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# ELECTRON CAPTURE STUDIES IN SHIELDED NUCLEI

Harold Jaffe

(Thesis)

April 5, 1954

Berkeley, California

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Electron Capture Studies in Shielded Nuclei

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April 5, 1954

ABSTRACT

A search for electron capture in shielded nuclei has been undertaken using, in most cases, a 10 inch bent crystal spectrometer to detect the characteristic x-rays. Electron capture decay has been confirmed in $Am^{242m}$, $Ir^{192}$, $Tl^{204}$ and $Np^{236}$ and detected for the first time in $Am^{242}$ and $Sb^{122}$. Relative abundances have also been determined. Upper limits for this type of decay have been set in $Np^{238}$, $Ag^{110m}$, $Ag^{110}$, $Cs^{134}$, $Tb^{160}$, $Tm^{170}$ and $Sb^{124}$. Certain features of the gamma ray spectra and decay schemes of several of the above nuclides have been studied.

An apparent discrepancy in the gamma ray spectrum of $Am^{241}$ has been resolved and several new transitions have been observed.

A reported anomaly in the L x-ray energies of uranium has been shown to be due to an instrumental effect.

# Electron Capture Studies in Shielded Nuclei

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## I. INTRODUCTION

A shielded nucleus $Z^A$ is one which has two stable neighboring isobars, $(Z - 1)^A$ and $(Z + 1)^A$. Such nuclei are always composed of an odd number of neutrons and an odd number of protons, and consequently have an even mass number, A. The two neighboring isobars are of the even-even type, and because they are both stable isotopes $Z^A$ will decay to $(Z + 1)^A$ by beta decay (negatron emission) and to $(Z - 1)^A$ by positron emission, electron capture or both. The extent to which these three modes of decay compete is governed mainly by the respective maximum energies of the emitted particle and parity and total angular momentum, or spin, changes involved in the transition. In the case of electron capture the neutrino is the emitted particle referred to above.

The only energetic requirement for the beta decay process is that the atomic mass of the parent nuclide exceeds that of the daughter. Electron capture further requires that this mass difference be equivalent to an energy larger than the binding energy of the particular electron that is captured. Positron decay is only possible when the mass difference exceeds twice the electron mass or 1.02 Mev.

It is generally accepted that the ground state of an even-even nucleus has zero spin and even parity. Recently it has been shown[1,2] that the first excited states of such nuclei usually have two units of angular momentum and even parity. Marked regularities have been observed in these and higher levels.[2,3,4] Since it is to be expected that competing modes of decay of a shielded nucleus populating equivalent spin states of the daughters would be of equal "order of forbiddenness" it should be possible to correlate, at least qualitatively or semiquantitatively, the relative rates of the three types of transitions with decay energy data. Of course some information is usually needed concerning the excited levels of the respective daughters.

The determination of beta decay energies is straightforward, but the corresponding measurement in electron capture is generally difficult to make. One of the easiest methods is by closed decay cycle calculations, but this is only applicable in the heavy elements where alpha radioactivity occurs. In lighter elements, if positron emission competes with electron capture the electron capture energy is readily determined from a knowledge of the decay scheme and the maximum kinetic energy of the positrons. Several measurements have been made[5,6,7] of the continuous gamma ray spectrum accompanying electron capture. Generally the entire decay energy is carried by the neutrino in electron capture, but occasionally it may be shared with a photon. The maximum energy of the resultant gamma ray spectrum equals the neutrino energy in the absence of this effect. This method is applicable only in certain optimum cases and is hampered by the relatively small probability for emission of continuous gamma radiation. Another method of measuring electron capture decay energies

is by precise determinations of the (p,n) reaction threshold. This allows calculation of the mass difference between two neighboring isobars. Of course, high precision mass spectrometry by the matched doublet technique can give the same information.

The detection of electron capture in shielded nuclei depends, as a rule, upon identification of the characteristic x-rays of the daughter emitted when the electron vacancy caused by the capture event is filled. Throughout this research the x-ray energies tabulated by Compton and Allison[8] and corrected by Dauben[9] for the latest values of the physical constants were employed. In the heavy elements ($Z > 92$) the energies were determined by a Mosely extrapolation of the data on thorium and uranium listed by Siegbahn.[10]

Two additional problems arising from previous work performed in this laboratory were studied and are discussed in the appendix. The first of these deals with major discrepancies in the gamma ray spectrum of $Am^{241}$ as determined by bent crystal spectrometer experiments[11] on the one hand and proportional counter studies[12] on the other. The second concerns an apparent anomaly[11] in the energy of the uranium L x-rays excited in the beta decay of $Pa^{232}$ and $Pa^{233}$.

## II. EXPERIMENTAL METHODS

In order to detect and identify the characteristic x-rays of an electron capture daughter a 10 inch bent crystal spectrometer of the Cauchois type was employed. This instrument has been described by Barton *et al.*[13] and somewhat modified by Browne.[11] The experimental approach was essentially the same used by the previous workers. The sample in solid form was powdered and mounted in a thin pyrex capillary.

The diameter of the capillary was about 0.015 inch and undoubtedly represented the limiting factor in the obtainable resolution. The capillary was secured in a groove on a lucite holder and this was mounted and aligned in the sample carriage.

A 0.008 inch thick topaz crystal elastically bent to a 10 inch radius was used to diffract the incident electromagnetic radiation. The 303 planes whose spacing was determined as 1.356 A in the unbent crystal[11] were employed and the energies in kev were calculated using a conversion factor based upon the latest values of the physical constants reported by Dumond and Cohen.[14]

The detector, mounted behind a collimator, consisted of a NaI (thallium activated) scintillation crystal with a RCA 5819 photomultiplier tube. A standard scaler and traffic counter were used to record the data.

In general the sample arm moved about the focal circle at a rate of $4^{\circ}$ per 400 minutes. Faster sweeping rates were possible but usually not profitable except in the study of activities of relatively short half-life. Photon lines were manifest by peaks in a plot of counting rate versus angular position. The latter was read from a graduated scale mounted about the focal circle. The Bragg angle and consequently the energy, was determined by observing the angular position of a line on both sides of the undiffracted beam or midpoint. Limits of error were calculated only taking into account the angular uncertainties. An accuracy of $0.01^{\circ}$ in the Bragg angle was considered the limit of the instrument.

The latter portion of the appendix describes some experiments which indicated the graduations on the above mentioned scale were somewhat non-uniform. Consequently, a new scale was accurately machined and mounted on the spectrometer. The $Np^{238}$, $Np^{239}$, and $Pu^{238}$ spectra were taken using the old scale and consequently the energy determinations in certain regions may be in error by an amount greater than the stated limits.

The line intensities were subject to correction for self absorption in the sample, reflectivity of the crystal, and absorbers in the path between sample and detector. The self absorption correction was based on an equation reported by Dixon[15] for cylindrical samples. It was necessary to know the absorption coefficients of the sample as a function of energy and in some cases these had to be estimated from Jönsson's "universal absorption curve."[16] If the energy of a line is very close to the K edge of the bulk sample matter or in the region of its three L edges, this correction can become quite severe and uncertain. Consequently, in those experiments where the activity could be obtained in essentially weightless form a low Z carrier was used.

Lind et al.[17] have found that the variation of reflectivity from a quartz crystal as a function of energy was inversely proportional to the square of the energy. Browne observed that lines widely spaced in energy maintained the same intensity ratio when reflected from both quartz and topaz indicating the same energy dependence for the latter crystal.

The correction for absorbers in path was based on the measured thickness of the matter in the path of the radiation and values of the absorption coefficients of this matter taken from the data listed in Compton and Allison.[18] This compilation was also used whenever possible in making the self absorption correction.

The useful range of the bent crystal spectrometer was 10 to approximately 110 kev. Millicurie activity levels were generally required.

In order to obtain absolute abundances of various x-rays and gamma rays, small samples were often counted in a $4\pi$ methane flow proportional counter to determine the disintegration rates. These were then examined with other instruments to be described shortly. A Tygon film covering a thin aluminum plate with a 0.25 inch center hole served as the sample mount. The activity was placed over the hole so that the film was the only backing material. The major portion of the Tygon on aluminum was removed by scraping to avoid electrostatic charge build up and its adverse effect on the proper functioning of the $4\pi$ counter.

In order to determine absolute line intensities samples of known disintegration rates were frequently studied with a NaI (thallium activated) crystal detector, 1.08 x 1.25 inch diameter, sealed to a Dumont 6292 photomultiplier tube. The output of this tube, after suitable amplification, was led to a 50 channel differential pulse height analyzer, thus allowing observation of the electromagnetic energy spectrum. The counting efficiency of a sodium iodide crystal of the above dimensions has been determined by McLaughlin and O'Kelley.[19] A geometry factor was measured by observing the intensity of the 59.6 kev radiation

associated with a known amount of $Am^{241}$. The absolute abundance of this gamma ray has been determined[12] as 0.40 ± 0.015 photon per alpha particle and its detection efficiency is 100 percent in a crystal of the above dimensions. Suitable corrections which were experimentally determined were applied for the escape peak effect resulting from the loss of an iodine K x-ray from the crystal. Such corrections are only important in the energy region below 150 kev. The scintillation spectrometer was calibrated using the well known gamma rays of $Am^{241}$, $Cd^{109}$, and $U^{235}$ as standards in the low energy region and those of $Cs^{137}$, $Co^{60}$, and $Na^{22}$ in the high region.

A proportional counter tube containing 90 percent xenon and 10 percent methane (1 atmosphere total pressure) was connected to the 50 channel analyzer and used to determine the absolute x-ray abundance in several experiments. A counting efficiency curve based on the L x-rays of the $Am^{241}$ decay has been determined for this counter by Hoff.[20] His data was corrected using the more recent intensity values of Beling, et al.[21] The radiations of $Am^{241}$ were also used as an energy standard, along with the silver K x-rays following the decay of $Cd^{109}$.

## III. EXPERIMENTAL RESULTS

### A. $Np^{238}$

The 2.1 day $Np^{238}$ decays to $Pu^{238}$ by negatron emission. Several levels of the daughter are populated, and gamma rays and internal conversion electrons have been observed.[22,23] Closed decay cycles[24] indicate approximately 120 kev are available for electron capture to beta stable $U^{238}$. With such a small disintegration energy at this Z one might expect the dominant mode of electron capture to be L capture.

Neutron irradiated neptunium oxide was dissolved and brought to 2 M HCl concentration. The neptunium was reduced to the IV state with formic acid and extracted into 0.4 M thenoyltrifluoroacetone (TTA) in benzene. The organic phase was washed twice with 2 M HCl and then the neptunium was back extracted into 10 M $HNO_3$. The aqueous solution was separated and evaporated to dryness and the neptunium was taken up in 2 M HCl. After two such cycles HF was added to the purified product in 2 M HCl and the fluoride was precipitated. Approximately 1 milligram of neptunium was powdered and mounted for the bent crystal spectrometer.

Five plutonium x-rays arising from the beta decay were observed. The data are listed in Table 1. A diagram of L x-ray nomenclature is shown in Figure 1.

Table 1

L X-Rays Following the Decay of $Np^{238}$

| Line | Transition | Observed energy (kev) | Siegbahn's extrapolated energy (kev) | Corrected relative intensity |
|---|---|---|---|---|
| Pu $L\gamma_6$ | $L_2$-$O_4$ | 22.28 ± 0.04 | 22.13 | 13 |
| Pu $L\gamma_1$ | $L_2$-$N_4$ | 21.49 ± 0.02 | 21.38 | 36 |
| Pu $L\beta_1$ | $L_2$-$M_4$ | 18.37 ± 0.01 | 18.27 | 100 |
| Pu $L\beta_2$ | $L_3$-$N_5$ | 17.36 ± 0.03 | 17.25 | 13 |
| Pu $L\alpha_1$ | $L_3$-$M_5$ | 14.32 ± 0.02 | 14.28 | 27 |

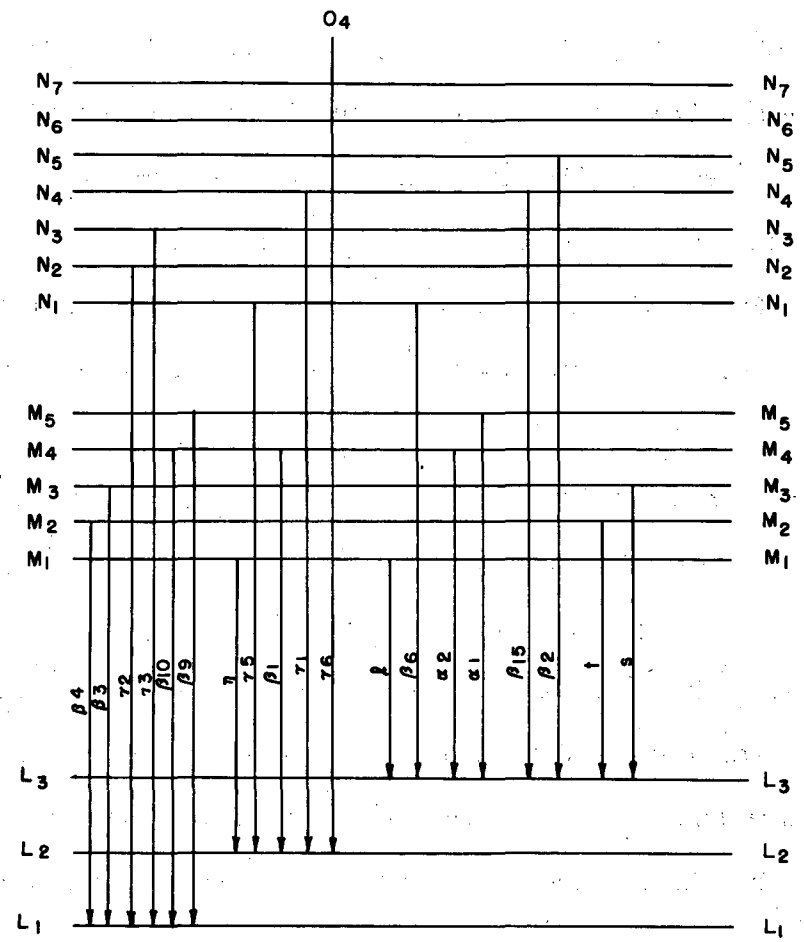


Fig. 1. Diagram of L x-ray nomenclature.

The relative quantum yields (photon intensities) of the $L_2$ and $L_3$ shells from these data are

$$L_2:L_3 = 3:1 .$$

A correction has been applied for the intensities of the $L_2$ and $L_3$ x-rays not observed. This correction is based on the abundance ratios of the L x-rays from each of the various L shells observed in the decay of $Am^{241}$ (see Appendix, Table 17). It is assumed that these ratios do not change rapidly with atomic number.

By far the majority of the observed x-rays arise from L conversion of a 44.3 kev gamma ray[23,25] in the plutonium daughter. This transition is also observed following the alpha decay of $Cm^{242}$.[26] In order to obtain the ratio of the $L_2$ and $L_3$ vacancies one must correct the observed x-ray intensities for fluorescence yield (fraction of the vacancies from a given shell that are filled by a radiative transition). Kinsey[27] has calculated these L shell yields using experimental data on total level widths combined with theoretical estimates of the radiation widths. His results extrapolated to Z = 94 indicate 0.61 and 0.43 for the fluorescence yield of the $L_2$ and $L_3$ shells, respectively. Using these figures the L vacancy ratio is

$$L_2:L_3 = 2:1 .$$

(The amount of $L_1$ excitation is unknown but presumed to be small.) This is to be compared to the value of 1.4 determined by Slätis, Rasmussen and Atterling,[23] and 2.0 from the data of Freedman, et al.[22] as modified by Mihelich.[25] The gamma ray giving rise to these L vacancies is an electric quadrupole transition.[26] The theoretical conversion coefficients[28] would indicate a ratio of approximately 1.1.

No x-ray lines of the electron capture daughter were observed. From the absolute L x-ray abundance calculated from conversion line data, and the maximum undetectable intensity of several uranium x-rays estimated from the observed spectrum it is possible to set an upper limit of 4 percent for $L_1$ electron capture and 2 percent for $L_2$ capture in $Np^{238}$. The actual amount of L capture is probably much less than this because of the small available energy.

## B. $Am^{242m}$

The 16 hour $Am^{242m}$ is an ideal isotope for study with the bent crystal spectrometer. Indeed, the first detailed investigation of this activity[29] included the use of this instrument and the proposed decay scheme, which consists of electron capture, isomeric transition and $\beta^-$ decay, was based mainly on the identification of the L x-rays of the three daughter nuclei. Several factors, including the fact that the first excited states of the two even-even daughters as determined by beta ray spectroscopy seemed somewhat out of line with the trend in this region,[2,3] prompted the reinvestigation of this isotope.

$Am^{241}$ in the form of the oxide, was neutron irradiated for five days in the Materials Testing Reactor at Arco, Idaho. A small portion of the sample was dissolved in HCl and several fluoride and hydroxide precipitations were carried out to separate the americium from the major fraction of the fission products. A Dowex-50 cation exchange column with 13 M HCl allowed the removal of rare earth activities.

The americium was then oxidized to the hexapositive state using acid persulfate and curium was removed by a $LaF_3$ scavenge. The purified americium was reduced with hydrazine and another fluoride-hydroxide precipitation cycle was carried out. The final product was converted to the oxide by heating and approximately 1.5 milligrams was mounted for the bent crystal spectrometer sample. Slightly more than 2 half-lives elapsed between the time of removal from the reactor and the mounting of the sample. The very high specific acitivity allowed a more detailed analysis of the spectrum than was previously possible.

The electromagnetic radiation in the region of 13 to 60 kev was examined with the bent crystal spectrometer. The 100 kev region of the K x-rays from K electron capture was not investigated because of time limitations and also because it was found that the intensity of the K x-rays relative to the $Am^{241}$ 59.6 kev gamma ray in the sample could be readily determined with the scintillation spectrometer. Since the 59.6 kev line was also observed on the bent crystal spectrometer the number of K x-rays relative to the L x-rays could be calculated.

The data on the K and L x-rays associated with the decay of the 16 hour $Am^{242m}$ are listed in Table 2. The intensities have been corrected in the usual fashion. L x-rays of neptunium arising from the decay of $Am^{241}$ in the sample were also observed. A portion of the spectrum showing the $L_\alpha$ complex is given in Figure 2.

