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FINAL REPORT  
PROPERTIES OF  
RADIOISOTOPE HEAT SOURCES

25 March 1965 to 31 August 1965

PART I  
(UNCLASSIFIED)

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Contract NAS 5-9156

by

COOK ELECTRIC COMPANY  
TECH-CENTER DIVISION  
MORTON GROVE, ILLINOIS

Prepared for

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION  
GODDARD SPACE FLIGHT CENTER  
ADVANCED POWER SOURCES SECTION  
GREENBELT, MARYLAND

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GREENBELT, MARYLAND

## **FINAL REPORT**

**TITLE:** Properties of Radioisotope Heat Sources

**CONTRACTOR:** Tech-Center Division  
Cook Electric Company  
Morton Grove, Illinois

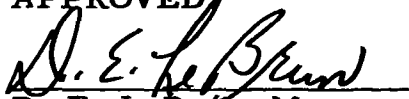
**PERIOD:** From 25 March 1965 to 31 August 1965

**CLIENT:** National Aeronautics and Space Administration  
Goddard Space Flight Center  
Advanced Power Sources Section  
Greenbelt, Maryland  
Mr. Dale Harris, Program Manager

**CONTRACT:** NAS 5-9156

  
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Instrumentation Section



## ABSTRACT

A report outlining the best available unclassified information on the nuclear, chemical, and physical properties of nine SNAP isotopes was prepared for NASA/GSFC Greenbelt, Maryland, Advanced Power Sources Section.

The isotopes reviewed are: Sr-90, Cs-134, Cs-137, Ce-144, Pm-147, Po-210, Pu-238, Cm-242, Cm-244. The properties reviewed were (1) Half Life; (2) Neutrons/Spontaneous Fission; (3) Neutrons from Spontaneous Fission; (4) Other sources of Radiation; (5) Energy Levels and Decay Schemes; (6) Fuel Forms; (7) Material Compatibility; (8) Effects of Impurities; (9) Thermal Conductivity; (10) Power Density; (11) Specific Power; (12) Heat Capacity; (13) Heat of Fusion; (14) Weight Density; (15) Melting Point; (16) Boiling Point; (17) Specific Activity; (18) Isotope Production, Availability, and Cost. At the end of each isotope section an annotated bibliography pertaining to that isotope is presented.

## ACKNOWLEDGMENT

The author wishes to express his appreciation to Dr. S. J. Rimshaw of the Oak Ridge National Laboratory and Mr. H. H. Van Tuyl of Battelle Northwest. Their constructive criticism and many helpful suggestions are gratefully acknowledged.

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## 1.0 INTRODUCTION

### 1.1 Purpose

This final report has been prepared for the Advanced Power Sources Section, Goddard Space Flight Center, Greenbelt, Maryland in partial fulfillment of the requirements set forth in NASA Contract NAS 5-9156. The portion of work on the subject contract covered by this report was initiated to satisfy two purposes. These were:

- (1) Edit the final reports generated by the Cook Electric Company under Contract NAS 5-3849 and by the Martin Company under Contract NAS 5-3879 and combine these reports into a single coherent volume.
- (2) Perform an unclassified literature search on Ce-144 from 1 January 1958 to the latest available information.
- (3) Include new references generated since the publication of the Cook and Martin reports.

### 1.2 Scope

Sections describing the nuclear, chemical, and physical characteristics, isotope production methods and costs of nine SNAP isotopes are presented. These isotopes are:

- (1) Strontium-90
- (2) Cesium-134
- (3) Cesium-137
- (4) Cerium-144
- (5) Promethium-147
- (6) Polonium-210
- (7) Plutonium-238
- (8) Curium-242
- (9) Curium-244

At the end of each section a bibliography for that isotope is presented. The bibliographies for all sections except Ce-144

cover the period from 1 January 1948 to 31 August 1965. The bibliography for Ce-144 begins with 1958.

### 1.3 General Program Background

For some time it has been of interest to the Advanced Power Sources Section of NASA to make available to those persons investigating power production through isotopes a single reference source describing the various properties of the most attractive SNAP isotopes. To accomplish this the Tech-Center Division of the Cook Electric Company and the Martin Company were contracted to prepare reports on these isotopes. The Cook effort was devoted mainly to a study of nuclear characteristics. The Martin Company effort dealt mainly with chemical and physical properties of the various isotopes. After the conclusion of these two programs the Tech-Center Division of Cook Electric Company contracted with NASA/GSFC to combine these two reports into a single coherent volume.

## 2.0 DESCRIPTION OF THE REPORT

### 2.1 Reference Sources

Two general sources of information were used in the compilation of the report. These were:

- (1) A review of unclassified government reports and scientific periodicals.
- (2) Letters of inquiry and personal visits to recognized authorities in the various fields.

### 2.2 Verification of Data Accuracy

After the completion of the various sections, a rough draft of each section was submitted for review to an authority in that particular field. A list of these persons and their field of interest is as follows:

- |     |                 |  |
|-----|-----------------|--|
| (1) | Strontium-90    | Dr. Thomas Bustard<br>Hittman Associates, Inc.<br>Baltimore, Maryland  |
| (2) | Cesium-134, 137 | Dr. R. E. McHenry<br>Dr. S. J. Rimshaw<br>Oak Ridge National Labs<br>Oak Ridge, Tennessee  |
| (3) | Cerium-144      | Dr. R. E. McHenry<br>Oak Ridge National Labs<br>Oak Ridge, Tennessee   |
| (4) | Promethium-147  | Dr. R. L. Moore<br>Dr. E. J. Wheelwright<br>H. H. Van Tuyl<br>Battelle Northwest<br>Pacific Northwest Labs<br>Richland, Washington |
| (5) | Polonium-210    | Dr. C. J. Kershner<br>Mound Laboratories<br>Miamisburg, Ohio   |

(7) Curium-242, 244 Dr. Butler  
Dr. S. J. Rimshaw  
Oak Ridge National Labs  
Oak Ridge, Tennessee

## 2.3 Report Format

Each section of the report has been divided into five major divisions. These are:

- (1) Nuclear Characteristics
- (2) Chemical Properties
- (3) Physical Properties
- (4) Production Method, Availability and Cost
- (5) Annotated Bibliography

The nine radioisotopes studied are arranged somewhat arbitrarily in order of ascending atomic weight. The data presented in each section are considered to be the best available in the unclassified literature. In some cases where no unclassified information was available for an element or compound, properties for a similar material were substituted. This is perhaps notable for Pu-238. Because of the necessity of these substitutions the reader is cautioned that these values are at best approximations and should be used with that fact in mind. In these cases if an exact value is needed it is mandatory that the classified literature be reviewed.

Articles and reports presented in each section's annotated bibliography are arranged in chronological order beginning with the most recent. In some cases double parentheses have been placed around various comments not present in the original article or report but added for clarification or explanation. Section 3.10 presents references that give detailed or extensive information on a number of isotopes in the bibliographies and so are placed there for general examination.

## 2.4 Explanation of Terms

### 2.4.1 Neutron Radiation

2.4.1.1 Neutrons from spontaneous fission: - A continuous spectrum of neutrons varying in energy from approximately 0.1 to

18 Mev is emitted in the spontaneous-fission process. An average of two to four neutrons are emitted per fission, depending on the fissioning isotopes.

2.4.1.2  $\alpha$ -neutrons: - Neutrons are produced from mixtures of alpha emitters and certain elements, including most of the light elements. The energy spectrum of the neutrons is a continuous distribution extending from very low energies up to a maximum energy that is slightly less than the sum of the alpha energy and the energy liberated in the reaction. The yield of neutrons varies with the target elements and the composition of the mixture. Beryllium produces the most neutrons per alpha. Aluminum, carbon and oxygen produce progressively fewer neutrons.

It is conservative to use the maximum neutron energy to calculate shielding and radiation levels if the spectrum is not known. Approximately  $5 \times 10^6$  neutrons/sec/curie with neutron energy up to 11 Mev are emitted from mixtures of beryllium and alpha emitters. The yields and energy increase with alpha energy. Yields and maximum neutron energies are lower for aluminum, oxygen and carbon.

