uranium is interpreted in terms of plutonium being a subtransitional point within the 5f transition series.

INTRODUCTION

AS Victoreen¹ has noted, comparatively little work has been done on the absorption of x-radiation by atoms. Such available data are scattered widely in the literature, and disagreement between observers is sufficiently great to make any single value doubtful. Thus, Victoreen concludes that there is great practical need for reasonably accurate values of the x-ray absorption coefficients of the elements.

This paper deals with the experimental determination and the theoretical interpretation of the x-ray absorption coefficients of thorium, uranium, and plutonium. Consideration is given to the discrepancy between the experimentally determined and the calculated value of the coefficient of water for Mo $K\alpha$ radiation.

EXPERIMENTAL DETERMINATION

Experimental Method

Characteristic x-rays, generated in an x-ray tube, were collimated, reflected at the proper Bragg angle from a freshly cleaved surface of LiF, and then passed through a $\frac{1}{32}$ -inch diameter hole drilled in two parallel steel plates. Wavelengths consisting of $\lambda/2$ were avoided by operating the x-ray tube at a sufficiently low voltage so that they were not generated. The x-rays, after passing through the hole in the steel plates, were detected by a scintillation counter which was coupled with a pulse-height analyzer. The absorption cells consisted of sealed bags made of 0.004 inch thick polyvinyl chloride sheet. Solutions of material to be analyzed were introduced into the sealed bags with a hypodermic syringe, the polyvinyl chloride being essentially self-sealing. Enough internal air pressure was

maintained in the absorption cells so that when these were inserted between the parallel steel plates the trapped air acted as a cushion insuring that the two sides of the absorption cell remained parallel. Use of a solution eliminated the objection of nonuniformity frequently encountered in using thin foils of elements of high atomic number for absorption work.

Method of Analysis

The reduction in intensity of an x-ray beam in passing through material is given by the formula

$$I = I_0 \exp(-\mu_m \rho t), \quad (1)$$

where I is the measured intensity (after passing through an absorption cell filled with solution and correcting for both radioactive and instrumentation background), I_0 is the original intensity (after passing through a blank absorption cell and correcting for instrumentation background), μ_m is the mass absorption coefficient, ρ is the density of the material, and t is the thickness of the material. For a two-component system this formula can be rearranged as

$$\ln(I_0/I) = \mu_1 \rho t w_1 + \mu_2 \rho t w_2, \quad (2)$$

where w_1 is the weight fraction of the component having the mass absorption coefficient μ_1 , and w_2 is the weight fraction of the component having the mass absorption coefficient μ_2 . Since $w_2 = 1 - w_1$, the formula can be reduced to

$$\ln(I_0/I)/\rho t = (\mu_1 - \mu_2)w_1 + \mu_2. \quad (3)$$

A plot of $\ln(I_0/I)/\rho t$ against w_1 results in a straight line, the intercept of which is equal to μ_2 and the slope of which is equal to $(\mu_1 - \mu_2)$. Thus, if solutions of varying concentrations; i.e., plutonium in 1M HClO₄, are prepared, the x-ray intensities measured, and the

* Work done under the auspices of the U. S. Atomic Energy Commission.

¹ J. A. Victoreen, J. Appl. Phys. 20, 1141 (1949).

data analyzed according to the above formula (where μ_2 is the mass absorption coefficient of $1M$ $HClO_4$) the mass absorption coefficient, μ_1 , of plutonium, is found by adding the experimental intercept to the experimental slope.

The practical application of the above equation is straightforward. Periodic checks were made to insure that I_0 and I were measured in the linear range of the recording instruments. The weight fraction w_1 , of thorium, uranium, or plutonium was calculated from the concentrations of the solutions under investigation. The densities of the solutions were experimentally determined by weighing known volumes.