There is some ambiguity in the assignment of the americium $L\beta_1$ and the plutonium $L\beta_3$ lines. In the former case the energy of the curium $L\beta_5$ ($L_3$-$O_{4,5}$) is expected to be 18.86 kev. From the ratio of the $L\beta_5$ to $L\alpha_1$ observed in the decay of $Am^{241}$ and the abundance of the

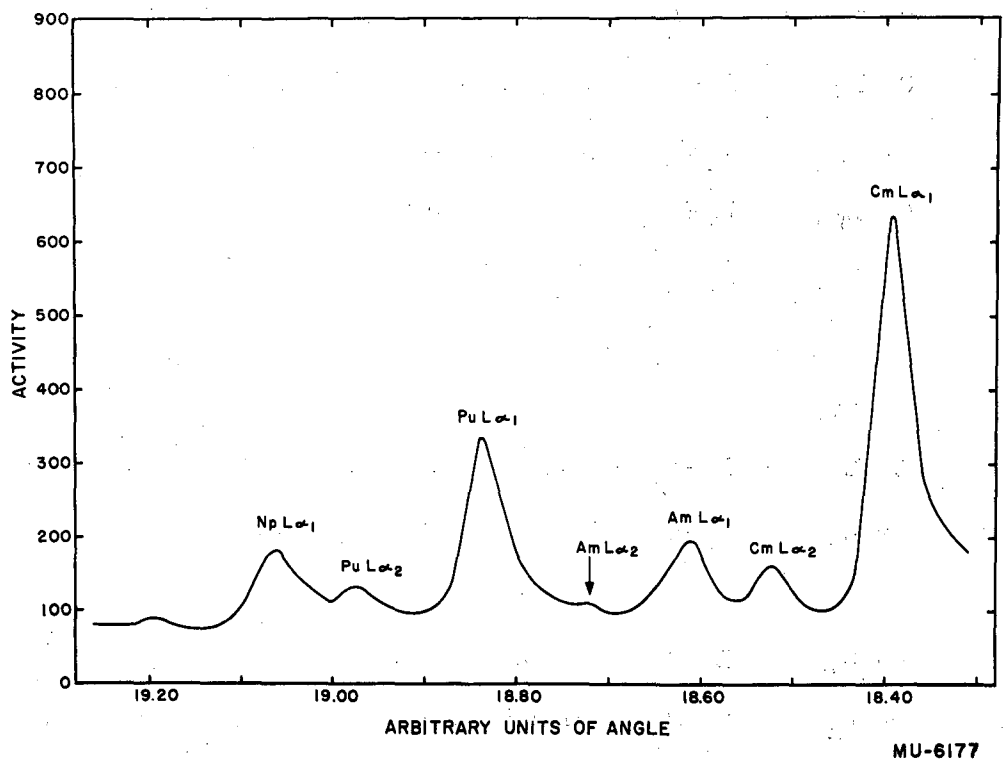


Fig. 2. $L_\alpha$ x-rays of curium, americium, plutonium, and neptunium from $Am^{241}$ and $Am^{242m}$.

Table 2

X-Rays Following the Decay of $Am^{242m}$

| Line | Transition | Observed energy (kev) | Siegbahn's extrapolated energy (kev) | Corrected relative intensity |
|---|---|---|---|---|
| Cm $L\alpha_2$ | $L_3$-$M_4$ | 14.75 ± 0.03 | 14.74 | 4 |
| Cm $L\alpha_1$ | $L_3$-$M_5$ | 14.97 ± 0.03 | 14.96 | 27 |
| Cm $L\beta_2$ | $L_3$-$N_5$ | 18.09 ± 0.02 | 18.10 | 16 |
| Cm $L\beta_1$ | $L_2$-$M_4$ | 19.47 ± 0.02 | 19.38 | 100 |
| Cm $L\gamma_1$ | $L_2$-$N_4$ | 22.79 ± 0.04 | 22.63 | 40 |
| Cm $L\gamma_3$ | $L_1$-$N_3$ | 23.30 ± 0.06 | 23.25 | 6 |
| Cm $L\gamma_6$ | $L_2$-$O_4$ | 23.62 ± 0.12 | 23.46 | 12 |
| Am $L\alpha_2$ | $L_3$-$M_4$ | 14.44 ± 0.06 | 14.41 | 1 |
| Am $L\alpha_1$ | $L_3$-$M_5$ | 14.61 ± 0.03 | 14.61 | 4 |
| Am $L\beta_1$ | $L_2$-$M_4$ | 18.89 ± 0.02 | 18.80 | 6 |
| Pu $L\alpha_2$ | $L_3$-$M_4$ | 14.08 ± 0.03 | 14.08 | 2 |
| Pu $L\alpha_1$ | $L_3$-$M_5$ | 14.28 ± 0.03 | 14.28 | 10 |
| Pu $L\beta_2$ | $L_3$-$N_5$ | 17.29 ± 0.03 | 17.25 | 9 |
| Pu $L\beta_1$ | $L_2$-$M_4$ | 18.33 ± 0.02 | 18.27 | 27 |
| Pu $L\beta_3$ | $L_1$-$M_3$ | 18.62 ± 0.04 | 18.52 | 7 |
| Pu $L\gamma_1$ | $L_2$-$N_4$ | 21.46 ± 0.04 | 21.38 | 16 |
| Pu $L\gamma_3$ | $L_1$-$N_3$ | 22.06 ± 0.10 | 21.97 | 7 |
| Pu $L\gamma_6$ | $L_2$-$O_4$ | 22.24 ± 0.10 | 22.13 | 7 |
| Pu K x-rays | | 102 | | 37 |

curium $L_{\alpha_1}$ it was possible to correct the intensity of the americium $L_{\beta_1}$ to the figure listed. The contribution of the curium $L_{\beta_5}$ to the observed line was 17 percent.

The calculated energy of the curium $L_{\beta_4}$ ($L_1$-$M_2$) is 18.57 kev and thus it may contribute to the plutonium $L_{\beta_3}$ line. The chosen assignment is based on the assumption that the $L_1$ lines of plutonium are more intense than those of curium because of $L_1$ electron capture. Since little $L_1$ conversion is expected from the low energy first excited state transitions in even-even nuclei the total intensity was assigned to the plutonium line.

There is considerable uncertainty in the abundance of the americium L x-rays. It was observed that all these lines decayed to some extent and then remained constant with time. The latter effect is undoubtedly due to self excitation of the bulk $Am^{241}$ in the sample, and a correction for this effect has been applied to the intensities. However, the presence of x-rays with the $Am^{242m}$ decay capable of exciting the americium $L_2$ and $L_3$ shells indicates the observed relative abundances are merely upper limits. At least part of the americium $L_3$ lines are due to fluorescence excitation by the intense curium $L_{\beta_1}$ which is only 1 kev above the americium $L_3$ edge (18.52 kev). Greater intensity of the americium lines found in the earlier study[29] was very likely due to greater sample mass and other effects which increased the amount of fluorescence excitation.

The relative L shell quantum yields of the two even-even daughters are:

$$\text{Cm} \quad L_2:L_3 = 160:50$$
$$\text{Pu} \quad L_2:L_3 = 50:20 \quad .$$

These figures are based on the observed intensities and were corrected for the abundance of the L lines not observed using the data on $Am^{241}$ (see Appendix, Table 17). The $L_1$ quantum yield has been omitted because of uncertainties in line assignments and low abundance.

Using fluorescence yields based on Kinsey's estimates[27] one can calculate the relative L shell vacancies. However, the possibility of nonradiative transitions which shift an electron vacancy from one L shell to another must be considered. Such transitions were suggested by Coster and Kronig[30] to explain certain x-ray satellite lines. The $L_1$-$L_3$ shift with the ejection of an M electron should be, according to Kinsey, the most prominent mode of filling the $L_1$ shell. Previous data obtained in this laboratory[11] and also the work on $Am^{241}$ indicates the magnitude of the Coster-Kronig coefficient is smaller than the estimated value. Because of these uncertainties, the following vacancy ratios were calculated ignoring this effect. The fluorescence yield of the plutonium K shell is estimated as 98 percent.[31,32]

$$\text{Cm} \quad L_2:L_3 = 250:110$$
$$\text{Pu} \quad K:L_2:L_3 = 40:80:50 \quad .$$

A correction must now be applied for the L vacancies in plutonium due to emission of K x-rays. Extrapolating the relative intensities of the K series radiations as listed by Compton and Allison[33] one finds approximately 25 percent of the K vacancies give rise to $L_2$ holes and

50 percent to $L_3$ holes at Z = 94. Taking this into account the corrected ratio of K vacancies to L vacancies in plutonium is as follows:

$$\text{Pu} \quad K:L_2:L_3 = 40:70:30 \quad .$$

The beta ray spectrometer studies by Passell[34] indicate the presence of two highly converted gamma rays and a beta spectrum of 628 kev maximum energy. His line data are summarized in Table 3.

Table 3

Conversion Lines from the Decay of $Am^{242m}$

| Electron energy | Conversion shell | Gamma ray energy (kev) | Best energy (kev) |
|---|---|---|---|
| 17.3 | Cm $L_2$ | 40.9 | |
| 22.0 | Cm $L_3$ | 41.0 | |
| 35.5 | Cm $M_2$ | 41.4 | 41.0 |
| 36.5 | Cm $M_4$-$M_5$ | 40.6 | |
| 39.9 | Cm N (or Pu M?) | 41.6 | |
| 20.9 | Pu $L_2$ | 43.2 | 43.4 |
| 25.5 | Pu $L_3$ | 43.6 | |

The L x-ray intensities lead to a ratio of curium to plutonium L conversion of 3.6, assuming only K or $L_1$ electron capture. This is to be compared to the value of 2.5 determined from the conversion electron abundances.

Beta L x-ray coincidence studies on $Am^{242m}$ by Frank Stephens[35] show 55 percent decay to the ground state and 45 percent to the first

excited state of $Cm^{242}$. Using this information, the reported branching ratio of 4.2 $\beta^-$/EC,[36] and the above conversion ratio, the electron capture branching ratio to the ground state and first excited state of $Pu^{242}$ is 1. This implies a $K/L_1$ capture ratio of 0.2. Close cycle calculations indicate the electron capture energy of $Am^{242}$ is 0.66 Mev. If the isomeric transition is 35 kev as discussed below, the electron capture energy of $Am^{242m}$ is 0.69 and 0.65 Mev to the ground state and first excited state of $Pu^{242}$, respectively. Such a low K/L capture ratio is very unusual for this amount of energy and certainly requires further study. An appreciable Coster-Kronig effect or $L_2$ and $L_3$ capture would tend to increase this value.

The $L_2$ to $L_3$ quantum yield of the americium lines is 3:2, and the calculated vacancy ratio is 1:1. On the basis of L conversion alone the isomeric transition occurs in 6 percent of the decays. However, this figure must be considered an upper limit because of fluorescence excitation.

Two gamma rays were observed on the bent crystal spectrometer and the results are compared to the beta spectrometer data of two investigators in Table 4.

Table 4

Gamma Rays Observed in the Decay of $Am^{242m}$

| Observed energy (kev) | Passell's[34] energy (kev) | Church's[37] energy (kev) | Intensity relative to Cm $L_{\beta 1}$ | $\alpha_L$ |
|---|---|---|---|---|
| 42.3 ± 0.2 | 41.0 | 42.2 | 0.7 | 500 |
| 44.8 ± 0.2 | 43.4 | 44.6 | 0.6 | 200 |

There were indications of a line at 43.6 kev, but this is very likely due to an instrumental effect. Because the entire spectrum was

examined at the slowest sweeping rate it was not possible to observe these lines more than once on each side of the spectrometer midpoint. The energies are in good agreement with those deduced from the electron lines.

The L shell conversion coefficients in Table 4 were calculated from the L vacancies and the intensities of the two gamma rays. $L_1$ conversion was not taken into account. The two gamma rays are very likely electric quadrupole transitions on the basis of comparison of these conversion coefficients with the theoretical values calculated by Gellman et al.[28] and represent the first excited state transitions in the two even-even daughters. The first excited state of $Cm^{242}$ had been inferred as about 40 kev from the alpha decay of $Cf^{246}$.[38]

At present it appears that the separation between the two $Am^{242}$ isomers is about 35 kev. This assumes that the measured beta particle energies of the two isomers are the respective ground state transitions. Since β L x-ray coincidence experiments indicate comparable population of the ground and first excited state of $Cm^{242}$ by both isomers the assumption is reasonable.

## C. $Am^{242}$

The present study of $Am^{242m}$ indicated that the conversion line assignments in the work of O'Kelley et al.[29] were somewhat in error. O'Kelley[39] had suggested that the long lived $Am^{242}$ electron captured to the same state as the 16 hour isomer on the basis of persistence of several electron lines which were thought to be due to L shell conversion in plutonium. These lines had decayed and then remained

constant with time. The beta ray spectrometer study by Passell[34] clearly indicated that the lines in question were M conversion electrons of the 16 hour activity superimposed upon electron lines of $Am^{241}$. Thus an experiment was undertaken using the scintillation counter to determine if K electron capture did compete with beta decay in the long-lived $Am^{242}$.

Approximately $10^7$ alpha counts per minute of intensely neutron irradiated americium was obtained from Drs. S. G. Thompson and E. K. Hulet. Mass analysis of this material had given the following isotopic abundances.

| | |
|---|---|
| $Am^{241}$ | 90.4 percent |
| $Am^{242}$ | 1.13 |
| $Am^{243}$ | 8.51 |

$Am^{243}$ is in equilibrium with the 2.3 day $Np^{239}$ which has associated with it prominent plutonium K x-rays. Since the object of this experiment was to try and detect any plutonium K radiation which might be associated with the $Am^{242}$ decay the neptunium was separated from americium by ion exchange techniques. The efficiency of the separation was readily determined on the scintillation counter by the absence of the prominent $Np^{239}$ radiation in the 200-300 kev region.

In order to facilitate detection of the plutonium K x-rays it was necessary to employ absorbers to decrease the intensity of the 59.6 kev gamma ray of $Am^{241}$. Using 700 mg per $cm^2$ of silver absorber, radiation at 105 kev was easily detected on the scintillation counter. The line had the structure expected for a K x-ray multiplet where the high energy components are accentuated by absorption. It was also possible to see

the 75 kev gamma ray of $Am^{243}$ on the high energy side of the $Am^{241}$ gamma ray. A typical spectrum is shown in Figure 3.

Making suitable corrections for absorption, escape peak, and the presence of the $Am^{243}$ gamma ray the ratio of 59.6 kev gamma ray to K x-rays was calculated as

$$\text{"59.6":K x-rays} = 100:1.7 \ .$$

When the sample was in equilibrium with $Np^{239}$ the x-ray peak intensity was increased by about a factor of two.

From the measured mass abundance of $Am^{241}$ and $Am^{242}$, the half-life[40] of $Am^{241}$, and the absolute abundance of the 59.6 kev gamma ray[12] the partial half-life for K electron capture in $Am^{242}$ was calculated as 850 years. The half-life of $Am^{242}$ is about 100 years,[41] indicating a $\beta^-$ to K capture ratio of approximately 9.

Passell has measured the beta energy of $Am^{242}$ as 588 kev. Beta L x-ray coincidence studies by Stephens[35] indicate comparable population of the ground state and the first excited state of $Cm^{242}$ so this energy is very likely the disintegration energy. Closed cycle calculations[24] indicate 660 kev are available for electron capture to the ground state of $Pu^{242}$.

## D. $Ir^{192}$

A very complex array of gamma radiation is associated with the beta decay of the 74 day $Ir^{192}$. Cork and co-workers[42] studying the conversion electron spectrum were the first to detect evidence of electron capture by noting that several transitions had K and L

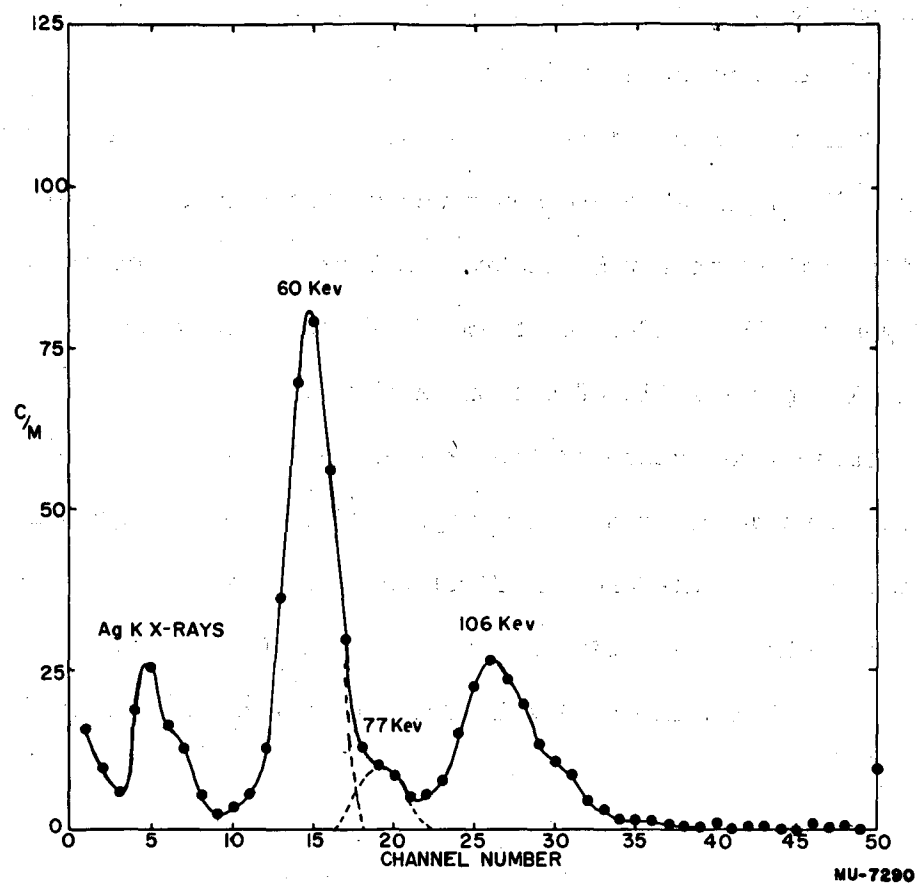


Fig. 3. Scintillation counter spectrum of $Am^{241}$-$Am^{242}$.

conversion lines whose separation was characteristic of osmium rather than platinum. Also several Auger transitions were found which could be ascribed to the electron capture daughter. Seventeen gamma rays were inferred from the electron line data and 14 of these were arranged in a reasonable decay scheme involving 5 excited states of platinum and 3 of osmium.

Muller, et al.[43] using a 2 meter bent crystal spectrometer were able to detect the K series radiation of the electron capture daughter as well as the platinum x-rays from internal conversion following the beta decay. They observed 11 of the lines reported by Cork and because of their very high precision they were able to substantiate many of the cascade-cross over conditions required by the proposed decay scheme. Gamma-gamma coincidence experiments[44,45] are also in agreement with this scheme.

Mims and Halben[46] investigated the possibility of positron emission by $Ir^{192}$ using a gamma-gamma coincidence apparatus to detect annihilation radiation. They were able to set a limit of $8 \times 10^{-3}$ percent for this mode of decay.

In an effort to determine the extent to which $Ir^{192}$ undergoes electron capture a quantity of this activity was obtained from the Oak Ridge National Laboratory. The $K_\alpha$ lines of osmium and platinum were observed with the bent crystal spectrometer but because of poor resolution it was difficult to obtain good relative intensities. Muller reported a value of 1.3 for the platinum to osmium K x-ray ratio and this figure will be used in calculations to follow.

A sample of $Ir^{192}$ was counted in the $4\pi$ counter and then the electromagnetic radiation was examined with the scintillation counter.

The observed lines, all multiplets, and intensities per beta are listed in Table 5 along with part of Muller's data. (The actual $4\pi$ count was corrected for the abundance of electron capture after the branching ratio was determined in order to get the true beta disintegration rate. It was assumed that the counting efficiency for electron capture was 100 percent but an error of a factor of 2 in this figure will only lower the intensity values by about 4 percent.) Since Muller's relative intensities are only stated as being good over a narrow energy range the two sets of data have been combined in column 5 of the Table. The gamma rays below 295 kev were seen with the scintillation spectrometer but intensity measurements were not possible because of the presence of a large Compton continuum.

A K x-ray peak was also observed and its intensity measured as 0.13 photon/$\beta^-$. Correcting this for fluorescence yield[32] one obtains 0.14 K vacancy/$\beta^-$. Using Muller's measured platinum to osmium x-ray ratio there are 0.08 platinum and 0.06 osmium K vacancies per beta. Bashilov and co-workers[47] have determined the abundances of the K conversion lines per beta particle and these are listed in column 6. The sum of the conversion lines of platinum (including several not listed in Table 5) is 0.076, in good agreement with the above value of 0.08. The K vacancies of osmium due to conversion are 0.005, indicating 0.055 K electron capture events per beta disintegration.

Column 7 lists the K conversion coefficients calculated from columns 5 and 6. Adjacent to this is the possible type of transition according to Rose's[48] theoretical K conversion coefficients. The latter values are listed in Table 6 for E1, E2, E3, and M1 transitions.