2.4.1.3 Photoneutrons: - A photon (see below) can eject a neutron when its energy exceeds the neutron binding energy. A photon energy greater than 7 Mev is required for all isotopes except deuterium, Be-9, C-14 and Li-6. Cross sections for the reaction are of the order of 1 mb and the neutrons generally have energy of 0.1 - 1 Mev.

2.4.1.4 n-2n neutrons: - If the bombarding neutron has sufficient energy, it is possible for the compound nucleus to emit two neutrons. The threshold energy is relatively high for all isotopes except deuterium and Be-9. Thus, beryllium, which may be used as a heat shield in space, may serve to multiply the neutron flux though, usually, only to a slight degree.

2.4.1.5 Neutron scattering and attenuation: - Most materials elastically and inelastically scatter neutrons through relatively large angles. In general, neutrons scatter more readily than gamma rays. Scattering at large angles is most predominant with heavier elements.

## 2.4.2 Gamma Radiation

2.4.2.1 Prompt gammas from spontaneous fission: - A continuous spectrum of gamma rays, varying in energy from 0.1 to 8 Mev, is

emitted in the process of spontaneous fission. Approximately 7.5 Mev is emitted in this fashion per fission.

2.4.2.2 Fission-product decay gammas: - Fission products formed in spontaneous fission ultimately emit a total of  $\sim 5.5$  Mev in gamma rays per fission. These gamma rays generally have lower energy than the prompt gammas from spontaneous fission. The fission-product gammas approach a steady state decay rate within a few hours after a spontaneously fissioning nuclide is isolated.

2.4.2.3 Inelastic gammas: - Inelastic scattering of neutrons, the predominant process of slowing down in the heavier elements, is accompanied by the emission of photons. The total photon energy is less than the incident neutron energy. Ordinarily the energy absorbed from the neutron is emitted as several photons.

2.4.2.4 Neutron-capture gammas: - Essentially all isotopes, with the exception of Li-6 and B-10, emit energetic gamma rays upon the capture of a neutron. This gamma radiation is especially important for those elements with high neutron capture cross sections since these elements compete most strongly for the available neutrons and are therefore strong gamma ray emitters.

One must know the neutron flux and spectrum in a shield to calculate inelastic and capture gammas.

2.4.2.5 Decay gammas: - Characteristic gamma rays are emitted in the decay of most radioisotopes; alpha, beta and positron decay and electron capture generally leave the product nucleus in an excited state, which subsequently decays to the ground state with the emission of one or more photons. These gamma rays vary widely in energy and abundance from one isotope to another. For example, abundant 1 to 2 Mev gamma rays accompany decay of Co-60 or Ce-144, while there is little gamma accompanying the decay of Pm-147.

2.4.2.6 Bremsstrahlung: - Continuous X-rays, or bremsstrahlung, are produced when high-energy charged particles decelerate in the atomic electric field. The order of 1 to 50% of the electron energy may be emitted as photons through this process. The gamma energy and differential spectrum increase approximately as the product of the electron energy and the average atomic number of the absorber. A few of the photons approach the energy of the incident electron with the majority having lower energy. The energy release and yield of photons in a given energy range for an alpha particle incident on a target are of

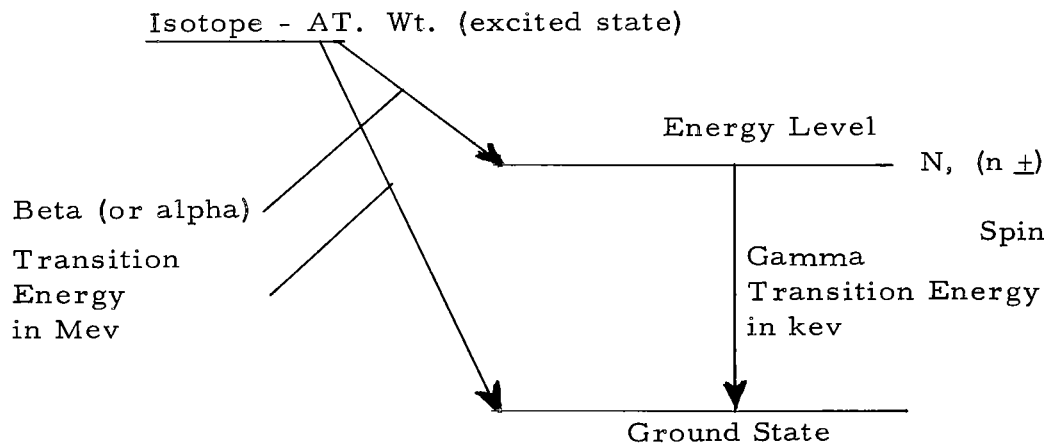


the order of a million less than for a beta particle of comparable energy. For this reason, bremsstrahlung from alpha emitters ordinarily is not significant. For the isotopes of interest, bremsstrahlung is most abundant and energetic when energetic beta particles, such as those accompanying decay of Sr-Y-90, interact with heavy elements.

2.4.2.7 Characteristic X-rays: - Produced following photoelectric interaction of gamma rays, or other processes in which the atomic electrons are disturbed. These X-rays are, in general, very soft and are of negligible consequence in shield design.

2.4.2.8 Gamma scattering and attenuation: - Of the three main ways gamma rays interact with matter (photoelectric effect, pair production and Compton scattering) two always result in secondary gammas. In pair production, the dominant effect for high-energy photons and elements of high atomic number, the subsequent annihilation of the positron, yields two 0.5 Mev photons. In the Compton effect the scattering of the photon by the electron merely decreases the photon's energy and changes its directions. This effect causes build-up of low-energy photons in shields and is most important for shielding materials of low atomic number.

#### 2.4.3 Explanation of Decay Scheme Notation



### 3.1 STRONTIUM-90, YTTRIUM-90

#### 3.1.1 Nuclear Characteristics

		Sr-90	Y-90
3.1.1.1	Atomic Number	38	39
3.1.1.2	Atomic Weight	90	90
3.1.1.3	Half Life		

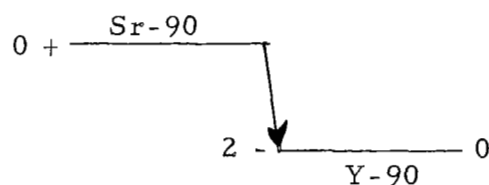
27.7 years

Reference 1

#### 3.1.1.4 Energy Levels and Decay Scheme

Sr-90  $\xrightarrow{\beta}$  Y-90 Reference 2

$\beta$ , 0.54 Mev (100%)

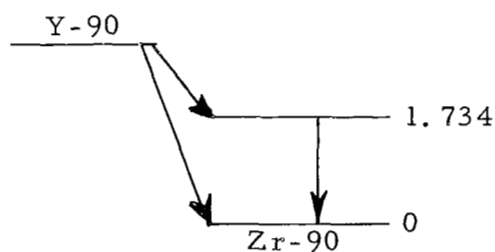


Reference 3

Y-90  $\longrightarrow$  Zr-90 Reference 4

$\beta$ , 0.446 Mev ( 0.02%)  
2.18 Mev (99.9+%)

$\gamma$  1.734 Mev



Reference 4

### 3. 1. 1. 5 Other Sources of Radiation - Bremsstrahlung

Although the gamma yield from Sr-Y-90 is negligible, considerable shielding is required to absorb the bremsstrahlung produced as the high energy betas are slowed down. Most of the bremsstrahlung are generated when the beta particles are slowed down in the compound or mixture of which the fuel pellet is made. A smaller number are generated by the betas which escape the pellet and are slowed down in the cladding material.

To allow accurate evaluation, a bremsstrahlung spectrum for Sr-Y-90 has been calculated (Ref. 5) and is given in Table I. This spectrum was used to calculate dose rates which were compared with results measured (Ref. 6) by Oak Ridge National Laboratories. The comparison of the calculated and measured results are extremely good. A complete description of this comparison is given in Reference 5.