The experiments were performed at room temperature of approximately $25^\circ C$. Extreme variations of $\pm 5^\circ C$ were unlikely but if they had occurred the densities would have been affected by only about $\pm 1\%$. The determination of the distance of separation between the steel plates is important and was accomplished experimentally by two different methods. An independent, experimental determination of the mass absorption coefficient of water for $Mo K\alpha$ radiation was found to be $0.985 \text{ cm}^2\text{g}^{-1}$. This value leads to an evaluation of the separation of $0.2208 \pm 0.0041 \text{ cm}$. By using a feeler gauge and a micrometer caliper the value of the

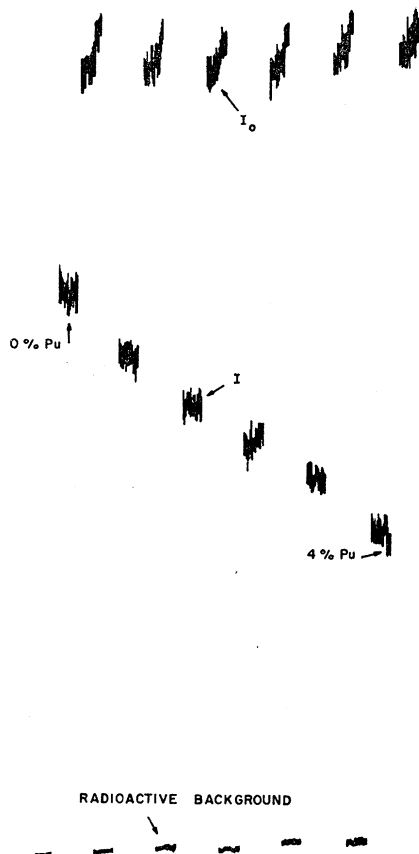


FIG. 1. Recording trace of solutions of plutonium in $1M$ $HClO_4$, $Mo K\alpha$ radiation.

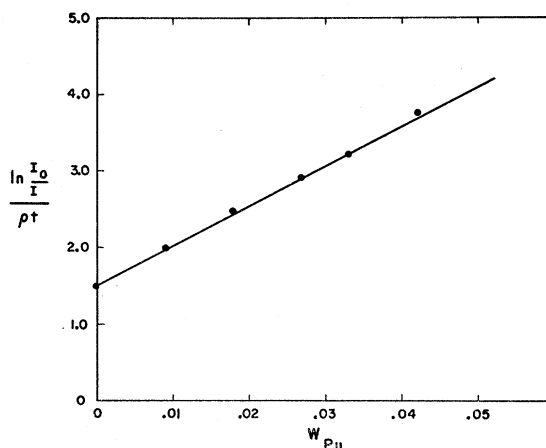


FIG. 2. $\ln(I_0/I)/\rho t$ vs w_{Pu} in $1M$ $HClO_4$ solutions, $Mo K\alpha$ radiation.

separation was found to be $0.2210 \pm 0.0025 \text{ cm}$. The agreement between the two methods was considered to be satisfactory.

It should be noted that any significant variation from the value of $0.985 \text{ cm}^2\text{g}^{-1}$ obtained for water for $Mo K\alpha$ radiation will result in values for the separation of the steel plates that will not be in acceptable agreement with the value determined by purely mechanical means. The absorption coefficient for water for $Mo K\alpha$ radiation as determined from the *Internationale Tabellen*² is $1.381 \text{ cm}^2\text{g}^{-1}$ and that from the *Handbook of Chemistry and Physics*³ is $1.132 \text{ cm}^2\text{g}^{-1}$. Use of either of these values will yield a totally unacceptable result for the evaluation of the separation of the steel plates. Thus, the author concludes that the absorption coefficients for water for $Mo K\alpha$ radiation as given by either the *Internationale Tabellen* or the *Handbook of Chemistry and Physics* are considerably in error. Additional support for this view is obtained from the work of Grodstein.⁴ Grodstein presents absorption coefficients for water corresponding to wavelengths of 0.413, 0.620, 0.827, and 1.240 Å. By interpolating Grodstein's values and applying the recommended limits of error, a value of $1.0 \pm 0.1 \text{ cm}^2\text{g}^{-1}$ is obtained for water for $Mo K\alpha$ radiation, which is in satisfactory agreement with the author's value of $0.985 \text{ cm}^2\text{g}^{-1}$.

EXPERIMENTAL RESULTS

A typical example of the experimental data is given in Fig. 1, which shows the recording trace obtained by examining, for $Mo K\alpha$ radiation, solutions of plutonium in $1M$ $HClO_4$ for which the weight fraction varies from 0.0% to approximately 4.0%. Information obtained from this recording trace is plotted in Fig. 2.

² *Internationale Tabellen zur Bestimmung von Kristallstrukturen* (Gebrüder Brontraeger, Berlin, 1935), Vol. II.

³ *Handbook of Chemistry and Physics* (Chemical Rubber Publishing Company, Cleveland, 1952), thirty-fourth edition.