Table 5

Gamma Rays Following the Decay of $Ir^{192}$

| Scintillation counter energy (kev) | Intensity photon/β | Muller et al.[43] energies (kev) | Muller et al. relative intensities | Abundance Photon/β | K conversion Electron/β[47] | $\alpha_K$ | Transition type |
|---|---|---|---|---|---|---|---|
| | | 295.94 | 380 | 0.30 | 0.016 | 0.053 | E2 |
| 310 | 1.37 | 308.45 | 370 | 0.29 | 0.015 | 0.052 | E2 |
| | | 316.46 | 990 | 0.78 | 0.034 | 0.044 | E2 |
| 470 | 0.49 | 467.98 | 300 | 0.46 | 0.0077 | 0.017 | E2 |
| | | 484.75 | 11 | 0.017 | 0.0005* | 0.029 | E2 |
| | | 588.40 | 11 | 0.10 | 0.00045 | 0.0045 | E1 |
| 600 | 0.27 | 604.53 | 14 | 0.13 | 0.0014 | 0.011 | E2 |
| | | 612.87 | 5 | 0.045 | 0.00056 | 0.012 | E2 |
| | | 205.74 | 75 | -- | 0.0043* | -- | -- |
| -- | -- | 201.31 | 10 | -- | 0.00065* | -- | -- |
| | | 136.33 | 4 | -- | -- | -- | -- |

*Conversion in osmium.

Table 6

Theoretical K Conversion Coefficients of the $Ir^{192}$ Gamma Rays

| Energy | E1 | E2 | E3 | M1 |
|---|---|---|---|---|
| 296 | $2.3 \times 10^{-2}$ | $6.5 \times 10^{-2}$ | 0.18 | 0.34 |
| 308 | $2.0 \times 10^{-2}$ | $5.8 \times 10^{-2}$ | 0.16 | 0.30 |
| 316 | $1.9 \times 10^{-2}$ | $5.6 \times 10^{-2}$ | 0.15 | 0.28 |
| 468 | $8.4 \times 10^{-3}$ | $2.3 \times 10^{-2}$ | $5.8 \times 10^{-2}$ | 0.10 |
| 485* | $6.8 \times 10^{-3}$ | $1.9 \times 10^{-2}$ | $4.7 \times 10^{-2}$ | $7.0 \times 10^{-2}$ |
| 588 | $5.0 \times 10^{-3}$ | $1.4 \times 10^{-2}$ | $3.3 \times 10^{-2}$ | $5.5 \times 10^{-2}$ |
| 605 | $4.8 \times 10^{-3}$ | $1.3 \times 10^{-2}$ | $3.1 \times 10^{-2}$ | $5.2 \times 10^{-2}$ |
| 613 | $4.6 \times 10^{-3}$ | $1.3 \times 10^{-2}$ | $3.0 \times 10^{-2}$ | $5.0 \times 10^{-2}$ |

*Conversion in osmium.

In light of Muller's relative intensities it appears reasonable that the gamma ray sequence in the electron capture daughter as suggested by Cork should be reversed. This would place the first excited state of $Os^{192}$ at 206 kev which is more in line with the trend of these levels in the lighter osmium isotopes.[2]

On the basis of the multipolarities listed in Table 5 the first three levels of $Pt^{192}$ form a 0+-2+-2+ sequence. The next two undoubtedly have even spin and parity, but more data on the lower energy transitions are required. These levels may both be 4+. The state at 1201 kev has odd parity on the basis of the E1 assignment to 588 kev transition.

The modified decay scheme of Cork et al. is shown in Figure 4. Several other gamma rays have been observed,[44,47] but these are of low intensity and are omitted from the decay scheme. Three beta groups of 0.67 Mev (44 percent), 0.54 Mev (40 percent), and 0.24 Mev (16 percent) have been reported by Bashilov. It is thought that the maximum energy group populates the 919 kev level indicating a beta disintegration energy of 1.59 Mev.

It is very interesting to note the analogy between the levels of $Os^{192}$ and $Pt^{192}$. Both the relative energies and the relative intensities of the gamma ray transitions in each daughter are qualitatively the same. Most likely the spin and parity assignments of the detected osmium levels are the same as the corresponding ones in platinum.

The partial half-life for K electron capture is 3.7 years on the basis of a $5.5 \times 10^{-2}$ $K/\beta^-$ branching ratio.

## E. $Ag^{110m}$ and $Ag^{110}$

The radiations associated with the 270 day $Ag^{110m}$ have been the subject of many experimental studies. Siegbahn,[49] and Cork et al.[50] have shown that a highly converted 116 kev gamma ray is the isomeric transition to the 24 second $Ag^{110}$ on the basis of the energy separation of its K and L shell conversion electrons. Miskel,[51] by rapid electrolytic separation, was able to prove the genetic relationship between the two isomers. The main mode of decay of the upper state, however, is to high energy excited levels of $Cd^{110}$ by the emission of two beta groups of 88 and 530 kev maximum energy, respectively. The short lived $Ag^{110}$ which is in equilibrium with its long lived parent

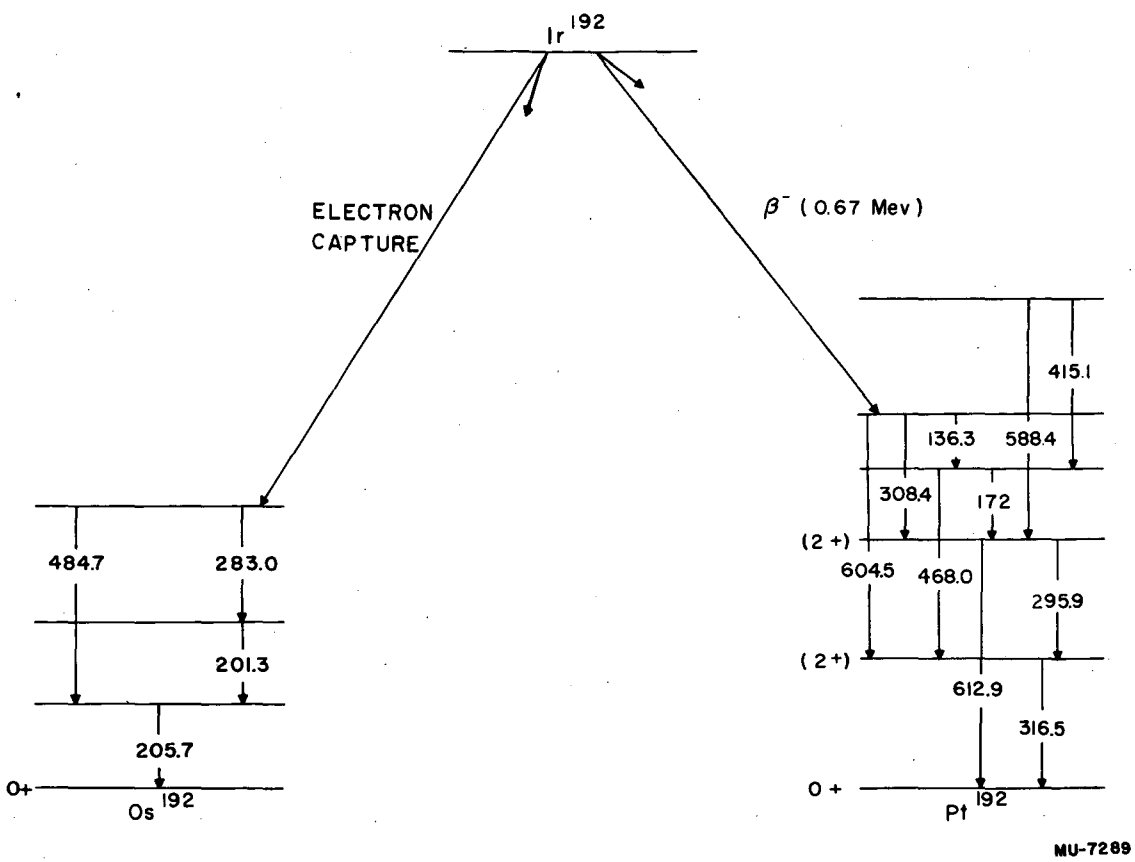


Fig. 4. Decay scheme of $Ir^{192}$.

decays by emission of two beta groups of over 2 Mev energy. From the abundance of these hard transitions Siegbahn estimated approximately 6 percent of the $Ag^{110m}$ undergoes an isomeric transition. By the same method Anton'eva and co-workers[52] arrived at a figure of 2 percent.

The electron capture decay of $Ag^{110m}$ to stable $Pd^{110}$ has not been reported. Deutsch[53] has set a limit of 3 percent for K electron capture on the basis of failure to observe any K radiation. A limit of 0.05 percent has been set on the $\beta^+/\beta^-$ ratio[46] by measurement of annihilation radiation coincidences.

A sample of $Ag^{110m}$ was obtained from the Oak Ridge National Laboratory and mounted for the bent crystal spectrometer. On sweeping the 20 kev region four lines were observed and the energies, and relative intensities of two of the four lines are listed in Table 7. The intensities of the $K\alpha_2$ radiations were not calculated because of poor resolution from the $K\alpha_1$ but were of the expected magnitude. A careful examination of the region of the palladium $K\alpha_1$ yielded negative results and a limit of 15 percent of the silver $K\alpha_1$ intensity can be set for this line. The spectrum is shown in Figure 5.

Table 7

K X-Rays Following the Decay of $Ag^{110m}$

| Line | Transition | Observed energy (kev) | Accepted energy (kev) | Corrected relative intensity |
|---|---|---|---|---|
| Ag $K\alpha_2$ | Ag $K\text{-}L_2$ | 22.06 ± 0.06 | 21.99 | -- |
| Ag $K\alpha_1$ | Ag $K\text{-}L_3$ | 22.19 ± 0.03 | 22.16 | 2.1 |
| Cd $K\alpha_2$ | Cd $K\text{-}L_2$ | 23.05 ± 0.12 | 22.98 | -- |
| Cd $K\alpha_1$ | Cd $K\text{-}L_3$ | 23.21 ± 0.04 | 23.17 | 1 |

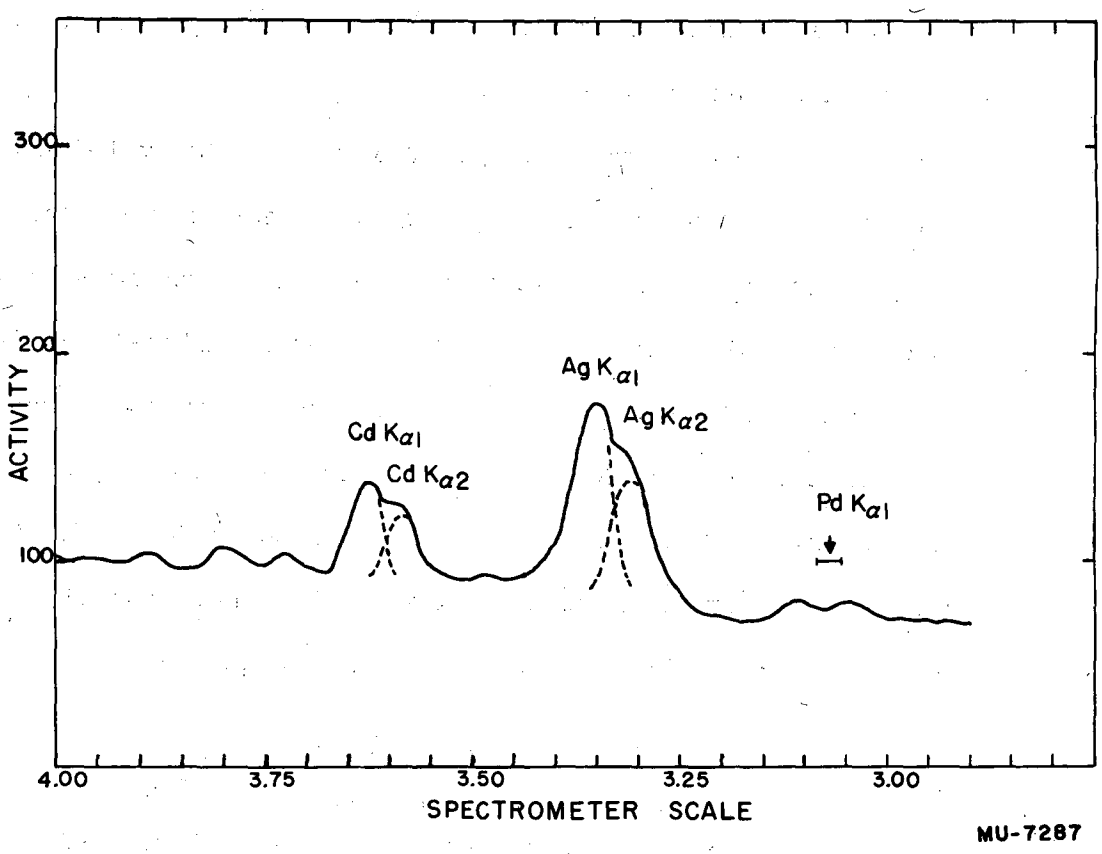


Fig. 5. K x-ray spectrum in the decay of $Ag^{110m}$.

The source of the silver K series radiation is the K conversion of the 116 kev isomeric transition. The cadmium lines are due to K conversion of the numerous gamma rays following the beta decay of $Ag^{110m}$.

In order to determine absolute intensities of the K x-rays a small sample was prepared on a Tygon film, counted in the $4\pi$ counter, and then examined in the calibrated proportional counter. When an absorber of 470 $mg/cm^2$ of beryllium was used to remove the electrons associated with the decay, a broad x-ray peak was observed at 22.5 kev and its corrected intensity was found to be 0.037 photons per disintegration. Using a figure of 0.81 for the K shell fluorescence yield,[32] this datum indicates 0.045 K vacancy per disintegration.

The 116 kev gamma ray is thought to be an M4 transition and as such would be essentially completely converted. From the relative intensities in Table 7 and the proportional counter data one calculates 0.031 silver K vacancy per disintegration. The K/L+M conversion ratio has been reported[49,52] as approximately 1.5, indicating 0.05 isomeric transition per disintegration. This is in excellent agreement with Siegbahn's estimate.

From the intensity limit of the palladium $K_{\alpha_1}$ line and the number of silver K vacancies, a limit of 0.5 percent can be set on the number of K electron capture events per disintegration. This corresponds to a partial half-life for this mode of decay greater than 150 years.

The number of cadmium K vacancies is 0.014 per disintegration. It is difficult, however, to compare this figure with data reported in the literature. The K conversion coefficient of the prominent 656 kev gamma ray has been found to be $2.5 \times 10^{-3}$,[49] and the author has determined

the intensity of this radiation as approximately 100 percent per beta decay by $4\pi$ counting and scintillation spectroscopy. However, corresponding information is not available on other lines. In order to get an estimate of the number of cadmium K vacancies the conversion electron spectrum given in Figure 1 of reference 49 was crudely integrated and the area of the cadmium K lines was compared to the area of the K conversion line of the 116 kev isomeric transition. The result indicates the silver to cadmium conversion ratio was roughly 1.7 and in view of the method of integration this is felt to be in excellent agreement with the measured ratio of 2.1.

Since the isomeric transition occurs in 5 percent of the decays of $Ag^{110m}$ and the limit 0.5 percent has been set on the number of K electron capture events per decay, a limit of 10 percent can be placed on the amount of K electron capture by the 24 second $Ag^{110}$.

## F. $Cs^{134}$

The 2.3 year $Cs^{134}$ is known to decay by beta particle emission to stable $Ba^{134}$. Many gamma rays are associated with the decay and these have recently been interpreted in terms of a new level scheme for $Ba^{134}$.[54]

The search for decay to $Xe^{134}$ has led to negative results. Waggoner, Moon and Roberts[55] in a study of the conversion electron spectrum of $Cs^{134}$ suggested that a 1.002 and 1.135 Mev electron line might be due to gamma ray transitions in $Xe^{134}$, and set a limit of 4 percent on the amount of electron capture in $Cs^{134}$ on the basis of their intensity. Both these transitions have been incorporated in the new level scheme of $Ba^{134}$. Siegbahn and Deutsch[56] tried to detect the

xenon K x-rays in a proportional counter and set a limit of 5 percent on the amount of K electron capture. Mims and Halben[46] using a gamma-gamma coincidence apparatus found that the $\beta^+/\beta^-$ ratio was less than 0.009 percent.

A high specific activity sample of $Cs^{134}$ (20 curies/gram) was obtained from the Oak Ridge National Laboratory. The cesium in chloride solution was dried on a platinum plate, powdered, and mounted in the usual fashion for the bent crystal spectrometer. The capillary contained approximately 10 millicuries of the activity.

Careful sweeps of the spectrum revealed only two weak barium lines. The energies of the radiations are listed in Table 8. Because of the low intensity of these lines and the relatively high background from the abundant gamma rays associated with the decay of $Cs^{134}$ a limit of only 1/3 the intensity of the barium K x-rays could be set for xenon K lines in the spectrum.

Table 8

K X-Rays Following the Decay of $Cs^{134}$

| Line | Transition | Observed energy (kev) | Accepted energy (kev) |
|---|---|---|---|
| Ba $K_{\alpha_2}$ | $K\text{-}L_2$ | 31.80 ± 0.07 | 31.80 |
| Ba $K_{\alpha_1}$ | $K\text{-}L_3$ | 32.16 ± 0.08 | 32.18 |

A small sample of the activity was $4\pi$ counted and then examined with the proportional counter. Use was made of beryllium absorbers to remove the beta particles and two lines were observed which

corresponded to the barium $K_\alpha$ and $K_\beta$ radiation at 32.2 and 36.5 kev, respectively. It was somewhat difficult to correct the observed intensities for counting efficiency because the $K_\beta$ group is above the K absorption edge of the xenon counter gas while the $K_\alpha$ group is below it. By considering only the $K_\alpha$ radiation and employing the $K_\alpha/K_\beta$ ratio for this atomic number as obtained from Compton and Allison[33] the intensity of the K x-rays was calculated as 0.6 percent per disintegration. Broyles[31] has determined the fluorescence yield of the K shell of barium as 0.87 and thus the number of K vacancies is 0.7 percent. As stated above, the limit on the intensity of the xenon x-rays from the decay of $Cs^{134}$ was 1/3 the number of barium K x-rays. Since the fluorescence yields will be roughly the same a limit of 0.2 percent can be set on the occurence of K electron capture. This corresponds to a partial half-life of greater than 1100 years.

The observed number of K vacancies is consistent with the available data on the relative intensities and K conversion coefficients of the prominent gamma rays of $Cs^{134}$, and the decay scheme and beta group abundances of Cork and co-workers.[54,57]

## G. $Tl^{204}$

The beta decay of $Tl^{204}$ has been the subject of many studies but it has only been recently that evidence for electron capture has been obtained. Mitchell and Caird[58] detected electromagnetic radiation of approximately the energy of the K x-rays of mercury in $Tl^{204}$ and upon comparison with a source of mercury x-rays concluded that this isotope did undergo electron capture. Lidofsky, Macklin and Wu,[59]

at the same time, also reported the presence of mercury K radiation on the basis of critical absorption experiments. der Mateosian and Smith[7] incorporated $Tl^{204}$ in a NaI (thallium activated) scintillation crystal and observed the x-ray peak in the spectrum. Under the conditions of the experiment, such a peak would only be expected if the radiation were not im prompt coincidence with the beta group. The latter authors determined a branching ratio of 0.015 ± 0.005 K capture per beta disintegration. In an effort to characterize the observed radiation definitely the following experiments were performed.