Table I (Ref. 5)

#### Grouped Bremsstrahlung Spectral Distribution of Sr-Y-90

Energy Group (Mev)	Number of Gammas per Disintegration
2.2 - 1.98	$7.42 \times 10^{-7}$
1.98 - 1.76	$6.75 \times 10^{-6}$
1.76 - 1.54	$2.89 \times 10^{-5}$
1.54 - 1.32	$8.51 \times 10^{-5}$
1.32 - 1.10	$2.09 \times 10^{-4}$
1.10 - 0.88	$4.37 \times 10^{-4}$
0.88 - 0.66	$8.87 \times 10^{-4}$
0.66 - 0.44	$1.82 \times 10^{-3}$
0.44 - 0.22	$4.19 \times 10^{-3}$
0.22 - 0	$1.65 \times 10^{-2}$

### 3. 1. 2 Chemical Characteristics

#### 3. 1. 2. 1 Fuel Forms

Possible strontium fuel forms are listed in Table II. Where possible the melting points of the compounds are listed.

Because the power density and specific power for the various compounds depend on the compound density and the percentage of strontium to the total compound weight, these figures are included for each fuel form.

Table II (Ref. 7)

Strontium Compounds

Compound	Molecular weight	Wt. % metal	Density, g/cm <sup>3</sup>	Specific power watts/g of compound	Power density watts/cm <sup>3</sup> of compound	Melting point °C
Sr	87.62	100.0	2.6	0.536	1.39	
SrO	103.62	84.56	4.70	0.453	2.13	2430
SrF <sub>2</sub>	125.62	69.75	4.24	0.374	1.59	
SrTiO <sub>3</sub>	183.52	47.74	5.12	0.256	1.31	
SrB <sub>6</sub>	152.49	57.46	3.42	0.308	1.05	
SrC <sub>2</sub>	111.64	78.48	3.26	0.421	1.37	
SrSi <sub>2</sub>	143.79	60.94	3.20	0.327	1.05	
SrSi	115.71	75.72	3.47	0.406	1.41	

It should be kept in mind that the specific power and power density figures are based on theoretical maximums for density.

### 3.1.2.2 Material Compatibility

Since the low power density of Sr-90 limits its use to comparatively low temperature heat source applications, usually less than 1000°C, the likelihood of chemical reaction between a strontium compound and container is small. In general, the simple salts such as the chloride and nitrate are more chemically corrosive than compounds such as the titanate, molybdate, and zirconate. The oxide is the anhydride of Sr(OH)<sub>2</sub> and very readily dissolves in water as do the chloride and nitrate.

No difficulty is anticipated as a result of valence change. The intermediate element, yttrium, has a normal valence of 3 which might result in a small excess of yttrium. The final decay element, zirconium, is identical in valence with strontium.

### 3. 1. 2. 3      Effects of Impurities on Nuclear Characteristics

The amount of Sr-90 in the fuel compound is relatively small. Reference 8, page 6 indicates a value of 50%. In addition to the other isotopes of strontium, a normal Sr-90 fuel slug contains trace quantities of various fission products which cannot be removed during processing. The following values were obtained from Reference 8.

Cerium-144	$3 \times 10^{-3}$ C/C* of Sr-90
Ruthenium-106	$6 \times 10^{-4}$ C/C of Sr-90
Zirconium and Niobium-95	$8 \times 10^{-4}$ C/C of Sr-90
Yttrium-91	$4 \times 10^{-2}$ C/C of Sr-90

Not only are these quantities small, but their radiation intensity as compared to the bremsstrahlung radiation produced by Sr-90 and its daughter product Y-90 is negligible and are not considered in the shielding calculations. The radiation from other isotopes of strontium, namely Sr-89, need not be considered either since its concentration should be quite small after storage for several half lives.

### 3. 1. 3      Thermal and Physical Properties

The physical properties of the Sr-90 fuel forms vary widely depending on the amount of impurities within the source, the attainable density of the fuel form, the "age" of the source material and the energy of the beta particles. Because of the possible variations with some of the aforementioned quantities a set of values will be presented as the "best" values for the weight density, energy of beta particles, "age" of the source and estimated isotope purity for the two fuel forms of interest and the power density and specific power will be calculated. If other information is available for the weight density, energy of the beta particles or the purity of the isotope the power density and the specific power can be easily calculated by following the set of calculations presented below. The following assumptions were made for the calculation of specific power and power density.

---

\*C/C = curies of contaminant per curie of Sr-90.

1. The strontium, yttrium are in equilibrium, i. e., for every beta emitted by Sr-90, there is a beta emitted by Y-90.
2. The initial Sr-90 source purity is 50%.
3. The strontium source is sufficiently "aged" so that the only radioactive sources are Sr-90 and daughter products.
4. The attainable densities of the fuel compounds are  $3.7 \frac{\text{gm}}{\text{cm}^3}$  for SrO and  $3.93 \frac{\text{gm}}{\text{cm}^3}$  for SrTiO<sub>3</sub>. (The theoretical values are higher.)
5. The maximum energy of the beta particles is 0.54 Mev for Sr-90 and 2.26 Mev for Y-90. The average beta energy per disintegration for Sr-90 is 0.17 Mev and for Y-90 is 0.925 (Ref. 10, page 698). The remaining energy given off per disintegration (0.37 Mev for Sr-90 and 1.335 Mev for Y-90) appears as highly penetrating particles (Ref. 11, page 539-540).
6. The half life of Sr-90 is 27.7 years.

Using the above assumptions, the number of curies per gram of pure Sr-90 (not including the Y-90) is

$$\frac{.693}{27.7 \times 3.16 \times 10^{10}} \times \frac{.602 \times 10^{24}}{90} = 142 \frac{\text{curies}}{\text{gm (Sr-90)}}$$

The power density of the SrO is

$$\begin{aligned}
 & 142 \left[ \frac{\text{curies}}{\text{gm Sr-90}} \right] \times \left\{ .17 \left[ \frac{\text{Mev}}{\text{dis Sr-90}} \right] + .925 \left[ \frac{\text{Mev}}{\text{dis Y-90}} \right] \right\} \\
 & \times 3.7 \times 10^{10} \left[ \frac{\text{dis}}{\text{sec}} \right] \times 1.603 \times 10^{-13} \left[ \frac{\text{watt}}{\text{Mev}} \right] \\
 & \times .5 \left[ \frac{\text{purity of}}{\text{Sr source}} \right] \times \left[ \frac{90}{106} \frac{\text{gm Sr}}{\text{gm SrO}} \right] \times 3.7 \left[ \frac{\text{gm SrO}}{\text{cm}^3} \right] = 1.45 \frac{\text{watts}}{\text{cm}^3 \text{ SrO}}
 \end{aligned}$$

$$\left\{ .17 \left[ \frac{\text{Mev}}{\text{dis Sr-90}} \right] + .925 \left[ \frac{\text{Mev}}{\text{dis Y-90}} \right] \right\} \times 3.7 \times 10^{10} \left[ \frac{\text{dis}}{\text{sec}} \right] \frac{1}{\text{curie}}$$

$$\times 1.603 \times 10^{-13} \left[ \frac{\text{watt}}{\text{Mev}} \right] \frac{1}{\text{sec}} = 6.5 \times 10^{-3} \left[ \frac{\text{watt}}{\text{curie}} \right] \text{ or } 6.5 \left[ \frac{\text{watt}}{\text{K curie}} \right]$$

By a similar calculation the power density for SrTiO<sub>3</sub> is 0.88  $\frac{\text{watts}}{\text{cm}^3}$ .

A complete listing of the thermal and physical properties of Sr-90, SrO, and SrTiO<sub>3</sub> is presented in Table III.