⁴ G. W. Grodstein, *X-ray Attenuation Coefficients from 10 kev to 100 Mev*, National Bureau of Standards Circular No. 583 (U. S. Government Printing Office, Washington, D. C., 1957).

TABLE I. Comparison between experimental and calculated x-ray absorption coefficients for thorium, uranium, and plutonium, cm^2g^{-1} .

Radiation	Wavelength, Å	Thorium		Uranium		Plutonium	
		Experimental	Calculated	Experimental	Calculated	Experimental	Calculated
Ag $K\beta$	0.4970	75±1	73	85±2	84	77±2	77
Ag $K\alpha$	0.5609	99±2	100	113±3	113	70±1	70
Mo $K\beta$	0.6322	88±1	88	96±2	95	94±2	94
Mo $K\alpha$	0.7107	110±1	110	125±4	128	53±1	54
Cu $K\beta$	1.3922	314±7	323	315±5	319	395±11	367
Cu $K\alpha$	1.5418	401±8	415	396±13	410	513±19	488
Co $K\beta$	1.6207	444±8	468	441±10	462	574±19	562
Co $K\alpha$	1.7902	562±6	586	568±7	579	737±28	739
Fe $K\beta$	1.7565	554±8	563	532±3	557	698±36	704
Fe $K\alpha$	1.9373	691±20	699	698±19	691	901±21	921
Cr $K\beta$	2.0848	812±10	805	854±11	800	1096±27	1112
Cr $K\alpha$	2.2909	1051±17	958	894±24	885	1240±22	1243

A value of $1.5 \text{ cm}^2\text{g}^{-1}$ for the intercept and $51.5 \pm 1.0 \text{ cm}^2\text{g}^{-1}$ for the slope may be calculated by passing a least squares straight line through the points. The absorption coefficient for plutonium for Mo $K\alpha$ radiation, $53 \pm 1 \text{ cm}^2\text{g}^{-1}$, is found by adding the intercept and the slope. This value is in good agreement with the value of $53 \pm 3 \text{ cm}^2\text{g}^{-1}$, reported by Williamson, Poole, and Marples.⁵ The absorption coefficient obtained for other elements and other wavelengths is given in Table I and is plotted graphically in Fig. 3.

Experimental Conclusions

Figure 4 shows the variation of the L and M absorption edges as a function of atomic number Z , taken from the work of Richtmyer and Kennard.⁶ Figure 4 predicts that for thorium the L_I absorption edge lies between Ag $K\alpha$ and Mo $K\beta$, the L_{II} edge lies between Mo $K\beta$ and Mo $K\alpha$, and the L_{III} edge lies on the long-

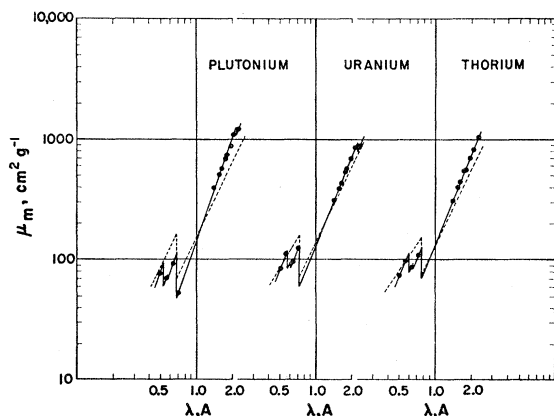


FIG. 3. Experimental x-ray mass absorption coefficients vs wavelength for thorium, uranium, and plutonium. The dotted line indicates the mass absorption coefficients of thorium and uranium and the estimated mass absorption coefficient of plutonium, as given in the *Internationale Tabellen*.

⁵ Williamson, Poole, and Marples, *J. Inst. Metals* **85**, 431 (1956-1957).

⁶ F. K. Richtmyer, and E. H. Kennard, *Introduction to Modern Physics* (McGraw-Hill Book Company, Inc., New York, 1947), fourth edition, p. 485.

wavelength side of Mo $K\alpha$; for uranium the L_I and L_{II} edges lie between Ag $K\alpha$ and Mo $K\beta$, the L_{III} edge lies just to the long-wavelength side of Mo $K\alpha$, and the M_I edge lies between Cr $K\beta$ and Cr $K\alpha$; for plutonium the L_I edge lies between Ag $K\beta$ and Ag $K\alpha$, the L_{II} edge lies between Ag $K\alpha$ and Mo $K\beta$, the L_{III} edge lies between Mo $K\beta$ and Mo $K\alpha$, and the M_I and M_{II} edges lie between Cr $K\beta$ and Cr $K\alpha$. The experimental points of Fig. 3 verify the predictions derived from Fig. 4. The M_I , M_{II} edges for plutonium are designated as a complex because the experimental equipment could not resolve any difference between them.