Ten millicuries of $Tl^{204}$ was received from the Oak Ridge National Laboratory. Because of the relatively low specific activity (450 millicuries/gram) about 1 millicurie was used for a bent crystal spectrometer sample. Although the intensities were low, two lines were observed and identified. The results are listed in Table 9. Because of resolution difficulties only approximate relative intensities could be determined.

Table 9

Mercury K X-Rays Following Decay of $Tl^{204}$

| Line | Transition | Observed energy (kev) | Accepted energy (kev) | Approximate relative intensity |
|---|---|---|---|---|
| Hg $K\alpha_2$ | $K-L_2$ | 68.79 ± 0.27 | 68.90 | 1 |
| Hg $K\alpha_1$ | $K-L_3$ | 70.93 ± 0.19 | 70.83 | 2 |

Aside from very weak indications of the mercury $K_\beta$ group no other electromagnetic radiation was observed. A search was made of the L x-ray regions with negative results.

Two small samples were prepared and $4\pi$ counted. The K x-rays were observed on the scintillation counter and their intensity was determined as 1.67 ± 0.1 percent relative to the $4\pi$ count (the error is the average deviation in two runs). The 59.6 kev radiation of $Am^{241}$ was employed as both an energy and intensity standard. Taking into account the 95 percent K shell fluorescence yield of mercury[32] the percentage K capture in $Tl^{204}$ is 1.76 ± 0.1.

When these samples were examined with a proportional counter the L x-ray multiplet at 11 kev was easily detected. Upon correction of its intensity for counting efficiency, counter wall absorption, and 470 $mg/cm^2$ of beryllium employed to remove the beta particles, the abundance of the L x-rays was found to be 0.55 percent per disintegration. Employing a weighted L fluorescence yield based upon Kinsey's estimates the total number of L vacancies is 1.8 percent. When the L vacancies due to the K x-rays are taken into account one arrives at the following K/L electron capture ratio in $Tl^{204}$

$$K/L = 3 .$$

der Mateosian and Smith were able to detect the continuous gamma ray spectrum associated with the electron capture decay and found an endpoint of about 250 kev. This energy plus the K absorption edge of mercury gives a disintegration energy of 333 kev.

## H. $Tb^{160}$

$Tb^{160}$ decays by beta particle emission to stable $Dy^{160}$. The transition is very complex and has been reported to involve three beta groups[60] of 0.86 (43 percent), 0.52 (41 percent), and 0.40 (16 percent)

Mev energy, and 12 gamma rays.[61] Of the latter, 5 are quite prominent and their energies are listed in Table 10 below. The other transitions are assumed to be weak because the other workers,[60] as well as the author, failed to detect them.

Table 10

Prominent Gamma Rays Reported in the Decay of $Tb^{160}$

| Line | Energy (kev) |
|---|---|
| $\Gamma_1$ | 86.5 |
| $\Gamma_2$ | 196.4 |
| $\Gamma_3$ | 297.8 |
| $\Gamma_4$ | 876 |
| $\Gamma_5$ | 962 |

A search for evidence of positron emission has yielded negative results[60] and there is no information in the literature concerning possible electron capture to $Gd^{160}$.

Several milligrams of very pure $Tb_4O_7$ powder were irradiated in the Materials Testing Reactor at Arco, Idaho, for an 8 day period. About 1 milligram of this material was packed in a capillary and mounted on the bent crystal spectrometer.

Four K series x-rays of dysprosium were observed and the measured energies and intensities are listed in Table 11.

Table 11

K X-Rays Following the Decay of $Tb^{160}$

| Line | Transition | Observed energy (kev) | Accepted energy (kev) | Corrected relative intensity |
|---|---|---|---|---|
| Dy $K\alpha_2$ | $K-L_2$ | 45.27 ± 0.12 | 45.19 | 50 |
| Dy $K\alpha_1$ | $K-L_3$ | 45.98 ± 0.08 | 45.98 | 100 |
| Dy $K\beta_1\beta_3$ | $K-M_2M_3$ | 52.13 ± 0.15 | 52.09-52.18 | 35 |
| Dy $K\beta_2$ | $K-N_2N_3$ | 53.46 ± 0.33 | 53.49 | 12 |

The relative intensities of the $K\beta_1\beta_3$ and $K\beta_2$ are somewhat uncertain because of the self absorption correction. According to the paper by Hill, Church and Mihelich[62] the K edge of terbium is at 52.00 kev and thus these two lines are subject to considerable absorption. An attempt has been made to correct for this effect but the magnitude of the absorption jump at the K edge could only be estimated.

It is of interest to compare the K x-ray intensities with those obtained by the interpolation of the data in Compton and Allison.[33] At Z = 66 the latter indicate the following ratios

$$K\alpha_1 : K\alpha_2 : K\beta_1\beta_3 : K\beta_2 = 100:50:33:12$$

which is in remarkable agreement with the data in Table 11. Since no self absorption correction has been applied to Compton and Allison's data this correction is either very small in electron bombardment x-ray sources or the agreement here is only fortuitous.

A careful search of the region of the gadolinium $K\alpha_1$ line indicated no detectable quantity of this radiation. There was evidence,

however, of terbium K series lines presumably due to fluorescence excitation in the sample. The maximum intensity of the gadolinium $K\alpha_1$ line is 4 percent of the dysprosium $K\alpha_1$.

Two gamma rays were observed on the bent crystal spectrometer. A relatively weak 48.75 ± 0.22 kev line was detected in the early sweeps of the K x-ray region. It was noticed that this line decayed relative to the x-rays with a half-life of 4 to 10 days. Cork et al.[63] have observed conversion electrons from a 49.0 kev gamma ray in the decay of the 6.8 day $Tb^{161}$ and this isotope, formed by two successive neutron captures in the high flux reactor, is presumably the source of the observed radiation. The x-ray intensities listed in Table 12 were determined after the decay of this second order product.

Table 12

Energies and Abundances of the Radiations Observed in the Decay of $Tb^{160}$

| Line | Energy (kev) | Intensity (photons/$\beta^-$) |
|---|---|---|
| K x-rays | 43 | 0.16 |
| $\Gamma_1$ | 87 | 0.12 |
| $\Gamma_2$ | 195 | 0.05 |
| $\Gamma_3$ | 290 | 0.14 |
| $\Gamma_4$ | 890 | 0.34 |
| $\Gamma_5$ | 960 | 0.34 |
| $\Gamma_6$ | 1180 | 0.25 |
| $\Gamma_7$ | 1270 | 0.11 |

A gamma ray of 86.91 ± 0.29 kev was observed in an abundance of 132 relative to the dysprosium $K_{\alpha_1}$ and is $\Gamma_1$ in Table 10. No other lines could be detected above this energy on the bent crystal spectrometer.

If one assumes that $\Gamma_1$ is the main source of K x-rays a K conversion coefficient can be calculated. The assumption appears to be substantiated by the magnitude of the K conversion electron peak of $\Gamma_1$ relative to that of other gamma rays as reported in the literature.[60] Employing a 91 percent fluorescence yield[32] to obtain the number of K vacancies leads to a value of $\alpha_K = 1.7$ which can be compared to 1.65 ± 0.2 determined by McGowan[64] and 1.5 calculated from scintillation counter data to be presented below.

A small sample of $Tb^{160}$ was $4\pi$ counted and then examined on the scintillation counter. The observed lines and their approximate intensities per beta particle are listed in Table 12. The two highest energy lines have not been previously reported, but were readily detected on the scintillation counter. A portion of the high energy spectrum is shown in Figure 6.

Preliminary gamma-gamma coincidence studies indicated that at least one of the two new lines is in coincidence with $\Gamma_1$. No other gamma radiation appears to follow or precede these hard transitions. The results on the other gamma rays are not as yet too clear. $\Gamma_4$ is in coincidence with $\Gamma_1$, while $\Gamma_5$ is not. It appears as if both $\Gamma_4$ and $\Gamma_5$ are in coincidence with at least one of the other low energy lines. More work is definitely needed on this decay scheme, but from the gamma ray intensities it appears that all the beta particles populate levels at least 960 kev above the ground state of $Dy^{160}$.

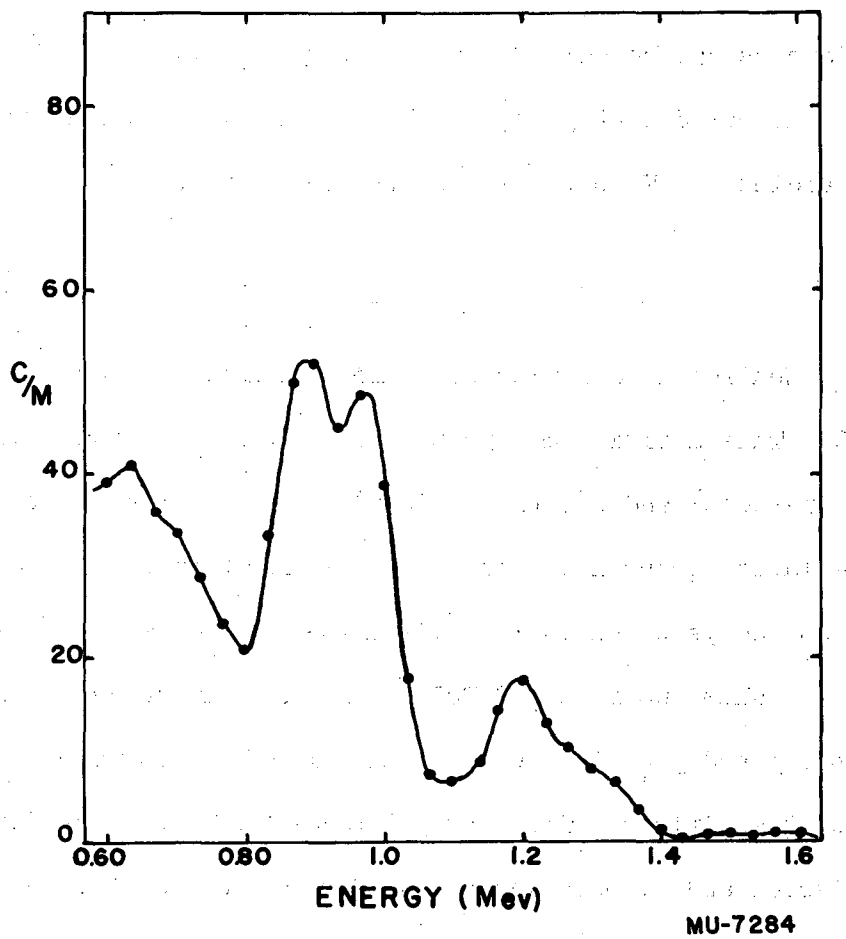


Fig. 6. High energy gamma rays in the decay of $Tb^{160}$.

From the measured abundance of the dysprosium K x-rays and the limit of 4 percent of this figure as the maximum intensity of gadolinium K radiation one obtains an upper limit of 0.0064 electron capture x-ray per beta particle. Correcting this for fluorescence yield the abundance of K capture in $Tb^{160}$ is less than 0.7 percent, indicating a partial half-life of greater than 30 years. The number of dysprosium K vacancies can be calculated from the absolute x-ray intensities and is 0.18 per disintegration. If $\Gamma_1$ is the major source of these vacancies its K conversion coefficient is 1.5.

## I. $Tm^{170}$

Neutron irradiation of thulium yields the 129 day isotope of mass 170. This activity decays by emission of two beta groups of 968 (76 percent) and 884 (24 percent) kev maximum energy, respectively.[65] The latter group leads to an 84 kev level in the ytterbium daughter which decays by gamma ray emission and internal conversion.

Graham, Wolfson and Bell[65] have sought evidence for electron capture and positron decay to stable $Er^{170}$ but were unable to detect such transitions. They set a limit of 0.01 percent for positron emission and by beta-gamma coincidence studies they were also able to set a limit of 0.3 percent for K electron capture.

A sample of $Tm_2O_3$ that had been neutron irradiated in the Materials Testing Reactor at Arco, Idaho, was made available by Mr. Peter Gray and Dr. S. G. Thompson. The activity had been chemically purified by ion exchange techniques. The thulium was precipitated as the hydroxide and mounted as previously described on the bent crystal spectrometer.

Four x-ray lines were observed and their energies and relative intensities are listed in Table 13. Again it must be said that the self absorption correction for the $K_{\beta_1\beta_3}$ and $K_{\beta_2}$ lines is uncertain because of their proximity to the thulium K edge. Also, the possibility exists that a small part of the observed ytterbium $K_{\alpha_2}$ peak is due to the thulium $K_{\alpha_1}$ caused by fluorescence excitation. A very small peak which could be the thulium $K_{\alpha_2}$ was observed. This might explain why the ytterbium $K_{\alpha_1}/K_{\alpha_2}$ ratio is somewhat smaller than its usual value of 2.

Table 13

K X-Rays Following the Decay of $Tm^{170}$

| Line | Transition | Observed energy (kev) | Accepted energy (kev) | Corrected relative intensity |
|---|---|---|---|---|
| Yb $K_{\alpha_2}$ | $K\text{-}L_2$ | 51.17 ± 0.20 | 51.34 | 60 |
| Yb $K_{\alpha_1}$ | $K\text{-}L_3$ | 52.24 ± 0.11 | 52.36 | 100 |
| Yb $K_{\beta_1\beta_3}$ | $K\text{-}M_2M_3$ | 59.38 ± 0.21 | 59.15-59.38 | 40 |
| Yb $K_{\beta_2}$ | $K\text{-}N_2N_3$ | 61.25 ± 0.6 | 60.88 | 15 |

A careful examination was made of the region of the erbium $K_{\alpha_1}$ but nothing above background variations was found. The maximum intensity of this radiation that could have escaped detection is estimated as about 5 percent of the ytterbium $K_{\alpha_1}$.

One line was observed on the bent crystal spectrometer which could be ascribed to a gamma ray and it is shown with the K x-ray spectrum in Figure 7. Its measured energy was 84.12 ± 0.27 kev which is in excellent

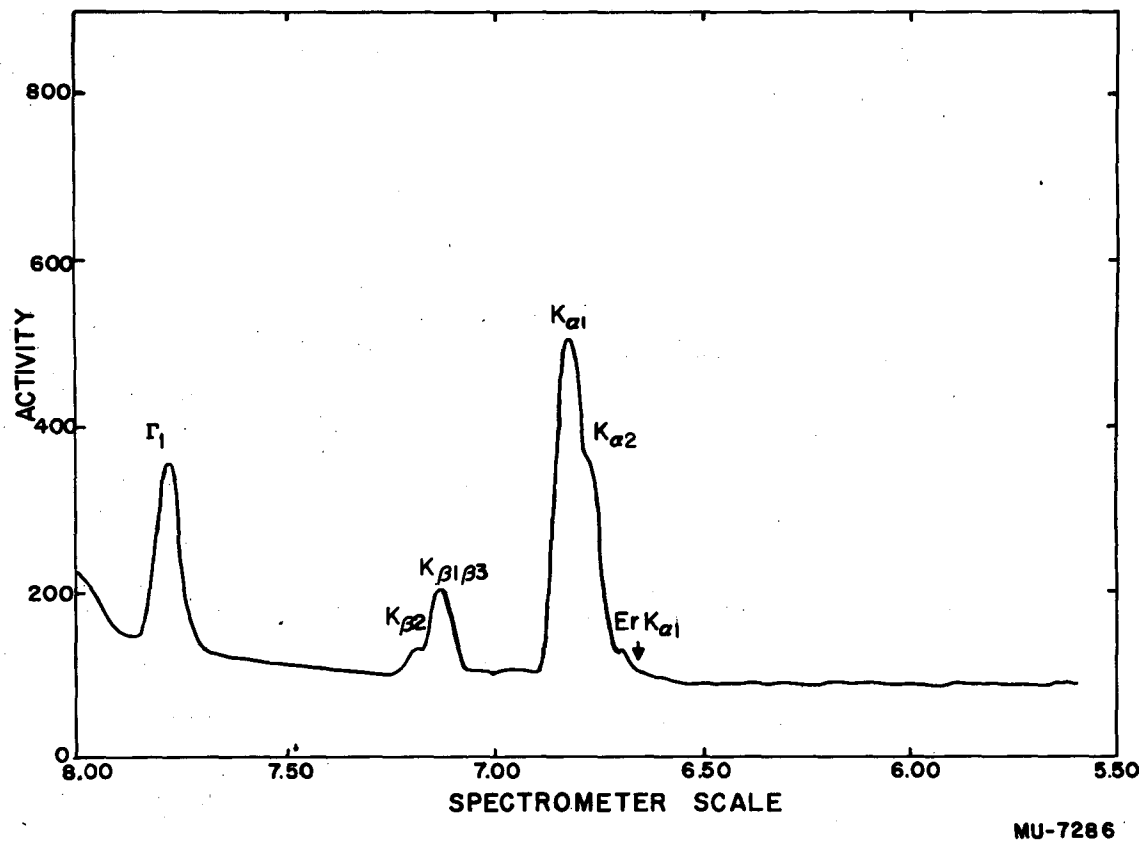


Fig. 7. K x-rays and gamma rays in the decay of $Tm^{170}$.

agreement with the conversion electron value of 84.1.[65] The intensity of this line was 210 relative to the ytterbium $K_{\alpha_1}$. Since the internal conversion of this gamma ray is the only source of K x-rays its K conversion coefficient will be given by

$$\alpha_K = \frac{\Sigma(\text{K x-ray intensities})}{(f_K)(\gamma \text{ intensity})}$$

where $f_K$ is the K fluorescence yield of ytterbium and can be estimated as 0.92.[32] The result is $\alpha_K = 1.6$ which can be compared to the value of 1.5 determined by McGowan,[64] 1.6 by Graham et al.,[65] and 1.5 determined on the scintillation counter as described below.

A small sample of $Tm^{170}$ was $4\pi$ counted and examined with the scintillation counter. Only the x-ray peak at 52 kev and the gamma peak at 86 kev were observed. By integrating these two lines and correcting for the escape peaks the intensities were found to be 3.7 percent and 2.6 percent, respectively, relative to the $4\pi$ count. Taking into account the fluorescence yield the K conversion coefficient of the gamma ray is 1.5. The intensity of the gamma ray can be compared to the value of 3 percent found by Graham, Wolfson and Bell.

From the absolute abundance of the K x-rays and the limit set on the erbium $K_{\alpha_1}$ relative to the ytterbium $K_{\alpha_1}$, the abundance of K electron capture is less than 0.2 percent per beta disintegration. This corresponds to a partial half-life of greater than 170 years.

## J. $Sb^{124}$

The 60 day $Sb^{124}$ decays by beta emission to several excited states of stable $Te^{124}$. This activity has received considerable

study[57] and a level scheme of the tellurium daughter has been proposed. Recently reported coincidence studies by Langer and Starner[66] have verified certain aspects of this scheme.

Langer, Moffat and Price[67] attempted to find evidence for K electron capture to $Sn^{124}$ by studying the Auger electron spectrum but with no success. These authors also found no evidence for positron emission.

A sample of $Sb^{124}$ was received from the Oak Ridge National Laboratory and was mounted for the bent crystal spectrometer. Unfortunately no lines could be detected because of low specific activity.

A small portion of the sample was then counted in the $4\pi$ counter and examined with the proportional counter. When 1 gram/$cm^2$ of beryllium was used to absorb the beta particles a small $K_\alpha$ peak was observed at 27 kev. There was also an indication of the $K_\beta$ group on the high energy side of this line. These radiations are undoubtedly the x-rays of tellurium from gamma ray conversion. The expected energy of the tellurium $K_\alpha$ group is 27.5 kev while that of the tin $K_\alpha$ group is 25.2 kev.