Table III

Thermal and Physical Properties

		<u>Sr</u>	<u>SrO</u>	<u>SrTiO<sub>3</sub></u>
Thermal Conductivity	$\frac{\text{cal cm}}{\text{sec}^\circ\text{C cm}^2}$	0.024(a) Ref. 12	0.017(b) Ref. 13	0.0128 Ref. 14
Specific Power	$\frac{\text{watts}}{\text{gm}}$	0.95 Ref. 15	0.80 Ref. 12	0.23 Ref. 15
Thermal Output	$\frac{\text{watts}}{\text{kilo curie}}$	6.5(d) Ref. 4	6.5(d) Ref. 4	6.5(d) Ref. 4
Heat Capacity	$\frac{\text{cal}}{\text{gm}^\circ\text{C}}$	0.176(e) Ref. 12	0.0847(f) Ref. 16	0.165(g) Ref. 17
Heat of Fusion	$\frac{\text{cal}}{\text{mole}}$	230 Ref. 18	-	-
Weight Density	$\frac{\text{gm}}{\text{cm}^3}$	2.63 Ref. 12	3.7 Ref. 19	3.93(c) Ref. 19
Melting Point	$^\circ\text{C}$	771 $\pm$ 1 Ref. 12	2430 Ref. 12	1660 Ref. 12
Boiling Point	$^\circ\text{C}$	1380	-	-
Specific Activity	$\frac{\text{kilo curie}}{\text{gm Sr-90}}$	0.142(h) Ref. 15	0.142(h) Ref. 15	0.142(h) Ref. 15

## Key to Thermal and Physical Properties (Table III).

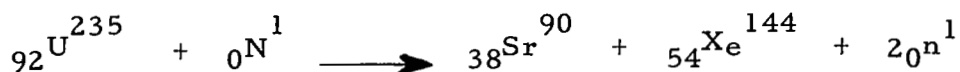
- a. Value for Y-90.
- b. The values stated are those for CaO since none could be found for SrO. The value stated is that at 970°C. Other values are 0.0185 at 560°C and 0.0265 at 140°C.
- c. The percentage of active isotope in the compound is 24%.
- d. Calculated value, see write up preceding table of physical properties.
- e. Value at 20°C.
- f. The value at 1727°C. Other values of the specific heat ( $C_p$ ) may be obtained by using the formula given in ref. 17 and given here in units of  $\frac{\text{calories}}{\text{mole } ^\circ\text{C}}$  to convert to  $\frac{\text{calories}}{\text{gm } ^\circ\text{C}}$  divide by the atomic weight.  
$$C_p = 8.69 + 0.16 \times 10^{-3} T - 0.76 \times 10^{-5} T^2$$
where  
$$298^\circ\text{K} < T < 2000^\circ\text{K}$$
- g. The value at 1115°C. Other values are .158 at 560°C and .143 at 140°C.
- h. Pure isotope.

### 3.1.4 Isotope Production, Availability and Cost

#### 3.1.4.1 Production Method

Sr-90 is produced by the fission of heavy elements, i. e., those elements heavier than lead. The fission may result from neutron bombardment or spontaneous decomposition of the nucleus. A typical fission reaction is:





Sr-90 is recovered by processing spent fuel elements from nuclear reactors. The fission products are separated from the acid solution containing the dissolved fuel elements after the unused fuel is recovered for recycling into the reactor. The various elements are separated from the fission product mixture. The strontium fraction contains the isotopes Sr-86, and Sr-88 (both of which are stable), Sr-89 (which has a half life of 50 days and is an emitter of very hard beta energy) and Sr-90. As recovered, the mixture of strontium isotopes should contain about 50% Sr-90 (Reference 8). A cooling period is generally allowed before final processing to permit the decay of the comparatively short-lived Sr-89. The Sr-88 is inert and remains with the active isotope as a diluent.

The breakdown of the strontium fraction of the fission product mixture thirty days after reactor discharge is (reference 20).

Sr-90	49.6%
Sr-89	13.4%
Sr-88	37.0%

#### 3.1.4.2 Availability and Costs

The prospective production level of Sr-90 as published by the Division of Isotope Development, USAEC, in January 1963 are presented below:

	FISCAL YEAR							
	<u>1964</u>	<u>1965</u>	<u>1966</u>	<u>1967</u>	<u>1968</u>	<u>1969</u>	<u>1970</u>	<u>1980</u>
Mega Curies	5	5	5	5	10	10	10	10
Kw (t)	32.5	32.5	32.5	32.5	65	65	65	65

The present cost of Sr-90 is approximately \$1.00 per curie and is expected to decrease to \$0.123 per curie at Hanford plant at a yearly production rate of 10 Mc. The above production rates are based on government facilities only. It is expected that by the year 1970 private power will make available 94 Kw (t) per year, and by 1980, close to 630 Kw (t) per year will be available (Reference 21).

## REFERENCES

1. The Half Life of Strontium-90, Canadian Journal of Physics 33, 133-7, March to April 1955.
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3. Table of Isotopes, Reviews of Mod. Physics 30, 2, II, April 1958.
4. Final Report, Radioisotope Evaluation Program MNSP-MK-5002, NASA-NAS 5-3879, Dec. 23, 1964.
5. Shielding Kilocurie Amounts of Strontium-90, MND-P-2529, A. M. Spamer, Martin Co. (March 1961).
6. Bremsstrahlung Absorption Measurements from  $\text{SrTiO}_3$ , ORNL-CF-61-1-25. J. A. Butler and E. E. Pierce.
7. Private Communication: S. J. Rimshaw, Oak Ridge National Laboratories, Sept. 1965.
8. Radioisotopic Heat Sources, HW-76323.
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10. Radiation Dosimetry, Brownell Hine, Academic Press, Inc., 1956.
11. The Atomic Nucleus, Evans, McGraw Hill Book Co., 1955.
12. Isotopic Power Sources, A Compendium MND-P-2581-3.
13. Thermal Conductivity, Journal American Ceramic Soc. Vol. 37, p. 107-110, 1954.
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17. High Temp Heat Contents of Metal and Orthotitanates of Barium and Strontium. J. P. Coughlin, R. E. Lorenson, Am. Chem. Soc. Vol. 75, 1953, pg. 530-1.
18. Nuclear Engineering Handbook, Etherington, McGraw Hill Book Co., 1958.
19. ORNL-3576 Handbook of Shielding Requirements and Radiation Characteristics of Isotopic Power Sources for Terrestrial, Marine, and Space Applications, E. D. Arnold, 1964.
20. Atomic Energy Waste, E. Gluechauf, Interscience Publishers, Inc., New York, 1961, pg. 41-46.
21. Radionuclide Power for Space, Vol. 21, No. 3 - March 1963.

### 3.1.5 Annotated Bibliography

- 3.1.5.1 ORNL-3802. REVIEW OF RADIOISOTOPES PROGRAM, 1964. J. H. Gillette, Ingrid B. Lane, comp. (Oak Ridge National Laboratories, Tennessee). May 1965. Contract W-7405-eng-26. 71 p.

Reactor-produced and cyclotron-produced isotopes, and radioisotope generators are discussed. Measurements of the half life of Cs-137 are reported. Calculations were made on the  $(n, \gamma)$  burnup of Pm-146 in Pm-147. Several computer programs for radioisotope calculations are described. The use of cesium tetraoxalate in Cs-137 processing is described briefly. Cooperative tests of a Co-powered thruster and a Sr-90 powered thermoelectric unit are also described. The preparation of Tc-99, Am-241, 242, Cm-BE, Ir-192 - ceramic, Pm-147, P-32, Sr-90, and other sources was studied.

- 3.1.5.2 HALF-LIVES OF Sr-90 and Cs-137. K. F. Flynn, L. E. Glendenin, A. L. Harkness, and E. P. Steinberg (Argonne National Laboratories, Lemont, Illinois). J. Inorg. Nucl. Chem. 27:21-3 Jan. 1965.

The half lives of Sr-90 and Cs-137 were measured both by specific activity and by direct decay over periods of 6 to 10 years. Results from the decay measurements are  $28.0 \pm 0.4$  years for Sr-90 and  $29.9 \pm 0.5$  years for Cs-137. Recommended "best" values for these half lives based on current literature values are  $28.1 \pm 0.3$  years for Sr-90 and  $30.0 \pm 0.2$  years for Cs-137.