The dotted line in Fig. 3 indicates the absorption coefficients of thorium and uranium, and the estimated absorption coefficient of plutonium, as given in the *Internationale Tabellen*. The high atomic number region of the absorption coefficients presented in the *Internationale Tabellen*, by failing to predict the L_I , L_{II} , and M_I absorption edges and yielding numerical values that at times disagree with experimental values by approximately 25%, lead the author to conclude that this region is probably considerably in error. The absorption coefficients in the *Internationale Tabellen* are based on Jonsson's⁷ "universal" absorption curve. In deriving this curve, Jonsson measured the absorption coefficients of five elements: aluminum, nickel, copper, silver, and platinum. While Jonsson's results can be assumed to be correct for the middle region of atomic numbers; i.e., $Z=20$ to 71, extrapolation to either lower or higher numbers may not be justified. Woernle⁸ has reported systematic deviations from Jonsson's method of representation for light elements ($Z=1$ to 18) and Fig. 3 indicates the amount of disagreement when extrapolation is made to high atomic numbers.

The mass absorption coefficients of thorium, uranium, and plutonium between absorption edges can be represented by an equation of the form

$$\mu_m = C\lambda^n. \quad (4)$$

Between the L_{III} and M_I edges the value of C is 138,

⁷ E. Jonsson, thesis, Upsala, 1928 (unpublished); see reference 9.

⁸ B. Woernle, *Ann. Physik* **5**, 475 (1930); see reference 9.

136, and 145 for thorium, uranium, and plutonium, respectively. The value of n was determined to be 2.44, 2.47, and 2.82 for thorium, uranium, and plutonium. These elements, therefore, were not found to obey the general λ^3 law. The results are consistent, however, with the observation of Compton and Allison,⁹ who note that for high Z numbers or long wavelengths, or both, the value of n is reduced from the "ideal" value of 3.

If the assumption is made that the absorption of x-ray energy by an atom is a function of the electronic configuration of the atom, particularly the configuration in the outermost shells, then an interesting correlation can be made between the way the absorption coefficient varies as a function of wavelength (the quantity n in $\mu_m = C\lambda^n$) and the electronic configuration in the outermost shells. Seaborg, Katz, and Manning¹⁰ propose a configuration in which the number of 5f electrons are 1, 3, 5 or 0, 3, 6 for thorium, uranium, and plutonium, respectively. See Table II. Zachariasen and Ellinger,¹¹ through the use of experimentally determined metallic radii, deduced that the number of 5f electrons in thorium, uranium, and plutonium are, respectively, 0, 0, and 3. If for thorium, uranium, and plutonium n increased in a fairly uniform manner, one might argue effectively for the constant increase in 5f electrons proposed by Seaborg *et al.* However, since n for plutonium is experimentally quite a bit greater than for

TABLE II. Correlation between absorption of x-ray energy and electronic configuration for thorium, uranium, and plutonium.

Z	n	5f electrons		
		Seaborg, Katz, Manning	Zachariasen, Ellinger	
90	2.44	1	0	0
92	2.47	3	3	0
94	2.82	5	6	3

thorium or uranium, the author favors the electronic configuration determined by Zachariasen and Ellinger on the basis of effective metallic radii.

The configuration proposed by Seaborg *et al.* is for the ground state of a neutral gaseous atom. As Seaborg points out, one cannot be sure that the configuration of a gaseous atom will be the same as the configuration of an atom in a compound or even as a hydrated ion in solution. However, the correlation between n and the configuration given by Zachariasen and Ellinger indicates that the metallic properties of thorium, uranium, and plutonium are retained upon dissolving up to 4 to 5 w/o of these elements in 1M HClO₄ and that the process of putting into solution either thorium, uranium, or plutonium, does not disturb appreciably the small energy differences between the 5f-6d-7s shells of these elements.