Integration of the observed peak and correction for absorber and counting efficiency yielded an abundance of K x-rays of about 0.4 percent relative to the $4\pi$ count. The K fluorescence yield at this Z is approximately 0.85,[32] and applying this factor a K vacancy abundance of 0.5 percent was calculated. Hutchinson and Wiedenbeck[68] have measured the intensity of the K conversion line of the prominent 603 kev gamma ray associated with the decay of $Sb^{124}$ and found 0.0036 K conversion electron per beta particle. Tomlinson et al.[69]

have found the other K lines to be about 16 percent of this, giving a total of approximately 0.4 K vacancy of tellurium per beta particle. Thus a limit of 0.1 percent can be set for K electron capture in $Sb^{124}$. This corresponds to a partial half-life of greater than 170 years.

K. $\underline{Sb^{122}}$

The radiations associated with the 2.8 day $Sb^{122}$ have been studied by Glaubman and Metzger.[70] These authors have reported three beta groups; 1.94 (26 percent), 1.36 (69 percent), and 0.73 (5 percent) Mev. They have also observed gamma rays of 0.563 (73 percent), 0.692 (3.5 percent), and 1.25 (0.8 percent) Mev. Gamma-gamma coincidence studies showed the first two gamma rays were in cascade. The weak 1.25 Mev radiation was assumed to be the cross-over transition. These data were interpreted in terms of two excited states in $Te^{122}$, 0.563 and 1.25 Mev above the ground state, and beta transitions to these levels and the ground state. There were also indications of a gamma ray at 1.15 Mev in 1 percent abundance. No mention was made of possible electron capture to $Sn^{122}$.

The 0.563 Mev transition is the dominant feature in the gamma ray spectrum according to Glaubman and Metzger and has a measured K conversion coefficient of 0.0049. From the measured intensity of the gamma ray there should be 0.0036 K vacancy per beta particle. Since the other transitions are both lower in abundance and higher in energy one might expect little additional K conversion. It then seemed reasonable to expect no more than approximately 0.4 percent K

vacancy due to conversion in tellurium or 0.34 percent K x-ray from this source.

Neutron irradiated antimony metal was obtained from the Oak Ridge National Laboratory. Since the specific activity was quite low it did not seem feasible to examine the x-rays on the bent crystal spectrometer. The antimony was dissolved in $HNO_3 + Br_2$ solution and several small samples were taken for $4\pi$ counting. On examination with the scintillation counter a fairly intense x-ray peak was observed at about 28 kev. Integration of this peak yielded an intensity of 2.6 percent relative to the $4\pi$ count. A more careful examination with the proportional counter showed a broad line at 24.5 kev whose integrated area corresponded to 2.5 percent of the $4\pi$ count when corrected for counting efficiency and beryllium absorber.

On intensity grounds alone it seemed obvious that K electron capture was competing with the beta decay to the extent of several percent. Also, the proportional counter measurement indicated the x-ray energy was closer to the tin $K_\alpha$ radiation (25.2 kev) than to the tellurium $K_\alpha$ lines (27.5 kev). To further check the nature of the radiation critical absorption experiments were performed on the scintillation spectrometer using palladium, silver and cadmium foils. The K absorption edge of these elements as well and the K series line energies of tin, antimony and tellurium are listed in Table 14.

It was indeed found that palladium behaved as a critical absorber while silver and cadmium did not. The presence of the excited K radiation from the absorbers did, however, prevent the experiment from being as clear cut as desired. Absorption curves taken on the 50 channel gamma ray analyzer are shown in Figure 8.

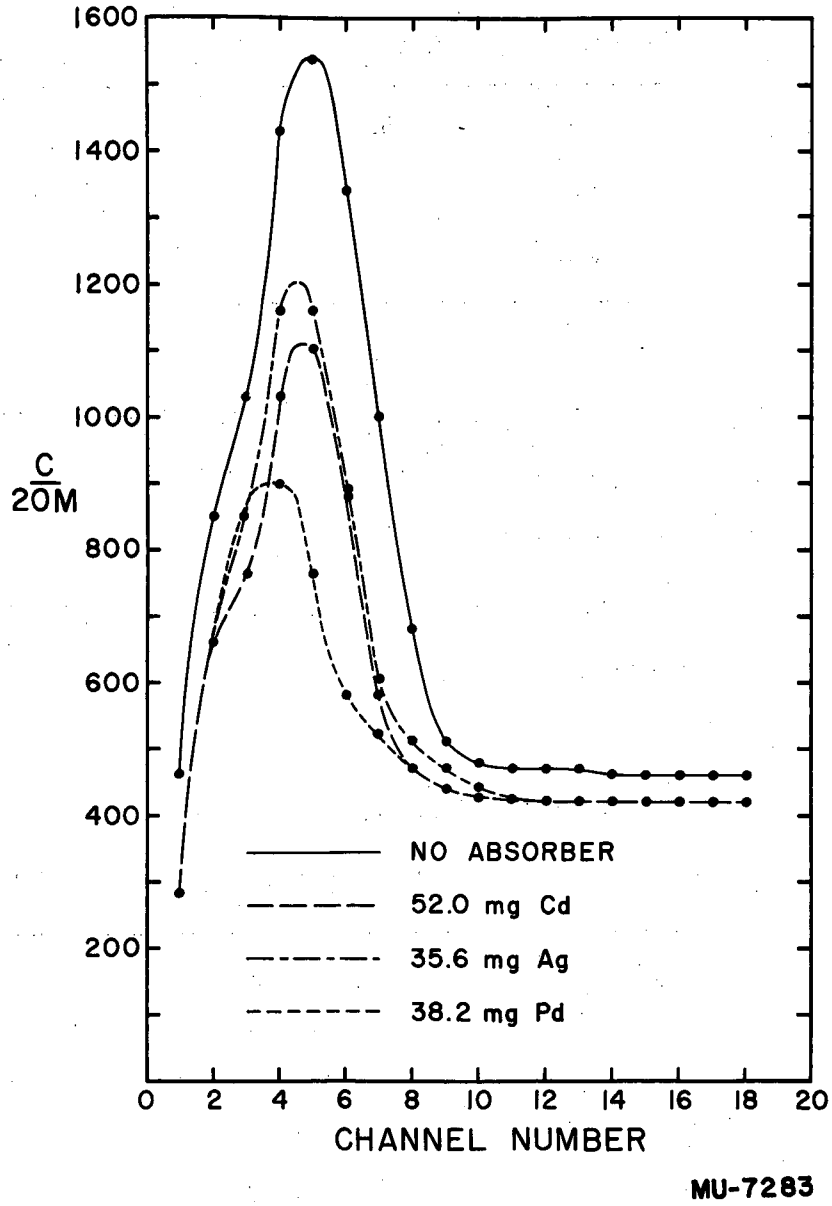


Fig. 8. Critical absorption of the K x-rays in the decay of $Sb^{122}$.

Table 14

Critical Absorption Data for the K X-Rays Following the Decay of $Sb^{122}$

| Absorbers | | | | | | |
|---|---|---|---|---|---|---|
| Element | K edge[62] | Element | $K_{\alpha_2}$ | $K_{\alpha_1}$ | $K_{\beta_1\beta_3}$ | $K_{\beta_2}$ |
| Pd | 24.36 | Sn | 25.04 | 25.27 | 28.46 | 29.11 |
| Ag | 25.54 | Sb | 26.11 | 26.36 | 29.72 | 30.34 |
| Cd | 26.71 | Te | 27.20 | 27.47 | 30.99 | 31.69 |

After these experiments were performed a sample of enriched antimony metal (99.4 percent $Sb^{121}$) was irradiated in the Materials Testing Reactor, Arco, Idaho, for about 32 hours. The high specific activity $Sb^{122}$ thus formed was examined with the bent crystal spectrometer and the observed lines are listed in Table 15.

Table 15

K X-Rays Following the Decay of $Sb^{122}$

| Line | Observed energy (kev) | Accepted energy (kev) |
|---|---|---|
| Sn $K_{\alpha_2}$ | 25.08 ± 0.05 | 25.05 |
| Sn $K_{\alpha_1}$ | 25.28 ± 0.02 | 25.27 |
| Sn $K_{\beta_1\beta_3}$ | 28.48 ± 0.03 | 28.44-28.49 |
| Sn $K_{\beta_2}$ | 29.09 ± 0.06 | 29.11 |
| Sb $K_{\alpha_2}$ | 26.14 ± 0.05 | 26.11 |
| Sb $K_{\alpha_1}$ | 26.39 ± 0.02 | 26.36 |
| Sb $K_{\beta_1\beta_3}$ | 29.74 ± 0.07 | 29.72 |
| Te $K_{\alpha_2}$ | 27.26 ± 0.05 | 27.20 |
| Te $K_{\alpha_1}$ | 27.52 ± 0.03 | 27.47 |
| Te $K_{\beta_1\beta_3}$ | 30.96 ± 0.07 | 30.99 |

The tin x-rays arise from electron capture in $Sb^{122}$ while the tellurium lines are due to K shell conversion of gamma radiation following the beta decay of this nuclide. The presence of antimony K radiation can be ascribed to fluorescence excitation in the sample. A portion of the spectrum is shown in Figure 9.

The ratio of tin to tellurium K x-rays in 6:1. This figure coupled with the measured total abundance of K x-rays mentioned above leads to a value of 0.022 tin x-rays and 0.0036 tellurium x-rays per disintegration. From these intensities and the fluorescence yield[32] of 0.84 at this Z the number of tin and tellurium K vacancies per disintegration are 0.026 and 0.0043, respectively. The former figure corresponds to a partial half-life of approximately 110 days for K electron capture.

It is possible that the unassigned 1.15 Mev gamma ray found by Glaubman and Metzger and not placed in their decay scheme may be associated with the electron capture decay. From the correlation of the first excited states of even-even nuclei[2] one might predict an energy of about 1.3 Mev for the lowest excited level in $Sn^{122}$. This value is based on a two point linear extrapolation ($Sn^{116}$ and $Sn^{120}$) and could be in error by a considerable amount. If the 1.15 Mev level is in $Sn^{122}$, 0.01 K electron capture events populate this state on the basis of the reported intensity of the gamma ray and the remaining 0.016 event lead to the ground state of $Sn^{122}$.

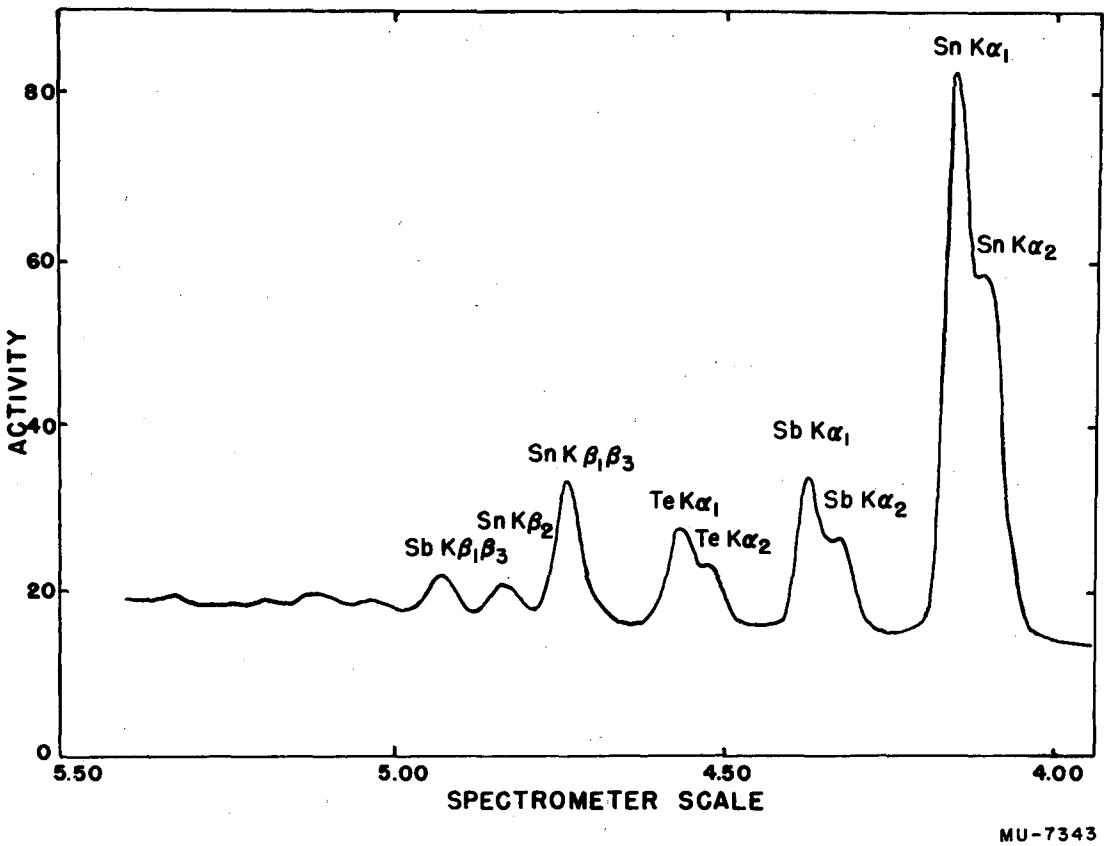


Fig. 9. K x-ray spectrum in the decay of $Sb^{122}$.

## L. $Np^{236}$

The decay of the 22 hour $Np^{236}$ has been studied by Orth and O'Kelley.[71] These authors found evidence of electron capture from Auger electron intensities and x-ray abundance determined by absorption techniques. A rather intense electron line was observed and assigned as the K conversion peak of a 150 kev gamma ray. Very weak indications of the L line were also found. Two beta groups of maximum energy 0.51 and 0.36 Mev were detected and it was concluded that the gamma ray followed the soft beta component and the electron capture populated the ground state of the $U^{236}$ daughter. The electron capture to beta emission branching ratio was estimated as 2.

Approximately 500 milligrams of $U^{235}$ (99+ percent pure) in the form of a metal foil of 21 milligrams/$cm^2$ surface density was bombarded for a total of 940 microampere hours of deuterons on the 60-inch cyclotron. Absorbers were employed to give a deuteron energy of 12 Mev. The $Np^{236}$ was formed by the following reaction

$$U^{235}(d,n)Np^{236}.$$

Because of its long half-life, the activity level of $Np^{235}$ formed by the (d,2n) reaction is expected to be small compared to the desired product.

The target was dissolved in HCl with a few drops of $H_2O_2$, and small amounts of formic acid and KI were then added to reduce neptunium to the quadrivalent state. After suitable adjustment of the HCl concentration, the solution was put on an ion exchange column. Under the experimental conditions most fission products passed through the resin while uranium and neptunium remained. The latter

were then eluted with dilute HCl and treated with $H_2O_2$ to destroy the $I^-$ ion. The solution was boiled with formic acid to bring the neptunium back to the (IV) state and the HCl concentration was adjusted to 2 M. The neptunium was then extracted into 0.4 M TTA in benzene, washed twice with 2 M HCl and back extracted into 8 M HCl. The extraction cycle was repeated and the neptunium was again contacted with the ion exchange resin giving the purified product.

Because of an unusually low cross section[72] there was insufficient activity for a study of the x-ray spectrum on the bent crystal spectrometer. Several small samples were prepared and examined on the scintillation counter. L x-rays at about 17 kev and K x-rays at 100 kev were observed as well as many low intensity lines up to 1.57 Mev. The latter radiation has been observed in the decay of $Np^{234}$,[73] and after following the decay of the K x-ray peak concluded that an appreciable amount of this activity was present. The $Np^{234}$ was not too much of a handicap because a suitable correction could readily be made for its presence.

A thorough examination of the 0-200 kev region revealed no indications of the reported 150 kev gamma ray. Passell[74] studying the beta spectrum, found that the line originally assigned to the K conversion of the 150 kev transition was actually a multiplet and could be interpreted in terms of two gamma rays of about 44 kev. By analogy to the decay of $Am^{242m}$ it was assumed that these two gamma rays represented the first excited states of the $Pu^{236}$ and $U^{236}$ daughters. If the lower energy line is due to conversion in plutonium the gamma ray

energies are 43.4 and 44.2 kev, respectively. The latter is in good agreement with the energy of 44 kev determined for the first excited state of $U^{236}$ from the alpha particle fine structure of $Pu^{240}$.[75] The electron intensities of these two transitions were found to be comparable.

In order to determine the K capture/beta branching ratio the alpha particle growth of $Pu^{236}$ was observed in the same sample used for the scintillation counter experiments. The growth curve gave a parent half-life of about 24 hours and alpha pulse analysis indicated only the 5.75 Mev alpha particles of $Pu^{236}$ were present. From the alpha particle disintegration rate after the $Np^{236}$ had decayed and the absolute abundance of the K x-rays at the beginning of the experiment a branching ratio of $K/\beta^- = 43 \pm 5\%/57 \pm 5\%$ was calculated. The K x-rays have been corrected for the presence of $Np^{234}$ and an estimated 97 percent fluorescence yield.

The intensity of the L x-rays was 0.6 relative to the K series lines. Correcting this figure for the mean L shell fluorescence yield of about 50 percent[27] the number of L vacancies is 1.2 per K vacancy. Since each K vacancy gives rise to roughly 0.72 L vacancy at this $Z^{33}$ there are at maximum 0.48 L vacancy from conversion per K capture event or 0.21 vacancy per disintegration. This figure is an upper limit on the amount of L conversion because L electron capture has been ignored. Since no abundant gamma rays which could be ascribed to the $Np^{236}$ were observed on the scintillation counter it appears safe to conclude approximately 80 percent of both the beta decay and K electron capture lead to the ground states of the respective daughters,

the remaining transitions populating the low lying first excited states. The decay scheme is shown in Figure 10.

## IV. DISCUSSION OF RESULTS

As mentioned earlier, the extent to which electron capture competes with $\beta^-$ decay in a given nuclide depends upon the respective decay energies of the two types of transitions and the spin and parity changes that occur. Decay to similar states of the two even-even daughters of a shielded nucleus should thus depend mainly upon the energy; the matrix element for the two transitions being similar. It has recently been pointed out by de-Shalit and Goldhaber,[76] however, that unusual decay rates may occur in regions of the closed shells because of mixing of the nuclear level configuration.

The ft values or comparative half-lives of the two transitions is a convenient criterion for comparing the respective rates of decay to similar daughter levels. This product should be independent of energy and, except for closed shell effects, be approximately the same for both electron capture and beta decay. The ft values employed below were determined using the nomogram of Moszkowski,[77] and the compilation of King, Dismuke, and Way.[78] The $f_1$ functions for "$\Delta I = 2$, yes" transitions were calculated from Marshak's equations[79] and the paper by Davidson.[80] In order to compare log $f_1t$ values for electron capture and beta decay it is necessary to divide Marshak's $B_c^{(1)}$ factor by 24 to obtain $f_{1_{EC}}$.