- 3.1.5.3 A RADIOMETRIC METHOD FOR THE ISOTOPIC ANALYSIS OF MIXTURES OF Ce-141 and Ce-144, Sr-89 and Sr-90 IN SAMPLES OF LOW TOTAL ACTIVITY. S. V. Belov and N. V. Sidorov. Zh. Analit. Khim. 19: 1049-52 (1964) (In Russian)

A method is described for the isotopic analysis of mixtures of Ce-141 and Ce-144 and Sr-89 and Sr-90 using a scintillating beta spectrometer and 4 pi counter. Rigid spectra of Ce-144  $\longrightarrow$  Pr-144 and Sr-90  $\longrightarrow$  Y-90 correspondingly. When samples of low total activity (about 1000 disintegrations per minute) are analyzed the method suggested has considerable advantages over the method of analysis of absorption curves in aluminum filters (the beta particles recording is more effective and the results obtained are more accurate).

3. 1. 5. 4     LARGE SOURCES OF BETA RADIATION FOR CHEMICAL PROCESSING. Isotopes Radiation Technology, 2:158-69 (Winter 1964-65).

The development and potential applications of beta-emitting isotopes for chemical processing are reviewed. Vitreous enamels were studied as source bearers for Ce-144, Pm-147, and Sr-90. It was concluded that some of these formulations are more resistant to chemical attack than Pyrex glass and can withstand considerable thermal and mechanical shocks. Beta-induced grafting of methacrylic acid-styrene copolymer on polyethylene was studied. Some beta irradiators were designed, and their uses are discussed.

3. 1. 5. 5     SHAPES OF ALLOWED AND UNIQUE FIRST-FORBIDDEN BETA-RAY SPECTRA: In-114, K-42, Rb-86, Sr-90, and Y-90. H. Daniel, G. Th Kaschl, H. Schmitt, and K. Springer (Max-Planck-Institut fur Kernphysik, Heidelberg). Phys. Rev., 136: B1240-2 (Dec. 7, 1964).

The beta spectra of In-114, K-42, Rb-86, Sr-90, and Y-90 (ground-state transitions) were measured with a double-lens spectrometer. The allowed spectrum of In-114 was found to have a purely statistical shape; the coefficient  $b$  of a possible  $b/W$  term turned out to be  $0.5 \pm 2.2 \times 10^{-2} \text{ mc}^2$ . The unique first-forbidden spectra of K-42, Rb-86, Sr-90, and Y-90 were found to show very small but definite deviations from the simple unique shape.

3. 1. 5. 6     BREMSSTRAHLUNG FROM Sr-90, Y-90 IN THIN ABSORBERS. Thomas S. Bustard (Martin Marietta Corp., Baltimore, Maryland), Joseph Silverman (University of Maryland, College Park, Maryland). Paper presented at the Symposium on Low Energy and Gamma Sources and Applications. Oct. 20, 21, 1964, Chicago, Illinois. Sponsored by AEC.

The bremsstrahlung yields obtained from targets ranging in atomic number from 13 to 73 using a thin, point source of Sr-90, Y-90 are presented. A novel method of obtaining these yields was used in that the bremsstrahlung spectra are measured as they are built up and attenuated through targets whose mass thicknesses are less than the range of the Y-90 maximum energy beta particle. The resulting bremsstrahlung yields are then compared to those calculable from the Evans\* approximation to thick target theory.

\*The Atomic Nucleus, Evans, R. D. McGraw Hill, 1955

3. 1. 5. 7     ISOTOPIC POWER DATA SHEETS.     Compiled by S. J. Rimshaw (Oak Ridge National Laboratory, Oak Ridge, Tennessee).     For presentation at Industry Information Meeting on Isotopic Power Development and Applications, Washington, D. C.     May 18-19, 1964.

## INTRODUCTION

Nuclear and physical properties are listed of some radioactive materials having potential application to terrestrial, marine, and aerospace power requirements.     The data here are limited to unclassified source materials of Co-60, Sr-90, Cs-137, Ce-144, Pm-147, Cm-242, and Cm-244.     It will be obvious from reviewing the data sheets that experimental information on the properties of radioactive materials is very incomplete.     Where such information is not available, literature values derived from the inactive materials or related materials have been used to indicate the probable order of magnitude of the value.

Future data compilations will include more isotopes and materials.     The information will be presented in the form of a manual or handbook in order to increase the space available for presenting and evaluating the data that are available.     Provisions will be made for additions or deletions of data to keep the information current and meaningful.

Source Material	SrTiO <sub>3</sub>
Half-life	Sr-90 - 27.7 years (Y-90 - 64.2 hrs)
Decay and Radiation Properties	Sr-90 $\longrightarrow$ Y-90 100% beta - 0.54 Mev Y-90 $\longrightarrow$ Zr-90 (stable) 100% beta - 2.26 Mev
Isotopic Composition	55% Sr-90, 43.9% Sr-88, and 1.1% Sr-86
Activity Concentration	33 curies of Sr-90 per gram of fission product SrTiO <sub>3</sub>

Radiochemical Purity	> 99.9% Sr-90 + Sr-89. Sr-89 is present in concentrations dependent on length of time since discharge from the reactor and is usually < 5% of the Sr-90 activity at time of fabrication of source pellets. Other radiochemical impurities such as Ce-144 can be neglected.
Chemical Purity	> 95% strontium. Major impurities are Ca and Ba.
Specific Power	0.223 watts per gram of SrTiO <sub>3</sub> , or 6.772 watts per kilocurie of Sr-90.
Thermal Energy	148 curies Sr-90 per thermal watt
Density	~ 3.7 g/cm <sup>3</sup> . The theoretical density is 5.0 g/cm <sup>3</sup> . Production values vary from 3.2 to 4.2 g/cm <sup>3</sup> , averaging about 3.7 g/cm <sup>3</sup> .
Power Density	0.825 watts/cm <sup>3</sup> from Sr-90 at a density of 3.7 g/cm <sup>3</sup> . Sr-89 will contribute in proportion to its concentration at a rate of 3.4 watts/1000 curies of Sr-89.
Thermal Conductivity	Values reported for inactive SrTiO <sub>3</sub> vary from 0.0132 to 0.0173 cal/sec cm °C at room temperature, depending on the density.
Coefficient of Expansion	$1.12 \times 10^{-5} \text{ }^{\circ}\text{C}^{-1}$
Melting Point	~ 1900°C
Mechanical Strength	Fair
Thermal and Radiation Stability	The thermal stability is good. The radiation stability is good as exhibited in two-year old samples.

Radiation Attenuation	Dose Rate, rads/hr at 100 cm	Shielding in Centimeters of Uranium for a Sr-90 Source Strength of		
		100 w	1000 w	10,000 w
	100	0.07	0.7	2.0
	10	1.0	2.3	3.8
	1	2.6	4.3	5.8
	0.1	4.4	6.2	8.0

Gas Evolution Due to  
Radioactive Decay  
Processes

None

Leach Rate

Inactive  $\text{SrTiO}_3$  leaches at a rate of 1 microgram/day  $\text{cm}^2$  in sea water. Tests on 18-month old pellets of fission product  $\text{SrTiO}_3$  indicate a leach rate of 1 mg/day  $\text{cm}^2$ .

Vapor Pressure

No data available

Resistance to Thermal  
Shock

No data available

Burnup Characteristics

Dispersibility poor

Capsule Compatibility

Excellent with usual capsule materials such as stainless steel and Hastelloy "C".

((This reference is reproduced in its entirety))

- 3.1.5.8 NP-14698. K VOPROSU OB OTNOSITEL'NYKH IZMERENIYAKH AKTIVNOSTI ISTOCHNIKOV BETA-IZLUCHENIYA. (Relative Measurements of Beta-Source Activities.) A. I. Denisikov (Gosurdarstvennyi Komitet po Ispol'zovaniyu Atomnoi Energii SSSR). (1963) 4 p.