THEORETICAL INTERPRETATION

In the previous sections the author discussed the problems involved and the results obtained in the experimental determination of the x-ray absorption coefficients of thorium, uranium, and plutonium. The following sections are an attempt to calculate the x-ray absorption coefficients of thorium, uranium, and plutonium by considering the atomic number, a screening constant, principal quantum numbers, and the atomic energy levels characteristic of these elements.

Method of Theoretical Calculation

Victoreen has shown that the mass absorption coefficient of an element is given by an equation of the form

$$\mu_m = \mu_a + \mu_s = \left[\frac{3\lambda^3}{\lambda_0\lambda_1\lambda_2} - \frac{\lambda^4}{\lambda_0\lambda_1\lambda_2\lambda_3} \right] \pi l_0^2 \frac{N_0}{A} + \sigma_0 Z \pi l_0^2 \frac{N_0}{A}, \quad (5)$$

where μ_m = mass absorption coefficient, μ_a = true absorption coefficient, μ_s = scattering coefficient, λ = incident wavelength in A, $\lambda_0 = e^2/mc^2 = 2.8197 \times 10^{-5}$ A, $\lambda_1\lambda_2\lambda_3$ = critical wavelengths in A characteristic of the atom under consideration, $l_0 = e^2/mc^2 = 2.8197 \times 10^{-18}$ cm, $N_0 = 6.0228 \times 10^{23}$ mole⁻¹, A = atomic weight (g/mole), σ_0 = dimensionless portion of scattering coefficient, and Z = atomic number.

The critical wavelengths $\lambda_1\lambda_2\lambda_3$, or more conveniently the critical wave numbers, $1/\lambda_1, 1/\lambda_2, 1/\lambda_3$, are calculated from the difference in atomic energy levels as follows:

$$1/\lambda = E_2/hc - E_1/hc, \quad (6)$$

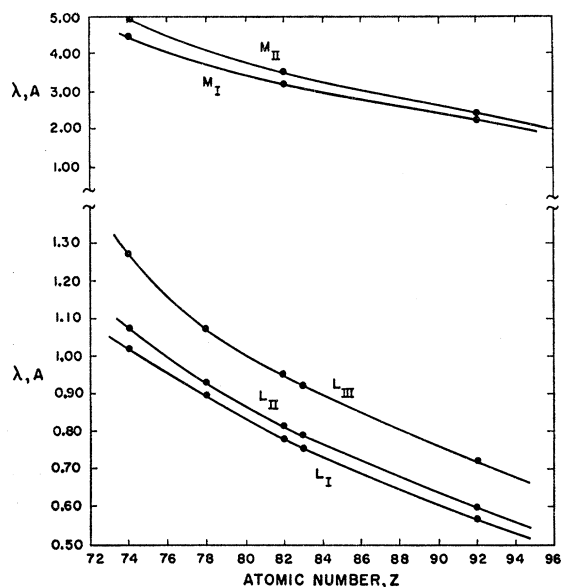


FIG. 4. Variation of L and M absorption edges as a function of atomic number, Z .

⁹ A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), pp. 526-542.

¹⁰ Seaborg, Katz, and Manning, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1950), National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV, Part II, p. 1509.

¹¹ W. H. Zachariasen and F. H. Ellinger, *J. Chem. Phys.* **27**, 811 (1957).

TABLE III. Screening constants and quantum conditions for thorium, uranium, and plutonium.

Z	K-L _I	L _I -L _{II}	L _{II} -L _{III}	s	L _{III} -M _I	M _I -M _{II}	M _{II} -M _{III}	λ ₁		λ ₂		λ ₃	
								n ₂	n ₁	n ₂	n ₁	n ₂	n ₁
90	13	22	23.5	36.5	1	∞	2	∞	2	∞	
92	11	...	22	38.5	40.2	...	1	∞	2	∞	2	∞	
94	19	27	27.7	41.3	...	43.5	1	∞	2	∞	3	∞	

where λ =wavelength in Å, h =Planck's constant (erg sec), c =velocity of light (Å/sec), and E_2, E_1 =energies in ergs of two quantum states within the atom.