Closed cycle calculations[24] indicate 0.12 Mev is available for electron capture in $Np^{238}$. The beta decay studies show that the first

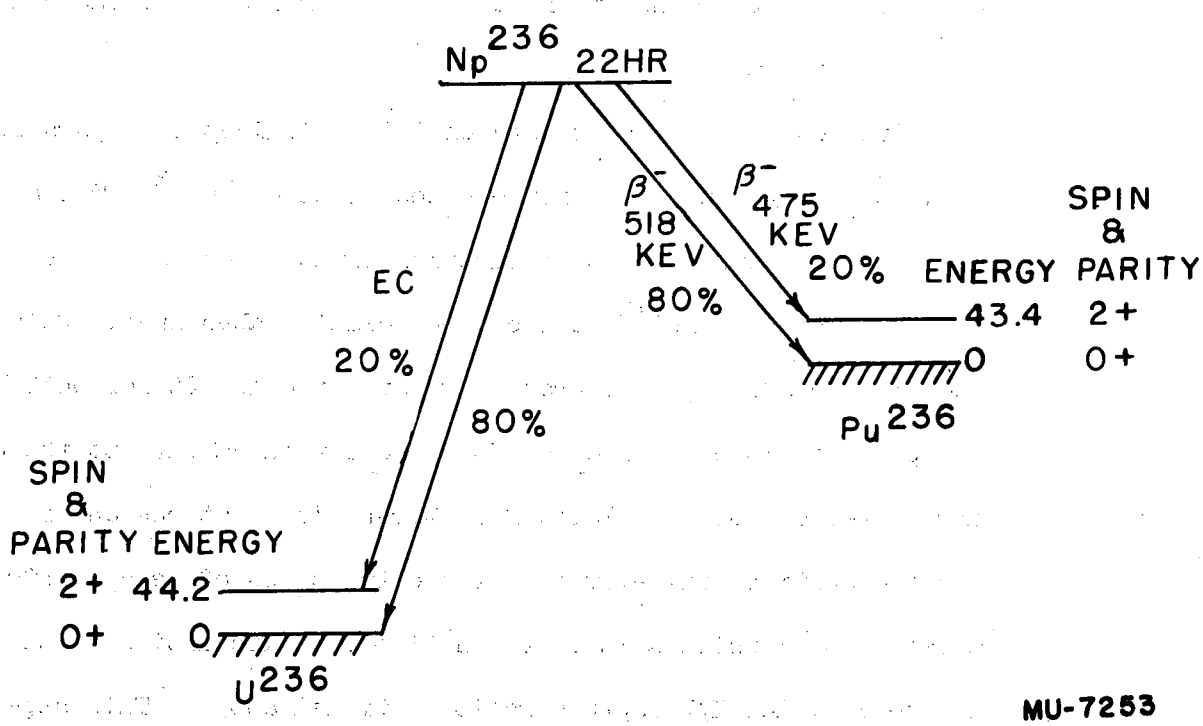


Fig. 10. Decay scheme of $Np^{236}$.

excited state of $Pu^{238}$ is populated by a transition whose log ft is 8.4. If this transition is of the "$\Delta I$ = 2,yes" type then the log $f_1t$ is 8.0. Electron capture to the first excited state of $U^{238}$ would involve a decay energy of roughly 0.08 Mev and only L or higher shells could be involved. If the electron capture were to have a log $f_1t$ of 8.0 the partial half-life for L capture would be approximately 800 years. This would correspond to an electron capture/beta branching ratio of $<10^{-3}$ percent and is consistent with the much larger limits of 4 and 2 percent for $L_1$ and $L_2$ capture determined from the bent crystal spectrometer data.

Stephens' coincidence data on $Am^{242m}$ indicates comparable beta decay to the ground and first excited states of $Cm^{242}$. The analysis of the x-ray data shows that electron capture populates the corresponding states of $Pu^{242}$ to the same relative abundance. Using the measured beta energy and branching ratio the log ft for both beta groups is about 6.9. The log ft for electron capture to the corresponding states can be calculated from Marshak's equations and is 7.2. Thus, the comparative half-lives of these two types of decay differ by only a factor of 2 using allowed f functions. A disturbing point, however, is the very low K/L capture ratio mentioned above. This aspect requires further study.

The long lived $Am^{242}$ beta decay has a log ft of 11.4 to be compared with the value of about 11.8 for K electron capture calculated from the measured partial half-life and the closed cycle decay energy. The former value takes into account the equal branching to the ground and first excited states of $Cm^{242}$. At present the relative population of

the electron capture daughter levels is not known. These ft values may indicate a second forbidden transition but not one of the type "$\Delta I = 3$, no." The log $f_2t$ of the beta decay for such a transition would be about 9 and Davidson[80] has shown that a "$\Delta I = 3$,no" decay should have a log $f_2t$ of approximately 12.

The spins of the two $Am^{242}$ isomers are still unknown. From the data accumulated in this laboratory it appears as if both activities decay in a very similar fashion except for half-life. Since the trend of excited states in this region indicates the presence of several higher spin levels that could energetically be reached by either isomer and radiations from these are not observed, it is thought that both activities have relatively low spin. However, the long isomeric transition must be explained. It is tempting to suggest a 0- and 0+ assignment to the 16 hour and long lived activity, respectively, but this brings on difficulties with the calculated ft values. At present the problem of spin and parity assignments must be left unresolved.

Since no information is available concerning the $Ir^{192}$-$Os^{192}$ mass difference it is not possible to calculate a log ft for electron capture in $Ir^{192}$. According to Bashilov et al.[47] the most energetic beta group of $Ir^{192}$ populates the fourth excited state of the platinum daughter. The electron capture leads to the third and probably lower excited states of osmium. Evaluation of these facts will require additional data.

Precision mass spectrometric measurements by Halsted[81] indicate $Pd^{110}$ is heavier than $Cd^{110}$ by 1.08 ± 0.26 millimass units or

1.00 ± 0.24 Mev. Siegbahn[49] has reported the beta disintegration energy of $Ag^{110m}$ as 3.02 Mev and this coupled with the above mass difference indicates 2 Mev is available for electron capture and positron emission. One can understand the absence of these types of decay in terms of the 5- spin assignment of $Ag^{110m}$.[82] In the decay to cadmium the lowest level populated directly by beta particles is 2.48 Mev above the ground state. The excited states of $Pd^{110}$ are not known but are probably not too much lower in energy than the corresponding cadmium levels. Thus electron capture would undoubtedly involve a more highly forbidden transition than the beta decay because a lower spin state would have to be populated. Positron emission may only be energetically possible to the ground and first excited states of $Pd^{110}$. A transition to the latter, whose position can be estimated as about 0.5 Mev above the ground state of $Pd^{110}$ from G. Scharff-Goldhaber's paper,[2] would be third forbidden and could hardly compete with the beta decay.

The 24 second $Ag^{110}$ ground state presumably has a spin of 1+ [49] and is unstable with respect to $Pd^{110}$ by 1.9 Mev. From the bent crystal spectrometer data a limit of 10 percent can be set on the abundance of electron capture in the decay of the short lived isomer. Likewise the positron abundance is less than 1 percent. Both beta groups of $Ag^{110}$, populating the ground and first excited states, respectively, of $Cd^{110}$ are allowed transitions. The former (2.82 Mev) has a log ft of 5.1 and the latter (2.24 Mev) has a log ft of 4.4. K electron capture to the ground state of $Pd^{110}$ with a log ft of 5.1 would have a partial half-life of approximately 5 hours, or an abundance

of 0.13 percent per beta decay. From the tables of Feenberg and Trigg[83] one can estimate the positron competition as about 10 percent of the electron capture or 0.01 percent per beta decay. Both these values are well below the set limits. K electron capture to the first excited state of $Pd^{110}$ with a log ft of 4.4 would have a partial half-life of about 2 hours or an abundance of 0.3 percent per beta decay. Positron competition to the first excited state is only expected to be about 1 percent of the electron capture decay or the order of 0.003 percent. These estimates are also within the experimental limits.

There is no available data concerning the $Cs^{134}$-$Xe^{134}$ mass difference. K. Way,[84] however, has compiled beta systematics curves from which one can estimate this difference as approximately 1 Mev. Goldhaber and Hill[82] have suggested that the spin of $Cs^{134}$ is 4. The first excited state (2+) of $Xe^{134}$ is expected to be about 1 Mev[2] above the ground state of $Xe^{134}$ and thus one would not expect any electron capture to this level on energy grounds alone. Electron capture or positron decay (if energetically possible) to the ground state of $Xe^{134}$ would involve a spin change of 4 and would certainly have a very long partial half-life. Qualitatively, therefore, neither electron capture nor positron emission can compete with the beta decay of $Cs^{134}$. The low experimental limits are in agreement with this conclusion.

The beta decay of $Tl^{204}$ to the ground state of $Pb^{204}$ is believed to be a "$\Delta I$ = 2,yes" transition[7,59] indicating the ground state of $Tl^{204}$ is 2-. The log $f_1t$ of the beta decay is 9.1, employing a

half-life value of 4.0 years.[85] The log $f_1t$ for K electron capture using the measured K capture abundance and the reported decay energy of 333 kev[7] is 8.3, indicating a difference of a factor of 5 in comparative half-lives. The observed K/L capture ratio of 3 can be compared to the theoretical value of 1.1. However, in view of the severe absorption corrections on the low energy L x-rays this discrepancy is not very meaningful.

Hogg and Duckworth[85] have measured the $Gd^{160}$-$2Se^{80}$ and $Dy^{160}$-$2Se^{80}$ mass differences from which one can arrive at a value of 3.3 ± 1.0 millimass units for the $Gd^{160}$-$Dy^{160}$ mass difference. This corresponds to 3.0 ± 0.93 Mev. From the intensity of the hard gamma rays following the beta decay of $Tb^{160}$ it appears that the highest energy beta group (0.86 Mev) populates a level at least 0.96 Mev above the ground state of $Dy^{160}$. Since the 1.2 Mev lines are at most only in coincidence with the 87 kev radiation the beta disintegration energy of this terbium isotope is in the range of 1.8 to 2.2 Mev and is probably closer to the former. It is therefore evident that very little if any energy is available for electron capture in $Tb^{160}$. On the basis of the above information $Gd^{160}$ may be unstable with respect to $Tb^{160}$ and decay to it with a very long half-life. At any rate the low limit for K electron capture can easily be understood.

There are no published experimental data from which one can calculate the energy available for K electron capture in $Tm^{170}$. K. Way's[84] beta systematics curves only indicate a small energy but with large uncertainties. The beta decay to the ground state of $Yb^{170}$ has a log ft of 9.0. If the 0.2 percent limit set for K electron

capture did represent the amount of capture then the electron capture energy for a transition of log ft of 9.0 would only be 150 kev. This energy represents an upper limit for the $Tb^{160}$-$Er^{160}$ mass difference if the comparative half-lives for transitions to the ground state of the two daughters are equal.

Precision mass spectrometry by Halsted[81] indicates the $Sn^{124}$-$Te^{124}$ mass difference is equivalent to 2.0 ± 0.2 Mev. Wall,[87] by measuring the Q value for the ground state (d,p) reaction on $Sn^{124}$ and $Te^{124}$ and combining these data with the decay energies of $Sn^{125}$ and $Sb^{125}$ arrived at a value of 2.4 ± 0.1 Mev for this difference. Since the beta disintegration energy of $Sb^{124}$ is 2.90 Mev[57] this nuclide is unstable with respect to electron capture to $Sn^{124}$ by approximately 0.7 Mev.

The ground state of $Sb^{124}$ is thought to be 3- on the basis of beta-gamma angular correlation studies.[82] The first excited state of $Sn^{124}$ is probably of the order of 1 Mev above the ground state and consequently only electron capture to the ground state of $Sn^{124}$ is energetically possible. Such a transition would be third forbidden and would not compete to an appreciable extent with the beta decay. This is consistent with the limit of 0.1 percent set for K electron capture.

Halsted's data on the $Sn^{122}$-$Te^{122}$ mass difference coupled with the beta disintegration energy of 1.94 Mev for $Sb^{122}$ indicate the latter is unstable with respect to $Sn^{122}$ by 1.4 ± 0.2 Mev. The spin of $Sb^{122}$ is 2-. The reason for the difference in spin of $Sb^{122}$ and $Sb^{124}$ probably lies in the spin of the odd proton of these nuclides.

The odd proton in $Sb^{121}$ is in a $d_{5/2}$ state while that of $Sb^{123}$ is in a $g_{7/2}$ state. de-Shalit and Goldhaber[76] have suggested an explanation of this on the basis of configuration mixing. Because of the 2- ground state and the large available energy electron capture occurs in $Sb^{122}$. If the weak 1.15 Mev radiation observed by Glaubman and Metzger[70] represents the first excited state of $Sn^{122}$, K electron capture populates this level 1 percent per disintegration and the ground state 1.6 percent per disintegration (the contribution of L capture is probably small). The log ft for K capture to the 1.15 Mev level is 6.6 which can be compared to the value of 7.5 for the corresponding beta transition to $Te^{122}$. Since the ground state of $Sn^{122}$ is 0+ the K electron capture to it will be a "$\Delta I$ = 2,yes" transition. The log $f_1t$ for this decay, assuming an abundance of 1.6 percent, is 7.8 while the corresponding beta group to the ground state of $Te^{122}$ has log $f_1t$ of 8.6.

Positron emission can only occur in decay to the ground state of $Sn^{122}$. Feenberg and Trigg's[83] curve for $f_{EC}/f_{\beta^+}$ indicates an abundance of less than 1 percent of the electron capture for an energy of 1.4 Mev at Z = 50. Therefore, one might expect the order of 0.01 percent $\beta^+$ in the decay of $Sb^{122}$.

Closed cycle calculations lead to an estimate of 0.92 Mev available for electron capture in $Np^{236}$. On the basis of the experimental branching ratio and the beta disintegration energy of 0.51 Mev the log ft for the transition to $Pu^{236}$ is 6.7. Likewise the log ft for K electron capture to $U^{236}$ is 7.0. Both these values ignore the small branching to the first excited state of the respective daughters.

A spin assignment of 1- to $Np^{236}$ appears reasonable in light of these first forbidden log ft values.

In conclusion it can be stated that the presence or absence of electron capture in the shielded nuclei studied is consistent with the reported or inferred disintegration energy and/or the suggested spin of the parent coupled with current ideas about the excited levels of even-even nuclei. When electron capture decay occurs to a state equivalent to one populated by the beta decay the comparative half-lives were found to agree within a factor of 8. Further refinements in the electron capture decay energy of these nuclides may lead to better agreement.

## V. ACKNOWLEDGMENT

The interest and guidance afforded the author by Professor I. Perlman throughout this study are very gratefully acknowledged. Discussions with and the assistance of Drs. C. I. Browne, F. Asaro, J. O. Rasmussen and Messrs. T. O. Passell and F. S. Stephens have also been most helpful in many phases of the research.

The author is indebted to Drs. S. G. Thompson, G. R. Choppin, B. B. Cunningham, Mr. P. R. Gray, the staff of the Materials Testing Reactor at Arco, Idaho, and the Oak Ridge National Laboratory for making available several of the activities studied and to Drs. W. W. T. Crane, H. G. Higgins, R. W. Hoff and Mr. R. J. Carr for assistance in certain aspects of the chemistry involved in sample preparation. The help of Messrs. H. P. Robinson and A. J. Hartzell in solving instrumental and electronic problems associated with the bent crystal spectrometer is also greatly appreciated.

This work was performed under the auspices of the U.S. Atomic Energy Commission.

# VI. APPENDIX

## A. $Am^{241}$

The gamma ray and L x-ray spectrum associated with the decay of the 470 year $Am^{241}$ has been investigated with a bent crystal spectrometer[11] and a proportional counter.[12] In the former experiment gamma rays of 26.43, 33.36, 38.00, and 59.78 kev were observed while only the 26.4 and 59.7 kev radiations were detected with the proportional counter. A 33 kev transition is called for by the decay scheme of Asaro, Reynolds, and Perlman.[26] In an attempt to resolve this discrepancy a reinvestigation of these radiations was undertaken using the bent crystal spectrometer.

It was noted in several preliminary experiments that the intensity of the 33.4 and 38.0 kev gamma rays varied considerably with respect to the 26.4 and 59.6 kev radiations in different samples, while the latter two lines maintained the same intensity ratio. Interestingly enough the anomalous lines correspond quite well in energy to the dominant K x-ray transitions of lanthanum, the usual carrier for americium. With this in mind portions of our samples were spectroscopically analyzed and a lanthanum impurity amounting to as much as 20 percent of the americium was found. Thus it seemed reasonable to ascribe at least the bulk of these two lines to fluorescence excitation in the impurity. Likewise, several lower energy lines which Browne noted correspond closely to the extrapolated energy of the dominant americium L x-rays are undoubtedly due to fluorescence excitation in the sample. These same conclusions were reached independently by Newton and Rose.[88]

Approximately three milligrams of carefully purified americium in the form of the chloride was obtained from Professor B. B. Cunningham. Spectroscopic analysis proved this material to be 99 percent pure with roughly 0.5 percent lanthanum. The americium was precipitated as the fluoride, flamed to the dioxide and mounted for the bent crystal spectrometer.

Fifteen neptunium L x-rays and 2 americium L x-rays were observed. The data are summarized in Table 16.

The intensities of these lines were corrected for absorbers in path, reflectivity efficiency, and self absorption.

The americium x-rays are due to fluorescent excitation of the sample by the 26.3 and 59.6 kev gamma rays, the $L\beta_1$ and $L\alpha_1$ being the most prominent lines from the $L_2$ and $L_3$ shells, respectively. Radiation from the $L_1$ shell of americium is not observed because of the low fluorescence yield and the more uniform intensity distribution of the $L_1$ x-rays.

The large number of L x-rays observed in this experiment makes it worth-while to calculate the relative intensities of the x-ray lines arising from a given L shell vacancy. These are listed in Table 17 along with Browne's data on $Pa^{233}$ and the figures listed in Compton and Allison for uranium.[33] Because of resolution difficulties in the $L\beta_2$-$L\beta_4$ and $L\gamma_2$-$L\gamma_3$-$L\gamma_6$ multiplets the intensities of these lines are somewhat doubtful.