Beta source activity or specific beta activity of solutions were measured by a relative measurement method (method of comparing the source activity with the standard activity of the same isotopic content). Curves of the counting rate as a function of aluminum filter thickness for thin sources, including Co-60 and Sr-90 + Y-90 sources, were evaluated.



3. 1. 5. 9 (ORNL-TM-198) ISOTOPES DIVISION QUARTERLY REPORT, OCTOBER-DECEMBER 1961. J. H. Gillette (Oak Ridge National Laboratories, Tennessee). June 5, 1962. Contract (W-7405-eng-26). 39 p.

Progress is reported on developments in the preparation, properties, and measurements of radioisotopes, radioactive sources, calutrons, and thermal diffusion studies in radioactive gases (B. O. G.). The feasibility of preparing a Sr-90 heat source, of  $8 \times 10^5$  curies, was studied. Information concerning the status and progress of engineering development is given.

3. 1. 5. 10 (CEA-tr-X-355) MESURE DES RAYONS PAR L'EMPLOI DES COMPOSES ORGANIQUES FLUORESCENTS. I. MESURE DU SPECTRE DE  $\beta$  A L'AIDE DES MONOCRISTAUX D'ANTHRACENE. (Measurement of Beta Rays by Use of Fluorescent Organic Compounds. I Measurement of  $\beta$  Spectra Using Anthracene Monocrystals.) T. Watanabe. Translated into French from Oyo Butsuri, 28; 377-80 (1959). 12 p.

It has been reported that the light pulse produced by mono-energetic electrons impinging on anthracene crystals is proportional to the electron energy within its limited range. In the present work, the response of a large single crystal of anthracene to impinging beta rays emitted by radioactive substances is discussed. The crystal used is of cylindrical shape, approximately 2.5 cm in diameter and 3.0 cm in height. It is glued with silicon grease to the photocathode of the Dumont 6292 photomultiplier tube. Output signals are proportionally amplified by an A-1 type linear amplifier and are analyzed by a single channel pulse analyzer. Beta spectra from pure beta emitting substances, such as S-35, RaD, RaE, RaF, or Sr-90 + Y-90, are observed and compared with that obtained by a lens type magnetic beta spectrometer with the result that the two spectra are similar except in the low energy part where the intensity of the former is higher. This seems attributable to both the beta bremsstrahlung and the imperfection of the crystal which was colored and opaque. The relation between the maximum energy of the beta rays and the pulse height is of good linearity from 0.16 to 2.2 Mev electron energy.

3. 5. 1. 11 ON THE BETA SPECTRUM AND DISINTEGRATION ENERGY OF Sr-90 and Y-90. Vaino Hovi. Ann. Acad. Sci. Fennicae Ser. A VI, No. 21, 1-12 (1959)

Values for the mean energy,  $\overline{E}_{\text{beta}}$  of the beta particles of the isotopes Sr-90 and Y-90 were determined by using different approximations of the shape correction factor and available spectroscopic data. It is shown that in the case of Sr-90 the theoretical mean energies corresponding to different approximations of the shape correction factor differ so much from each other that by using modern micro-calorimetric techniques at low temperatures it would be possible to get an independent verification of the shape factor. This might be important for clearing up the actual form of the interaction in beta-decay. In the case of Y-90, because of the small relative differences between the values of  $\overline{E}_{\text{beta}}$ , independent calorimetric verification could hardly lead to as significant results. ((Detailed information on decay energy was obtained from Mr. T. Bustard, Hittman Associates, Baltimore, Maryland.))

3.1.5.12 NACHWEIS DER POLARISATION DER BETA TEILCHEN VON Sr-90 + Y-90 by Joachim Heintze, Zeitschrift Fur Physik 148, 560-3, 1957. ((The entire article is in German and no attempt at translation has been made.))

3.1.5.13 M6472 STRONTIUM-90 BIBLIOGRAPHY. Braden, C. H., L. Slack, and F. B. Spull, Dec. 31, 1957, 10 p.

The following sources were checked for references relating to Sr-90: Chemical Abstracts, Nuclear Science Abstracts, and Library files. ((All applicable references contained in this document are contained elsewhere in this section.))

3.1.5.14 DESIGN AND PERFORMANCE OF A THIN MAGNETIC LENS BETA RAY SPECTROMETER. T. D. Nainan, H. E. Deware, and Ambui Mukerji (Tata Institute of Fundamental Research, Bombay, India). Proceedings of the Indian Academy of Science, 44A, 111-122, 1956

Construction of a thin magnetic lens beta ray spectrometer is described. A baffle system which takes advantage of the ring focus has been designed on the basis of the calculations of electron projectory limited to an angle of acceptance of 9-10.5°. The results of the studies of the beta ray spectra of Sr-90 and Y-90, Pm-147, Tm-170, are presented.

((Source purity: Mixture of Sr-89, 90. Sr-89 allowed to die down to negligible intensity. Sample measured on thin backing of mica. The measured beta spectrum is as follows:  $E_{\text{beta max}} = 0.541 \pm 0.008$  Mev.))

3.1.5.15 FORBIDDEN BETA SPECTRA OF Rb-86, S-90, Y-90,  
AND Tl-204. E. E. Berlovick, D. M. Kahai, and A. V.  
Savateev. Izvestia, Akad, Nauk, 20-275-88, March 1956

In solving the problem of beta interaction an important part is played by elucidation of the forbidden beta spectra. Such investigations are also of interest in connection with the study of the nuclear shell model. In the present work we studied the shape of the forbidden spectra of R-86, S-90, and Tl-204. All these nuclei have an even mass number and for them the transitions to the ground state of the daughter nucleus occur between even-even and odd-odd nuclei, or vice-versa.

The radioactive isotope Sr-90 with a half life of 19.9 years\* is one of the members of the radioactive chain beginning with Br-90 that is produced by the bombardment of U-235 with slow neutrons. Sr-90 is separated chemically from the disintegration products together with Sr-89 (half life = 54.5 days) after decay of the latter, Sr-90 can be obtained together with its daughter Y-90 which is formed in the ground state. Y-90 with a half life of 2.54 days decays to the stable nucleus Zr-90 in the ground state. In our measurements we use samples of Sr-90 with a very high specific activity separated from the uranium disintegration products and cooled for a sufficient time to insure Sr-89 content under 1a. The source material was applied in the form of an ultra thin layer on a strip of aluminum foil one micron thick.

\*The currently accepted value for the half life of Sr-90 is 27.7 years.

((Cited as best value for decay energy in Rev. Mod. Phys. Vol. 30, #2 Pt. II, April 1958.))

3.1.5.16 THERMAL NEUTRON FISSION YIELDS AT MASSES 90  
AND 91. George W. Reed (Argonne National Laboratories,  
Lemont, Illinois). Physical Review 98, 1327 - 9 June 1955

Thermal neutron fission yields in U-235 have been determined for Sr-90 and Y-90. The fission yield at Sr-90 is 4.02 per cent based on a half-life of 20 years. The yield for Y-91, 5.35%, agrees with experimental error with that previously reported for Y-91. The apparent low yield at mass 90 is discussed. ((The discussion of the fission yield at Sr-90 is based on a 19.9 year half life. The presently accepted value is 27.7 years.))

- 3.1.5.17 THE HALF LIFE OF STRONTIUM-90. D. M. Wiles and R. H. Tomlinson (McMaster University, Hamilton, Ontario, Canada). Canadian Journal of Physics 33, 133-7, March to April 1955.

The value of  $27.7 \pm 0.4$  years has been obtained for the half life of Sr-90 from the absolute disintegration rate of a known number of atoms. The disintegration rate was measured with a 4 pi proportional counter and the number of atoms was determined with a mass spectrometer using isotope dilution.

((The half life determined by these workers is the currently accepted value.))