The atomic energy levels of Eq. (6) are calculated by means of the expression (Victoreen¹)

$$\frac{E}{hc} = R \left[\frac{(Z-s)^2}{n^2} + \left[\frac{\alpha^2(Z-s)^4}{4n^4} + \frac{\alpha^4(Z-s)^6}{8n^6} + \frac{\alpha^6(Z-s)^8}{12.8n^8} + \frac{\alpha^8(Z-s)^{10}}{18.28n^{10}} + \frac{\alpha^{10}(Z-s)^{12}}{24.35n^{12}} + \dots \right] \right], \quad (7)$$

where R =Rydberg's constant ($0.00109737 \text{ Å}^{-1}$), Z =atomic number, s =screening constant, n =a principal quantum number, and $\alpha=2\pi e^2/ch$, the fine structure constant. The bracketed term in Eq. (7) is the relativity correction which must be carried to the higher powers of α^2 until the contribution of all the following terms may be neglected.

It may be seen that the bracketed term in Eq. (5) is dimensionless and that the units for μ_m are cm^2g^{-1} . The scattering coefficients for thorium, uranium, and plutonium are of the order of magnitude of $0.15 \text{ cm}^2\text{g}^{-1}$ and are therefore negligible in comparison to either the true absorption coefficients or the mass absorption coefficients.

By substituting and collecting numerical values for the constants and neglecting the scattering coefficient, Eq. (5) can be reduced to

$$(\mu_m)_Z = C\lambda^3 - D\lambda^4. \quad (8)$$

Results of Theoretical Calculations

In Table III are listed the screening constant, s , applicable to a given wavelength region (i.e., $s=36.5$ between the L_{III} absorption edge and the M_I absorption edge for thorium) and the principal quantum number, n , used to determine a critical wave number by use of Eq. (7).

TABLE IV. Values of C and D for use in $\mu_m = C\lambda^3 - D\lambda^4$ for thorium, uranium, and plutonium.

	Thorium		Uranium		Plutonium	
	C	D	C	D	C	D
K-L _I	818	444	987	615	709	165
L _I -L _{II}	483	212	440	81
L _{II} -L _{III}	440	188	530	242	420	75
L _{III} -M _I	183	45	180	44	163	19
M _I -M _{II}	154	35
M _{II} -M _{III}	138	15

Table IV lists the values of C and D used in Eq. (8), obtained by application of the values of Table III to Eqs. (7), (6), and (5).

Table I allows comparison of the calculated coefficients obtained from Eq. (8) with the experimental coefficients.

Utilizing the numerical values of Tables III and IV, good agreement is obtained between the calculated coefficients and the experimentally observed coefficients as given in Table I and graphically illustrated in Fig. 5 for the L_{III} - M_I region.

In determining the values listed in Table III, it was found that the quantum conditions used to calculate a critical wavelength (i.e., $1/\lambda_2=2$ to ∞ for thorium) remained the same on both sides of an absorption edge

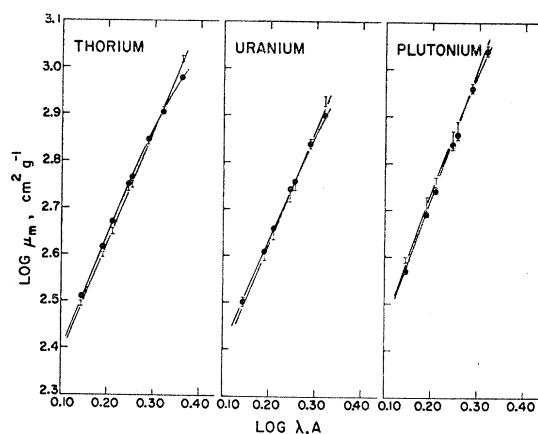


Fig. 5. Comparison of experimental (I) and calculated (•) x-ray absorption coefficients for thorium, uranium, and plutonium in the L_{III} - M_I region.

but that the value of the screening constant, s , changed (i.e., s jumps from 23.5 to 36.5 across the L_{III} edge for thorium). Minor changes in quantum conditions at an absorption edge; for example, $1/\lambda_2=2$ to 10 instead of 2 to ∞ (representing a 3.5% decrease in the final value of $1/\lambda_2$), even when compensated for by changes in the screening constant, s , do not yield calculated coefficients that are in acceptable agreement with the observed coefficients.

A comparable situation was found by Victoreen for gold in which the observed and calculated coefficients were matched by having identical quantum conditions on either side of an absorption edge while the value of s changed at an absorption edge.