The quantum yield ratios of the neptunium x-rays calculated from the x-ray intensities are

$$L_1:L_2:L_3 = 1:3.2:1.9 \ .$$

Table 16

L X-Rays Observed in the Decay of $Am^{241}$

| Line | Transition | Observed Energy (kev) | Siegbahn's extrapolated energy (kev) | Corrected relative intensity |
|---|---|---|---|---|
| Np $L_{\alpha_2}$ | $L_3$-$M_4$ | 13.79 ± 0.04 | 13.76 | 7 |
| Np $L_{\alpha_1}$ | $L_3$-$M_5$ | 13.98 ± 0.04 | 13.94 | 64 |
| Np $L_{\eta}$ | $L_2$-$M_1$ | 15.90 ± 0.06 | 15.85 | 2 |
| Np $L_{\beta_6}$ | $L_3$-$N_1$ | 16.15 ± 0.06 | 16.11 | 1 |
| Np $L_{\beta_2}$ | $L_3$-$N_5$ | 16.89 ± 0.02 | 16.88 | 13 |
| Np $L_{\beta_4}$ | $L_1$-$M_2$ | 17.10 ± 0.02 | 17.06 | 16 |
| Np $L_{\beta_5}$ | $L_3$-$O_{4,5}$ | 17.58 ± 0.06 | 17.51 | 3 |
| Np $L_{\beta_1}$ | $L_2$-$M_4$ | 17.78 ± 0.01 | 17.74 | 100 |
| Np $L_{\beta_3}$ | $L_1$-$M_3$ | 18.02 ± 0.02 | 17.98 | 13 |
| Np $L_{\gamma_5}$ | $L_2$-$N_1$ | 20.15 ± 0.08 | 20.09 | 2 |
| Np $L_{\gamma_1}$ | $L_2$-$N_4$ | 20.82 ± 0.02 | 20.77 | 34 |
| Np $L_{\gamma_2}$ | $L_1$-$N_2$ | 21.12 ± 0.04 | 21.09 | 8 |
| Np $L_{\gamma_3}$ | $L_1$-$N_3$ | 21.31 ± 0.04 | 21.34 | 7 |
| Np $L_{\gamma_6}$ | $L_2$-$O_4$ | 21.51 ± 0.04 | 21.49 | 10 |
| Np $L_{\gamma_4}$-$\gamma_4'$ | $L_1$-$O_{2,3}$ | 22.19 ± 0.04 | 22.19 | 3 |
| Am $L_{\alpha_1}$ | $L_3$-$M_5$ | 14.66 ± 0.03 | 14.61 | 10 |
| Am $L_{\beta_1}$ | $L_2$-$M_4$ | 18.76 ± 0.04 | 18.80 | 6 |

Table 17

Relative Intensities of the L X-Rays Arising from a Given L Vacancy

| Vacant shell | Line | Transition | Relative intensity of x-radiation from this shell vacancy | | | Percentage abundance in $Am^{241}$ |
|---|---|---|---|---|---|---|
| | | | $Am^{241}$ | $Pa^{233}$ | C and A | |
| $L_1$ | $L\beta_4$ | $L_1$-$M_2$ | 123 | 135 | 98 | 35 |
| | $L\beta_3$ | $L_1$-$M_3$ | 100 | 100 | 100 | 28 |
| | $L\beta_{10}$ | $L_1$-$M_4$ | -- | 7 | 0 | -- |
| | $L\beta_9$ | $L_1$-$M_5$ | -- | 10 | 0 | -- |
| | $L\gamma_2$ | $L_1$-$N_2$ | 58 | 55 | 36 | 17 |
| | $L\gamma_3$ | $L_1$-$N_3$ | 49 | 66 | 33 | 14 |
| | $L\gamma_4$ | $L_1$-$O_3$ | 21 | 24 | 0 | 6 |
| $L_2$ | $L\eta$ | $L_2$-$M_1$ | 2 | 2 | 2 | 1.5 |
| | $L\beta_1$ | $L_2$-$M_4$ | 100 | 100 | 100 | 67 |
| | $L\gamma_5$ | $L_2$-$N_1$ | 2 | 1.5 | 0 | 1.3 |
| | $L\gamma_1$ | $L_2$-$N_4$ | 34 | 36 | 24 | 23 |
| | $L\gamma_6$ | $L_2$-$O_4$ | 10 | 15 | 4.4 | 7 |
| $L_3$ | $L\ell$ | $L_3$-$M_1$ | -- | -- | 3.4 | -- |
| | $L\alpha_2$ | $L_3$-$M_4$ | 12 | 13 | 11 | 8.5 |
| | $L\alpha_1$ | $L_3$-$M_5$ | 100 | 100 | 100 | 72 |
| | $L\beta_6$ | $L_3$-$N_1$ | 2 | 2 | 1.6 | 1.5 |
| | $L\beta_2$ | $L_3$-$N_5$ | 20 | 35 | 28 | 14 |
| | $L\beta_7$ | $L_3$-$O_1$ | -- | -- | 0.4 | -- |
| | $L\beta_5$ | $L_3$-$O_{4,5}$ | 5 | -- | 6.4 | 4 |

Employing Kinsey's figures for the fluorescence yields of the 3 L shells but ignoring the Coster-Kronig effect for the moment, the L vacancy ratios are

$$L_1:L_2:L_3 = 1.0:0.9:0.76 .$$

Aside from the gamma ray study on the bent crystal spectrometer use was made of the NaI (thallium activated) scintillation spectrometer. A sample of 2.5 micrograms of the purified americium was used for this latter investigation. In order to examine the energy region above 60 kev an absorber of 1.1 grams of silver was employed to decrease the intensity of the 59.6 kev line. Under these conditions four new gamma rays were observed. The data re summarized in Table 18. The first three entries were detected with the bent crystal spectrometer and the remaining lines were observed with the scintillation counter.

Table 18

Gamma Rays Following the Decay of $Am^{241}$

| Gamma ray energy (kev) | Intensity (photon/alpha) | Intensity per 100 Np $L_{\beta_1}$ |
|---|---|---|
| 26.38 ± 0.04 | 0.04 | 35 |
| 59.65 ± 0.13 | 0.40 | 360 |
| 59.62 ± 0.06 (2nd order) | -- | -- |
| 102 | $5.6 \times 10^{-4}$ | -- |
| 128 | $5.6 \times 10^{-5}$ | -- |
| 168 | $6.4 \times 10^{-6}$ | -- |
| 207 | $7.6 \times 10^{-6}$ | -- |

All intensities were measured relative to the 59.6 kev transition and its intensity has been determined as 0.40 ± 0.015 photon per alpha particle.[12] A careful search of the 30-40 kev region on the bent crystal spectrometer yielded negative results. The maximum intensity of any radiation in this region is 2 percent of the 59.6 kev gamma ray or 0.008 photon per alpha particle. A portion of the spectrum observed with the bent crystal spectrometer is shown in Figure 11 and the scintillation counter spectrum is shown in Figure 12.

The 102 kev gamma ray agrees with the expected energy of the neptunium K x-rays. In an attempt to determine whether the line was a gamma ray or the K x-ray multiplet the width of the peak was compared with the K x-ray peak found in the long lived $Am^{242}$. The width of the latter was more than 50 percent larger than that of the former and thus it is certain that at least the bulk of the 102 kev radiation is of nuclear origin. Gamma-gamma coincidence studies by Frank Stephens[89] indicate this gamma ray is in coincidence with the 60 kev radiation. Several low intensity conversion lines from a 99.0 kev gamma ray have been observed.[37,90]

The 128 kev transition has not been previously reported. It is interesting to note that the alpha particle fine structure of $Am^{241}$ indicates two levels separated by 126 kev.[26]

Both a 165 and a 207 kev gamma ray have been observed in the beta decay of $U^{237}$.[91] Thus their appearance in the $Am^{241}$ gamma ray spectrum is not too surprising in view of the fact that the daughter of both decays is $Np^{237}$. Because of the extremely low intensity of these transitions coincidence studies were not possible. The decay scheme

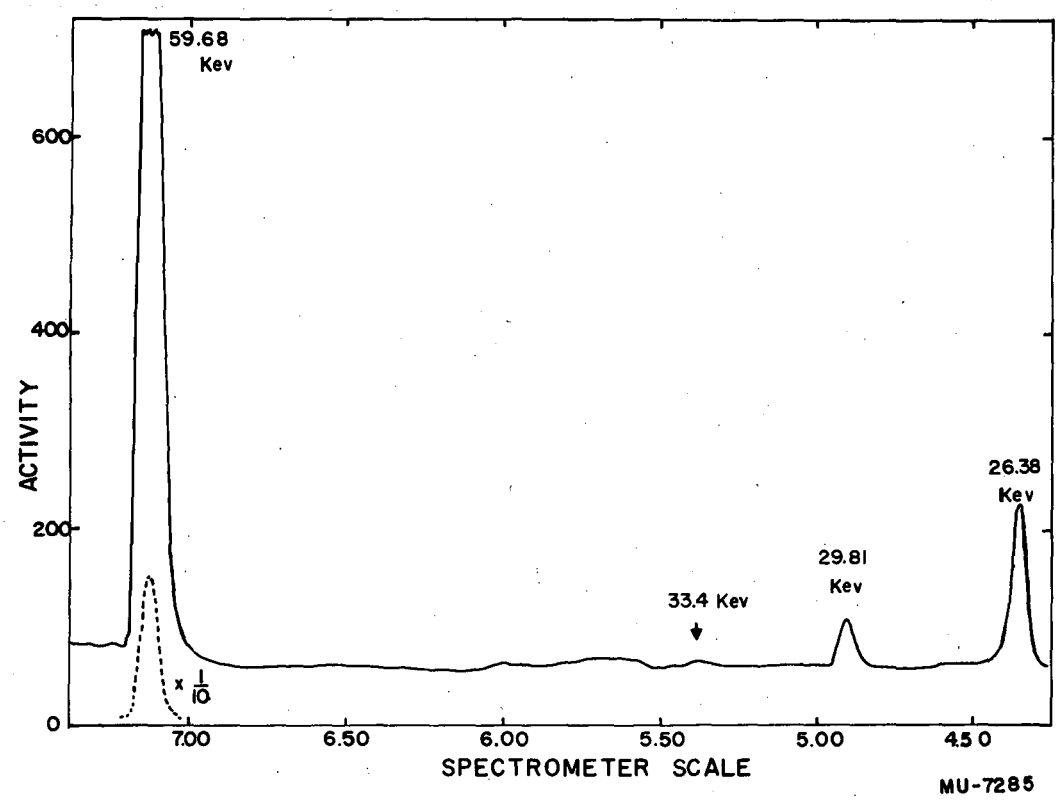


Fig. 11. Spectrum of the 25-70 kev region in the decay of $Am^{241}$.

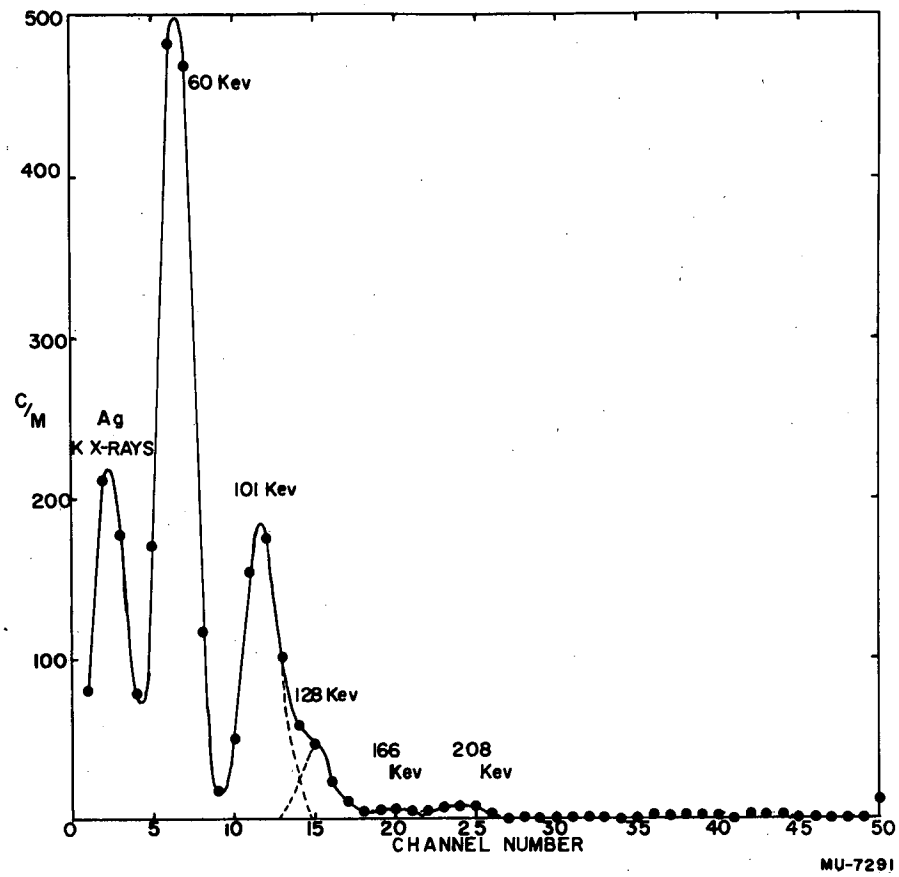


Fig. 12. Scintillation counter spectrum of $Am^{241}$.

of $Am^{241}$ is shown in Figure 13. The level previously reported 11 kev below what is now called the ground state of $Np^{237}$ has been omitted on the basis of recent experimental work.[92] Several lines not discussed above are included in the decay scheme as the basis of electron line studies.[37,90]

The relative intensities listed in Tables 16 and 18 allow one to calculate the ratio of L x-rays to 59.6 kev gamma rays. The number thus obtained is 0.8. This is to be compared with the value of 1.0 determined by Beling et al.[21]

The conversion coefficient of the 59.6 kev gamma ray has been determined as 0.92,[90] and its absolute abundance as 0.40 photon per alpha particle. Since essentially every decay passes through this state and the total number of 59.6 kev transitions is 0.77 per alpha particle, the intensity of the 26.4 kev transition is 0.23 per alpha particle. From the measured photon intensity the total conversion coefficient of the 26.4 kev gamma ray is 5. Clearly these two radiations originating from the 59.6 kev level are both electric dipole transitions on the basis of their low conversion coefficients.

One can now discuss the possibility of observing 33.2 kev photons from the transition called for by Asaro's alpha particle fine structure. The intensity of this transition is for all intents the same as that of the 26.4 kev precursor. We know from the fact that both the 26.4 and the 59.6 kev transitions are electric dipole and populate the initial and final states, respectively, of the 33.2 kev transition that the latter gamma ray must involve no parity change and a spin change of 1 or 2 units of angular momentum. A magnetic dipole transition of

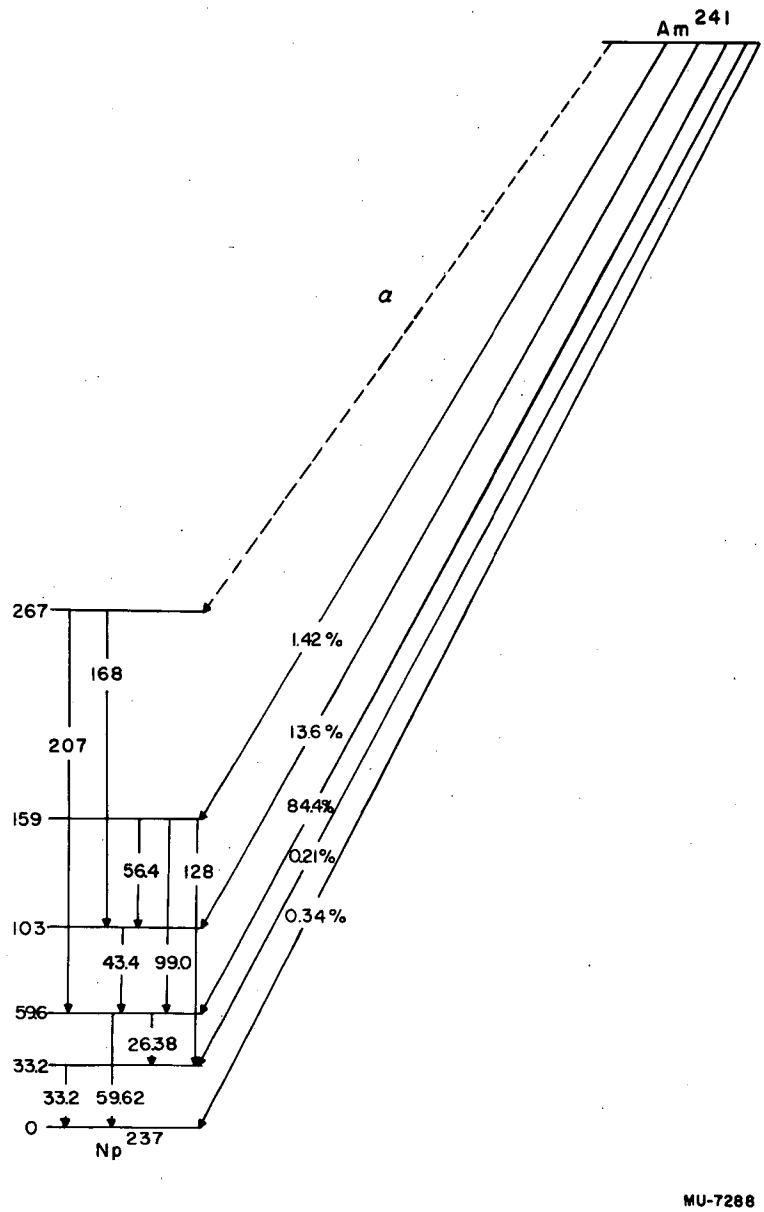


Fig. 13. Decay scheme of $Am^{241}$.

this energy would have an L conversion coefficient of about 100 according to the theoretical calculations.[28] Electric quadrupole radiation would be more highly converted. Thus from 0.23 transition one might expect to have less than 0.002 33.2 kev photon as $\alpha_L$ is less than the total conversion coefficient. This is consistent with failure to observe the radiation is these experiments.

Passell[90] has arrived at a rough figure for the $L_1$ to $L_3$ conversion ratio in the decay of $Am^{241}$ and finds 3:1. Coupled with the calculated L vacancy ratios this indicates an upper limit of approximately 40 percent for an $L_1 \longrightarrow L_3$ Coster-Kronig transition. It must be realized that this limit is very crude because of the difficulty of resolving the $L_1$ and $L_2$ conversion lines. Browne's[11] data on $Pb^{210}$ indicates a value of 16 percent for such a transition at Z = 83 which can be compared to the predicted value of 70 percent.[27] These discrepancies with Kinsey's estimates indicate, as Browne has pointed out, the large uncertainties in the various Auger transition probabilities of this type. High resolution beta spectroscopy coupled with the use of the bent crystal spectrometer should allow the accurate determination of the magnitude of the Coster-Kronig coefficient and lead to more reliable L shell fluorescence yields.

## B. $Np^{239}$

The 2.33 day $Np^{239}$ which decays by beta emission to $Pu^{239}$ has been reported[57] to have several low energy gamma rays as determined by beta spectrometer studies. Aside from the inherent interest in these the possibility of producing large quantities of this isotope in a

nuclear reactor by the following reaction

$$U^{238}(n,\gamma)U^{239} \xrightarrow[23.5\ \text{min}]{\beta^-} Np^{239}$$

affords an opportunity to study the L x-ray spectrum of the plutonium daughter. The L x-rays arise from the filling of the various L shells vacated by conversion of several of the low energy gamma rays.

Uranium metal was neutron irradiated in the Oak Ridge reactor. The sample was dissolved in HCl and the neptunium was reduced to the (IV) state by heating with formic acid. After adjusting the HCl concentration to 2 M the neptunium was extracted into 0.4 M TTA in benzene, washed twice with 2 M HCl and then back extracted into 10 M $HNO_3$. The sample was evaporated to dryness and taken up into 2 M HCl and the cycle was repeated. Finally 0.5 milligram of $FeCl_3$ was added and the iron was precipitated as the hydroxide carrying the neptunium activity. This material was powdered and packed in a pyrex capillary for the bent crystal spectrometer.

In an attempt to increase the resolution of the spectrometer a new collimator was employed throughout this experiment. The collimator was constructed of wolfram plates 0.010 inch thick spaced 0.010 inch apart at the end of the collimator closest to the topaz crystal. In later experiments it was found that warpage of the wolfram plates was causing appreciable absorption. Unfortunately when this effect was noted it was no longer feasible to rerun the spectrum. Consequently the relative intensities of the various lines are not thought to be too reliable unless the energies are close together.

The data on the K and L x-rays observed in the decay of $Np^{239}$ are summarized in Table 19. For comparison the energies of several plutonium L lines observed in the decay of $Am^{242m}$ are included. The latter experiment was performed after the new and more accurately graduated spectrometer scale was incorporated on the instrument. The x-ray energies obtained by a Mosely extrapolation of the data tabulated by Siegbahn for uranium and thorium are also listed. The K x-ray energies have been obtained from the extrapolated values of the absorption edges.[62]

It will be noticed that the energy of the plutonium $K\alpha_1$ is considerably higher than the calculated value while good agreement is obtained with the $K\alpha_2$. A possible explanation of this is the presence of an unresolved gamma ray of approximately 105 kev.[57] However, it is felt that the bulk of this line is due to the plutonium $K\alpha_1$ because of the presence of the $K\alpha_2$ line. Also, from data listed in Compton and Allison[33] the $K\alpha_1/K\alpha_2$ intensity ratio in lighter elements is 2, and apparently very insensitive to Z. The ratio observed here is 2.2.

Because of the collimator absorption it is not meaningful to calculate the quantum yields and vacancy ratios. An initial attempt to do this yielded insufficient $L_3$ vacancies to account for all the $K\alpha_1$ radiation.