- 3.1.5.18 FISSION YIELDS OF STABLE AND LONG LIVED ISOTOPES OF CESIUM, RUBIDIUM, AND STRONTIUM IN NUCLEAR SHELL THEORY. Canad. Jour. of Phys. 31, 419, 1953

The relative yields of the isotopes of cesium, rubidium, and strontium from thermal neutron fission of U-235 have been determined mass spectrometrically. The cesium isotope yields are combined with those obtained previously for the xenon isotopes to give high precision yields for the mass change from 131 to 137. In this work, neutron capture reactions have been considered and corrections made where these take place to an appreciable extent. The results give further evidence of abnormal yields in the 82 neutron shell region. The half life of Cs-137 was determined and found to be  $33 \pm 2$  years. Cesium has been reported to have a half life from 33 to 37 years. The Cs-133 to 137 ratio will therefore increase with time. The data of Table 4 ((not shown)) may be used to calculate the half life of Cs-137. The results of these calculations are given in Table 5. It should be pointed out that some variation in the 133 mass yield may occur owing to neutron capture reaction, etc., which would introduce an error in the half life determination particularly for samples radiated at high flux values. ((A condensation of Table 5 is presented as follows.))

5 sample runs: results; 31, 31, 35, 35 years;  
average value =  $33 \pm 2$  years.

- 3.1.5.19 FURTHER STUDIES ON Sr-90 and Y-90. L. E. Glenderin and C. D. Coryell, Unclassified. NNES (IV) 9 Book 2, 687-91, Auth 1951, ANL.

Absorption curves of the Sr-90 and Y-90 radiations in aluminum indicate maximum beta energies of 0.6 Mev for Sr-90 and 2.2 Mev for Y-90. The half life of Sr-90 has been estimated from fission-yield considerations to be 24 to 30 years. The best values taken at 25 years. Based on Reports CC-529 and CC-1112 of 1943.

3.1.5.20 IDENTIFICATION OF Sr-90 and Y-90 in URANIUM FISSION. R. W. Nottorf, Unclassified NNES (IV) 9 Book 2, 682-6, 1951

The discovery of a long lived strontium activity and its shorter-lived yttrium daughter is reported. The yttrium isotope was identified with the known 60 hour Y-90 and its half life was shown to be 65 hours. Decay measurements of Sr-90 over 30 months indicated a period of  $23 \pm 3$  years.

Based on reports CC-521 and CC-725 of 1943.

3.1.5.21 FIRST-FORBIDDEN-BETA-SPECTRA AND THE BETA SPECTRUM OF STRONTIUM-90 - YTTRIUM-90. Al Jackson Laslett, E. N. Jensen, and A. Paskin (Institute for Atomic Research and Department of Physics, Iowa State College, Ames, Iowa). Physical Review 79, 412, 1950.

((Author states)) that a more adequately separated source of Sr-90 was obtained since our first work on this activity\*\*. Upper limit of 0.54 Mev (kinetic energy) is obtained for the Sr-90 beta spectrum ((the half life of Sr-90 is taken at 19.9 years in this article. Presently accepted half life for Sr-90 is 27.7 years)).

\*\* Phys. Rev. 75, 1949, 1949

3.1.5.22 THE HALF LIFE OF STRONTIUM-90. Ruth I. Powers and A. F. Voigt (Institute for Atomic Research and Chemistry Department, Iowa State College, Ames, Iowa). Physical Review 79, 175, 1950\*

The two samples used, after separation from cyclotron bombarded Uranyl nitrate, were purified by repeated carbonate precipitations and mounted as Strontium carbonate. In order not to measure any of the 55 day Sr-89 which was originally present, sample one was read with 746 mg/cm<sup>2</sup> of aluminum absorber while sample two was read with 714 mg/cm<sup>2</sup>. The decay of several samples which were prepared

have now been followed for seven years by means of a Lauritsen electroscope and the half life resulting from these measurements is sufficiently different from that currently used to warrant its publication at this time. The method of least squares was used to draw the best possible straight line through each set of data. From this a half life of 20.0 years was obtained with sample one while sample two gave 19.7 years. Therefore, the half life of Sr-90 may be taken as  $19.9 \pm 0.3$  years. ((This was the original experiment in which the half life of Sr-90 was determined to be 19.9 years. The presently accepted half life of Sr-90 is 27.7 years.))

\*Also published as government report No. AECU 840, 1 May 1950.

3. 1. 5. 23 THE FORBIDDEN BETA DECAY OF STRONTIUM-90 AND YTTRIUM-90. Charles H. Braden, Louis Slack, and Franklin D. Shull. (Washington University, St. Louis, Missouri.) Physical Review 75, 1964 - 5 June 1949

The isotope Sr-90 is a fission product which decays by beta emission to Y-90. The half life is  $8 \times 10^8$  seconds ((25.4 years)) and the energy release including rest mass is  $2.04 \text{ mc}^2$  (kinetic energy 5.71 kev). The daughter product Y-90 decays by beta emission to Zr-90. The half life of this transition is  $2.25 \times 10^5$  seconds and the energy release is  $5.40 \text{ mc}^2$  (kinetic energy 2.25 Mev). No gamma radiation is observed.

A carrier free sample of Sr-90 was obtained from Oak Ridge. This sample had been aged to permit an associated 55 day activity of Sr-89 to die out. Three spectrum were measured. One, Sr-90 and Y-90 in equilibrium together; two Sr-90 alone; and three, Y-90 alone. For the latter two runs, the isotopes were chemically separated by a method due to Kurbatov and Kurbatov. Samples were prepared for the spectrometer by evaporation from solution on thin zapon films following the insulin technique of Langer. The spectra were measured in the double focusing spectrometer described by Kuri, Osoba, and Slack.

3. 1. 5. 24 SHAPE OF THE BETA-SPECTRA OF STRONTIUM-90 AND YTTRIUM-90. E. N. Jensen and L. Jackson Laslett. (Institute for Atomic Research and Department of Physics, Iowa State College, Ames, Iowa.) Physical Review 75, 1949 - 1949.

The beta spectra of Sr-90 and Y-90 have been examined in this laboratory using the magnetic lens spectrometer previously described, and were found to exhibit a shape similar to that recently reported by Langer and Price for the forbidden transition of Y-91. The upper limits found for the Sr-90 and Y-90 spectra are  $2.05 \text{ mc}^2$  (0.53 Mev kinetic energy) and  $5.37 \text{ mc}^2$  (2.23 Mev kinetic energy), respectively. These values determined in light of the special form found for the spectra differed not inappreciably from those reported by Meyerhoff\* whose data also, however, give some indication of the same type of departure from the conventional spectral shape as that reported here.

\*Phys. Rev. 74, 621-2, Sept. 1948

- 3.1.5.25 ANGULAR CORRELATION OF SUCCESSIVE GAMMA-RAY QUANTA II. Edward L. Brady and Martin Deutsch. (Massachusetts Institute of Technology, Cambridge, Massachusetts.) Physical Review 74, 1541-2, November 1948. ((Article is concerned with the angular correlation observed by an improved method using scintillation counters for six radioactive substances. These substances were nickel, yttrium, magnesium, barium, strontium, and paladium.))
- 3.1.5.26 NOTES ON THE BETA-SPECTRA OF YTTRIUM-90 AND STRONTIUM-90. Walter E. Meyerhoff. (Department of Physics, University of Illinois, Urbana, Illinois.) Physical Review, 74, 621-2, September 1948.

The rather wide discrepancies in the literature values of the beta end point energy of Y-90 made it desirable to repeat the measurement of this energy. Since the Y-90 (60 hours), can be obtained in secular equilibrium with Sr-90 (25 years) in a practically carrier free form, the value for the beta end point energy of Sr-90 was obtained also.

Sources of the order of  $1 \text{ mg/cm}^2$  thickness were deposited on  $0.02 \text{ mg/cm}^2$  thick zapon film and mounted in a conventional  $180^\circ$  type spectrometer with a Neary slit type system. An end window geiger counter carrying a fixed slit which was covered with a  $0.8 \text{ mg/cm}^2$  mica window served as a detector. The resolution of the spectrometer was 3%. Curie plots of the beta spectra gave  $2.35 \pm 0.03 \text{ Mev}$  for Y-90 end point and  $0.61 \pm 0.01$  for the Sr-90 end point.