Discussion of Theoretical Calculations

In Fig. 6 the screening constant, s , is shown as a function of atomic number, Z . The increase in the screening constant for plutonium over that for thorium and uranium is interpreted in a manner analogous to Compton and Allison's¹² treatment for the $4f$ electrons. If the screening constant of Sommerfeld¹³ is plotted as a function of Z and a line drawn through the points one can observe a sharp increase in the slope of the line at $Z=57$, a decrease in the slope at $Z=70$ and a further decrease in the slope at $Z=80$. An explanation of these changes in slope is obtained by considering that the contribution to the screening constant by a single electron is greater in a shell of smaller radius than in one of larger radius. At $Z=47$, where the screening constants of Sommerfeld start, added electrons enter into the $5s$ or $5p$ shells, but at the beginning of the

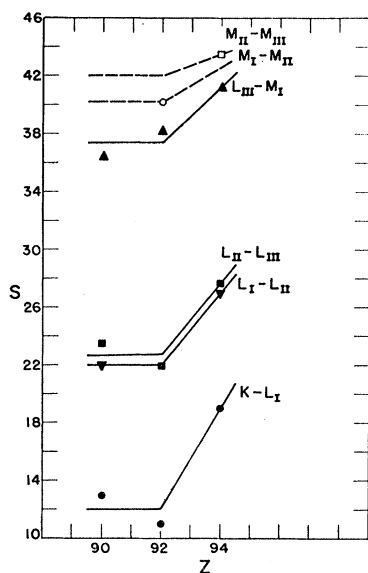


FIG. 6. Variation of the screening constant, s , with atomic number for thorium, uranium, and plutonium.

rare earths, at $Z=57$, added electrons begin to enter the $4f$ shells, which are presumably of smaller effective radii than the $5s$ or $5p$ shells, and thus an increase in the slope is observed. At $Z=70$ when the $4f$ shell is filled, added electrons enter into the $5d$ shell which has a larger effective radius than the $4f$ shell and a decrease in slope is observed. At $Z=80$ when the $5d$ shell is filled added electrons enter the $6p$ shell which has a larger effective radius than the $5d$ shell and the slope is thus observed to decrease still further.

Seaborg, Katz, and Manning propose an electronic configuration in which the number of electrons in the

$5f$ shell is either 1, 3, 5 or 0, 3, 6 for thorium, uranium, and plutonium, respectively. On the other hand, Zachariasen and Ellinger, through the use of experimentally determined metallic radii, find a configuration which has 0, 0, and 3 electrons in the $5f$ shell for thorium, uranium, and plutonium. By interpreting the increase in the screening constant as shown in Fig. 6 as evidence for entrance of electrons into the $5f$ shell, the author concludes that the configuration of Zachariasen and Ellinger is to be preferred over that of Seaborg *et al.*

Through a consideration of the number of electrons in the valence states and in the $5f$ shell as given by Zachariasen and Ellinger, Elliott¹⁴ has concluded that plutonium is a transition element within the $5f$ series occupying a unique position as an intermediate between a "thoride" subseries represented by atomic number 90 to 93 and an "actinide" subseries consisting of those elements having atomic numbers greater than 94.

Since in thorium and uranium, electrons seem to prefer the valence states (4 and 6 electrons respectively for thorium and uranium) it would appear that the $5f$ shell represents a higher energy condition than the valence states. However, in americium electrons seem to prefer the $5f$ shell (6 electrons) instead of the valence states (3 electrons) making the $5f$ shell representative of a lower energy condition than the valence states. Plutonium, having nearly equal number of electrons in the $5f$ shell (3 electrons) and the valence states (5 electrons) is thus an intermediate with the $5f$ shell representative of a lower energy than the $5f$ shell of thorium and uranium but of higher energy than the $5f$ shell of americium.

Support for this scheme of $5f$ shell energy levels can be obtained from an examination of the quantum conditions in Table III, which give rise to the critical wave numbers used in the calculation of absorption coefficients. For thorium and uranium λ_3 is given by the difference in energy levels obtained from Eq. (7) when $n=2$ and $n=\infty$, while for plutonium λ_3 is given by the difference in energy levels when $n=3$ and $n=\infty$. Since the energy level characterized by $n=2$ is greater than one characterized by $n=3$, this evidence conforms to the $5f$ shell energy level scheme outlined above and thus tends to confirm Elliott's conclusion that plutonium is a transition point in the $5f$ series of elements.

ACKNOWLEDGMENTS

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¹² See reference 9, p. 627.

¹³ A. Sommerfeld, *Atomic Structure and Spectral Lines* (Methuen and Company Ltd., London, 1934), third edition.

¹⁴ R. O. Elliott, thesis, University of California, 1958 (unpublished).