Table 19

X-Rays Following the Decay of $Np^{239}$

| Line | Transition | Observed energy (kev) | Energy observed in decay of $Am^{242m}$ (kev) | Siegbahn's extrapolated energy (kev) | Corrected relative intensity |
|---|---|---|---|---|---|
| $L\alpha_2$ | $L_3$-$M_4$ | 14.13 ± 0.03 | 14.08 ± 0.03 | 14.08 | 6 |
| $L\alpha_1$ | $L_3$-$M_5$ | 14.33 ± 0.02 | 14.28 ± 0.03 | 14.28 | 36 |
| $L\beta_2$ | $L_3$-$N_5$ | 17.32 ± 0.01 | 17.29 ± 0.03 | 17.25 | 24 |
| $L\beta_4$ | $L_1$-$M_2$ | 17.62 ± 0.03 | -- | 17.56 | 6 |
| $L\beta_5$ | $L_3$-$O_{4,5}$ | 18.02 ± 0.01 | -- | 17.95 | 5 |
| $L\beta_1$ | $L_2$-$M_4$ | 18.36 ± 0.01 | 18.33 ± 0.02 | 18.27 | 100 |
| $L\beta_3$ | $L_1$-$M_3$ | 18.60 ± 0.01 | -- | 18.52 | 7 |
| $L\gamma_5$ | $L_2$-$N_1$ | 20.75 ± 0.03 | -- | 20.68 | 2 |
| $L\gamma_1$ | $L_2$-$N_4$ | 21.50 ± 0.02 | 21.46 ± 0.04 | 21.38 | 37 |
| $L\gamma_2$ | $L_1$-$N_2$ | 21.82 ± 0.02 | -- | 21.70 | 5 |
| $L\gamma_3$ | $L_1$-$N_3$ | 22.07 ± 0.04 | 22.06 ± 0.1 | 21.97 | 5 |
| $L\gamma_6$ | $L_2$-$O_4$ | 22.25 ± 0.02 | 22.24 ± 0.1 | 22.13 | 9 |
| $L\gamma_4$ | $L_1$-$O_3$ | 22.97 ± 0.04 | -- | 22.88 | 2 |
| $K\alpha_2$ | K-$L_2$ | 100.0 ± 0.4 | -- | 99.50* | 110 |
| $K\alpha_1$ | K-$L_3$ | 105.6 ± 0.5 | -- | 103.69* | 240 |

*Calculated from the extrapolated energy[62] of the K and L edges.

Two gamma rays were observed in the spectrum and their energies and intensities relative to the plutonium $L_{\beta_1}$ x-ray are given in Table 20.

Table 20

Gamma Rays Observed in the Decay of $Np^{239}$

| Gamma ray energy (kev) | Intensity |
|---|---|
| 57.60 ± 0.3 | 5 |
| 61.83 ± 0.3 | 25 |

It is not possible to use the data reported above for the calculation of conversion coefficients of any of the numerous gamma rays which are associated with the decay. However, the intensity of the two observed lines indicates they are probably dipole radiation.

## C. $Pu^{238}$

In the study of the electromagnetic radiation associated with both $Pa^{232}$ and $Pa^{233}$ Browne[11] observed a shift in the energy of the uranium $L_{\beta_1}$ line ($L_2$-$M_4$) relative to the energy observed by electron bombardment. Although the discrepancy was small (60 volts) it appeared to be well outside of the experimental error. The uranium $L_{\beta_1}$ was also observed in the alpha decay of $Pu^{239}$ in very low intensity and its measured energy was in better agreement with the literature value. Since both protactinium isotopes are beta emitters it appeared worth-while to investigate another alpha active plutonium isotope.

In an effort to resolve the apparent discrepancy a study of low energy electromagnetic radiation associated with the alpha decay of $Pu^{238}$ was undertaken. An intense source of $Pu^{238}$ was obtained from Professor B. B. Cunningham and the fluoride was precipitated and mounted in the usual fashion.

Table 21 presents the energies and the relative intensities of the observed uranium L x-rays. Only $L_2$ and $L_3$ lines were seen. The source of these x-rays is the L shell conversion of a 43.8 kev gamma ray in the uranium daughter. This radiation has been shown to be an electric quadrupole transition[93] and according to the theoretical conversion coefficients[28] should predominantly excite the $L_2$ and $L_3$ levels.

Table 21

L X-Rays Following the Decay of $Pu^{238}$

| Line | Transition | Observed energy (kev) | Browne's[11] energy $Pa^{233}$ (kev) | Siegbahn's extrapolated energy (kev) | Corrected relative intensity |
|---|---|---|---|---|---|
| U $L_{\gamma_6}$ | $L_2$-$O_4$ | 20.91 ± 0.05 | 20.85 ± 0.04 | 20.85 | 11 |
| U $L_{\gamma_1}$ | $L_2$-$N_4$ | 20.22 ± 0.03 | 20.75 ± 0.02 | 20.16 | 41 |
| U $L_{\beta_1}$ | $L_2$-$M_4$ | 17.28 ± 0.01 | 17.27 ± 0.01 | 17.22 | 100 |
| U $L_{\beta_2}$ | $L_3$-$N_5$ | 16.46 ± 0.02 | 16.49 ± 0.02 | 16.43 | 15 |
| U $L_{\alpha_1}$ | $L_3$-$M_5$ | 13.63 ± 0.02 | 13.65 ± 0.01 | 13.61 | 19 |

The agreement of the energy of the uranium $L_{\beta_1}$ with that found in previous experiments is excellent. An examination of the original data on the L x-rays in the $Pu^{239}$ decay showed that within the experimental

error the $L_{\beta_1}$ energy agreed with both these data and the electron bombardment studies. Thus it was thought possible that the apparent shift was due to an instrumental effect. Since other x-rays have been seen on this instrument in good agreement with published energies a reasonable explanation seemed to be a non-linearity in the spectrometer scale from which the Bragg angle was determined. Such a situation could give rise to an error in a narrow region without affecting the remainder of the energy spectrum. To test this hypothesis another experiment to be described shortly was planned.

The quantum yield ratio of the two L shells can be calculated from the above relative intensities after correction for the fraction of the $L_2$ and $L_3$ x-rays not observed. The result is

$$L_2 : L_3 = 4:1 \ .$$

Asaro[93] has found that the absolute intensity of L x-rays in the decay of $Pu^{238}$ is 0.13 per alpha disintegration. Combining this with the above ratio and Kinsey's fluorescence yields the absolute number of vacancies can be calculated as

$$L_2 = 0.16 \text{ vacancy/alpha particle}$$

$$L_3 = 0.065 \text{ vacancy/alpha particle} \ .$$

The absolute abundance of the 43.8 kev gamma ray has been determined as $3.8 \times 10^{-4}$ and $4.2 \times 10^{-3}$.[93,94] Because of the experimental method used the former figure should be more accurate. Employing this intensity the following L conversion coefficients were calculated:

$$\alpha_{L_2} = 420 \qquad \alpha_{L_3} = 170 \qquad \alpha_{L_{total}} = 590 \ .$$

The total L conversion coefficient is in good agreement with the predicted value of about 700 for an E2 transition of this energy at Z = 92, but the $L_2$ to $L_3$ ratio is roughly 2.5 times the theoretical value.

It should be mentioned that two other gamma rays have been observed by Asaro in the decay of $Pu^{238}$. However, the population of the levels from which they originate are so small that the L vacancies due to their conversion are completely negligible.

A careful search was made of the 40-50 kev region in an attempt to detect the 43.8 kev radiation but without success. This was to be expected from the observed x-ray intensity and the reported low abundance of the gamma ray.

## D. Fluorescence Excitation in Uranium

The 470 day $Cd^{109}$ decays by electron capture[57] to a metastable state of $Ag^{109}$. The half-life of this state is 39 seconds so that it will be in equilibrium with the cadmium parent. From the high K/L capture ratio of $Cd^{109}$ and the large K conversion coefficient of the isomeric transition it is evident that abundant K series radiation of silver will accompany the decay. The energy of the silver $K_{\alpha_1}$ line (22.16 kev) which is the dominant feature in the K multiplet is only slightly greater than the uranium $L_2$ absorption edge (20.94 kev) and is expected to be fairly efficient in exciting fluorescence in this shell. The uranium $L_{\beta_1}$, excited in this fashion and observed on the spectrometer, could certainly indicate the reality or instrumental nature of the above mentioned energy discrepancy.

To these ends a 20 mil silver foil was bombarded on the 60-inch cyclotron with 19 Mev deuterons for a total of 1,100 microampere hours. The short lived silver activities were allowed to decay for about two months. At that time the silver was dissolved in nitric acid and the chloride was precipitated. The supernate contained approximately $5 \times 10^9$ disintegrations per minute of $Cd^{109}$. To this solution was added several milligrams of uranium in the form of the nitrate and uranium hydroxide was precipitated. Under these conditions 80 percent of the cadmium followed the uranium. The precipitated was darkened by a small amount of silver which had not been completely removed. The sample was dried under heat lamp and mounted for the spectrometer.

The silver $K_{\alpha_1}$ line was observed and in addition a relatively weak uranium $L_{\beta_1}$ was detected. The energy of the latter was determined on two separate alignments of the spectrometer and is listed in Table 22.

Table 22

Fluorescent Excitation of Uranium by $Cd^{109}$ Radiation

| Line | Observed energy (kev) | Literature energy[9] (kev) |
|---|---|---|
| Ag $K_{\alpha_1}$ | 22.23 ± 0.04 | 22.16 |
| U $L_{\beta_1}$ | 17.29 ± 0.02<br>17.28 ± 0.02 | 17.22 |

Since the $L_{\beta_1}$ energy obtained by fluorescence excitation in the uranium agreed so well with that determined in previous experiments it was clear that the discrepancy was of an instrumental nature. To correct this effect a new spectrometer scale was machined and very accurately graduated. The old scale was replaced by this new one and it was aligned so as to be concentric with the sample arm to 0.0001 inch. Re-examination of the $Cd^{109}$-uranium spectrum gave the following results:

| | |
|---|---|
| Ag $K_{\alpha_1}$ | 22.19 ± 0.04 |
| U $L_{\beta_1}$ | 17.24 ± 0.02 . |

Both lines are still somewhat too high in energy but check with the published values within experimental error.

## VII. REFERENCES


1. M. Goldhaber and A. W. Sunyar, Phys. Rev. 83, 906 (1951).
2. G. Scharff-Goldhaber, Phys. Rev. 90, 587 (1953).
3. F. Asaro and I. Perlman, Phys. Rev. 87, 393 (1952).
4. F. Asaro and I. Perlman, Phys. Rev. 91, 763 (1953).
5. D. Maeder and P. Preiswerk, Phys. Rev. 84, 595 (1951).
6. C. E. Anderson, G. W. Wheeler, and W. W. Watson, Phys. Rev. 87, 668 (1952).
7. E. der Mateosian and A. Smith, Phys. Rev. 88, 1186 (1952).
8. A. H. Compton and S. K. Allison, X-Rays in Theory and Experiment. (D. Van Nostrand Company, Inc., New York, 1935) pp. 784-790.
9. C. Dauben, Tables of Energies in Kev of the K and L X-Rays, unpublished report (January 1953).
10. M. Siegbahn, Spektroskopie der Rontgenstrahlen (Verlag Von Julius Springer, Berlin, 1931).
11. C. I. Browne, Jr., Ph.D. Thesis, University of California Radiation Laboratory Declassified Report UCRL-1764 (June 1952).
12. J. K. Beling, J. O. Newton, and B. Rose, Phys. Rev. 86, 797 (1952).
13. G. W. Barton, Jr., H. P. Robinson, and I. Perlman, Phys. Rev. 81, 208 (1951).
14. J. W. M. DuMond and E. R. Cohen, Revs. Modern Phys. 25, 691 (1953).
15. W. R. Dixon, Nucleonics 8, 68 (1951).
16. A. H. Compton and S. K. Allison, op. cit., p. 540.
17. D. A. Lind, W. J. West, and J. W. M. DuMond, Phys. Rev. 77, 475 (1950).
18. A. H. Compton and S. K. Allison, op. cit., p. 800.

19. P. M. McLaughlin and G. D. O'Kelley, California Research and Development Company Report MTA-40 (1953).

20. R. W. Hoff, private communication (July 1953).

21. J. K. Beling, J. O. Newton and B. Rose, Phys. Rev. 87, 1144 (1952) (erratum).

22. M. S. Freedman, A. H. Jaffey, and F. Wagner, Jr., Phys. Rev. 79, 410 (1950).

23. H. Slätis, J. O. Rasmussen, Jr., and H. Atterling, Phys. Rev. 93, 646 (1954).

24. G. T. Seaborg, R. A. Glass and S. G. Thompson, J. Am. Chem. Soc., to be published (1954).

25. J. W. Mihelich, Phys. Rev. 87, 646 (1952).

26. F. Asaro, F. L. Reynolds, and I. Perlman, Phys. Rev. 87, 277 (1952).

27. B. B. Kinsey, Can. J. Research 26A, 404 (1948).

28. H. Gellman, B. A. Griffith, and J. P. Stanley, Phys. Rev. 85, 944 (1952).

29. G. D. O'Kelley, G. W. Barton, Jr., W. W. T. Crane, and I. Perlman, Phys. Rev. 80, 293 (1950).

30. D. Coster and R. de L. Kronig, Physica 2, 13 (1935).

31. C. D. Broyles, D. A. Thomas, and S. K. Haynes, Phys. Rev. 89, 715 (1953).

32. E. H. S. Burhop, The Auger Effect and Other Radiationless Transitions (Cambridge University Press, 1952), p. 48.

33. A. H. Compton and S. K. Allison, op cit., pp. 640-644.

34. T. O. Passell, private communication (February 1953).

35. F. S. Stephens, private communication (February 1954).

36. G. H. Higgins and S. G. Thompson, unpublished data (February 1953).

37. E. L. Church, Argonne National Laboratory Classified Report ANL-5141 (November 1953).

38. D. C. Dunlavey, private communication (1952).

39. G. D. O'Kelley, Ph.D. Thesis, University of California Radiation Laboratory Unclassified Report UCRL-1243 (May 1951).

40. B. G. Harvey, Phys. Rev. 85, 482 (1952).

41. K. Street, Jr., A. Ghiorso, and G. T. Seaborg, Phys. Rev. 79, 530 (1950).

42. J. M. Cork, J. M. LeBlanc, A. E. Stoddard, W. J. Childs, C. E. Branyan, and D. W. Martin, Phys. Rev. 82, 258 (1951).

43. D. E. Muller, H. C. Hoyt, D. J. Klein, and J. W. M. DuMond, Phys. Rev. 88, 775 (1952).

44. K. I. Roulston and R. W. Pringle, Phys. Rev. 87, 930 (1952).

45. F. S. Stephens, private communication (October 1953).

46. W. Mims and H. Halban, Proc. Phys. Soc. (London) 64A, 311 (1951).

47. A. A. Bashilov, N. M. Anton'eva and B. S. Dzhelepov, Izvest. Akad. Nauk Sci. Fiz. SSSR 16, 264 (1952).

48. M. E. Rose, G. H. Goertzel, and C. L. Perry, Oak Ridge National Laboratory Unclassified Report ORNL-1023 (June 1951).

49. K. Siegbahn, Phys. Rev. 77, 233 (1950).

50. J. M. Cork, W. C. Rutledge, C. E. Branyan, A. E. Stoddard, W. J. Childs, and J. M. LeBlanc, Phys. Rev. 80, 286 (1950).

51. J. Miskel, Phys. Rev. 79, 403 (1950).

52. N. M. Anton'eva, A. A. Bashilov, and B. S. Dzhelepov, Doklady Akad. Nauk SSSR 77, 41 (1951) (as reported in J. M. Hollander, I. Perlman, and G. T. Seaborg, Revs. Modern Phys. 25, 469 (1953).

53. M. Deutsch, Phys. Rev. 72, 527 (1947) (A).

54. J. M. Cork, J. M. LeBlanc, W. H. Nester, D. W. Martin, and M. K. Brice, Phys. Rev. 90, 444 (1953).

55. M. A. Waggoner, M. L. Moon, and A. Roberts, Phys. Rev. 80, 420 (1950).

56. K. Siegbahn and M. Deutsch, Phys. Rev. 73, 410 (1948).

57. The data are summarized in J. M. Hollander, I. Perlman, and G. T. Seaborg, Revs. Modern Phys. 25, 469 (1953).

58. A. C. G. Mitchell and R. S. Caird, Phys. Rev. 87, 388 (1952).

59. L. Lidofsky, P. Macklin and C. S. Wu, Phys. Rev. 87, 391 (1952).

60. S. B. Burson, K. W. Blair, and D. Saxon, Phys. Rev. 77, 403 (1950).

61. J. M. Cork, C. E. Branyan, W. C. Rutledge, A. E. Stoddard, and J. M. LeBlank, Phys. Rev. 78, 304 (1950).

62. R. D. Hill, E. L. Church, and J. W. Mihelich, Rev. Sci. Instruments 23, 523 (1952).

63. J. M. Cork, J. M. LeBlanc, W. H. Nester, and F. B. Stumpf, Phys. Rev. 88, 685 (1952).

64. F. K. McGowan, Phys. Rev. 85, 151 (1952).

65. R. L. Graham, J. L. Wolfson, and R. E. Bell, Can. J. Phys. 30, 459 (1952).

66. L. M. Langer and J. W. Starner, Phys. Rev. 93, 253 (1954).

67. L. M. Langer, R. D. Moffat, and H. C. Price, Jr., Phys. Rev. 79, 808 (1950).

68. D. R. Hutchinson and M. L. Wiedenbeck, Phys. Rev. 88, 699 (1952).

69. E. P. Tomlinson, S. L. Ridgeway, and K. Gopalakrishnan, Phys. Rev. 91, 484 (1953) (A).

70. M. L. Glaubman and F. R. Metzger, Phys. Rev. 87, 203 (1952) (A), verbal report in Nuclear Science Abstracts 6, No. 19 (1952), Supp. II.

71. D. A. Orth and G. D. O'Kelley, Phys. Rev. 82, 758 (1951).

72. W. W. T. Crane and G. H. Higgins, private communication (February 1954).

73. R. W. Hoff, Ph.D. Thesis, University of California Radiation Laboratory Declassified Report UCRL-2325 (September 1953).

74. T. O. Passell, private communication (February 1954).

75. F. Asaro and I. Perlman, Phys. Rev. 88, 828 (1952).

76. A. de-Shalit and M. Goldhaber, Phys. Rev. 92, 1211 (1953).

77. S. A. Moszkowski, Phys. Rev. 82, 35 (1951).

78. R. W. King, N. M. Dismuke, and K. Way, Oak Ridge National Laboratory Unclassified Report ORNL-1450 (October 1952).

79. R. E. Marshak, Phys. Rev. 61, 431 (1942).

80. J. P. Davidson, Jr., Phys. Rev. 82, 48 (1951).

81. R. E. Halsted, Phys. Rev. 88, 666 (1952).

82. M. Goldhaber and R. D. Hill, Revs. Modern Phys. 24, 179 (1952).

83. E. Feenberg and G. Trigg, Revs. Modern Phys. 22, 399 (1950).

84. K. Way and M. Wood, Revs. Modern Phys., to be published. I am indebted to the authors for making a preprint of this paper available.

85. G. Harbottle, Phys. Rev. 91, 1234 (1953).

86. B. G. Hogg and H. E. Duckworth, Can. J. Phys. 32, 65 (1954).

87. N. S. Wall, Phys. Rev. 92, 1526 (1953).

88. J. O. Newton and B. Rose, Phys. Rev. 89, 1157 (1953).

89. F. S. Stephens, private communication (1953).

90. T. O. Passell, private communication (June 1953).

91. F. Wagner, Jr., M. S. Freedman, D. W. Engelkemeir, and J. R. Huizenga, Phys. Rev. 89, 502 (1953).

92. F. Asaro and I. Perlman, Phys. Rev., to be published (1954).

93. F. Asaro and I. Perlman, University of California Radiation Laboratory Declassified Report UCRL-2419 (November 1953).

94. G. W. Reed, Jr., U. S. Atomic Energy Commission Declassified Report AECD-3185 (January 1951).