## 3.2 CESIUM-134

### 3.2.1 Nuclear Characteristics

#### 3.2.1.1 Atomic Number 55

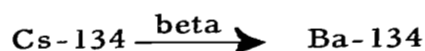
#### 3.2.1.2 Atomic Weight 134

#### 3.2.1.3 Half Life

2.050  $\pm$  0.004 years      Reference 1  
(2.1 years on latest GE chart  
of the nuclides)

(Cs-134m) 2.93  $\pm$  0.05 hours      Reference 2

#### 3.2.1.4 Energy Levels and Decay Scheme

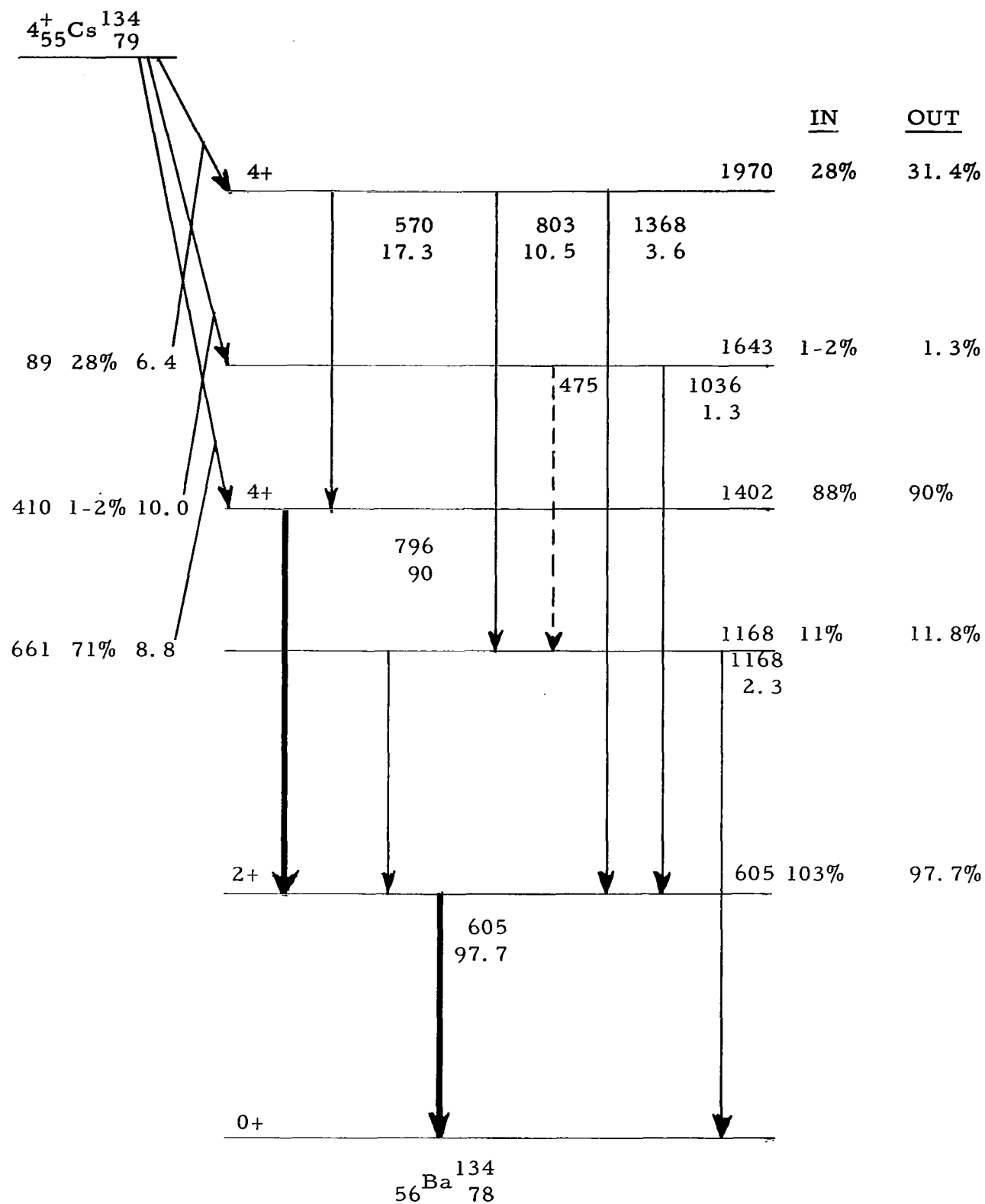


Beta      0.662 Mev (71%)  
            0.410 Mev (1%)  
            0.089 Mev (28%)

Gamma      1.368, 1.168, 1.036,      Reference 3  
            0.803, 0.796, 0.605,  
            0.570, 0.563, 0.475 Mev

$Q_{\text{beta}}$       2.059 Mev





### 3.2.1.5 Other Sources of Radiation

Since Cs-134 is produced by irradiation of Cs-133, the resultant product can be expected to be virtually free of any contaminants except the Cs-133 fraction which is inert. Therefore, no other source of radiation except that associated with the cesium-barium decay chain would be expected.

### 3.2.2 Chemical Characteristics

#### 3.2.2.1 Fuel Forms

Cs-134 fuel forms would be identical to those of Cs-137 shown in Section 3.3.0. Again, the polyglass would be the most probable form to be used as a heat source.

#### 3.2.2.2 Material Compatibility

For details on cesium fuel form compatibility refer to Section 3.3.0 on Cs-137.

#### 3.2.2.3 Effects of Impurities on Nuclear Characteristics

Because Cs-134 is produced by neutron irradiation in a reactor of stable Cs-133, the resultant Cs-134 fraction will be free of any contaminants other than the stable Cs-133 isotope.

### 3.2.3 Thermal and Physical Properties of Cs-134

The thermal and physical properties of Cs-134 glass are found in Table IV.

The specific power, specific activity and power densities listed in the table as the best available values are calculated on the basis of the following assumptions.

- (a) The density of the cesium glass is  $3.2 \text{ gm/cm}^3$ .
- (b) There is 0.16 gm Cs-134 per gm of cobalt glass. (This figure may be too high. Experiments are now in progress.) Reference 4.
- (c) The half life of Cs-134 is 2.05 years.

- (d) There is 1.7 Mev per disintegration consisting of 1.58 Mev due to gammas and 0.12 Mev due to betas.

TABLE IV

Thermal and Physical Properties of Cs-134 Glass

Thermal Conductivity	$\frac{\text{cal Cm}}{\text{sec } ^\circ\text{C Cm}^2}$	.0025 (a)	Ref. 5
Power Density	$\frac{\text{watts}}{\text{gm}}$	2.036 (b)	Ref. 6, Ref. 4 see above
Specific Power (Pure Cs-134)	$\frac{\text{watts}}{\text{kilocurie}}$	10.1 (b)	Ref. 6, Ref. 4 see above
Heat Capacity	$\frac{\text{cal}}{\text{gm } ^\circ\text{C}}$	0.44 (c)	Ref. 6
Heat of Fusion	$\frac{\text{cal}}{\text{mole}}$	--	
Weight Density	$\frac{\text{gm}}{\text{cm}^3}$	3.2 3.1	Ref. 7
Melting Point	$^\circ\text{C}$	$\sim 1275$ (d)	Ref. 5
Boiling Point	$^\circ\text{C}$	--	
Specific Activity (Theoretical)	$\frac{\text{kilocurie}}{\text{gm Cs-134}}$	0.190	Ref. 8

Key to Thermal and Physical Properties of Cs-134

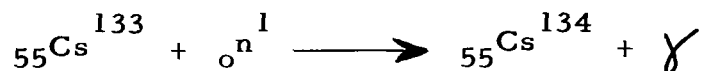
- (a) This value holds between 1000 $^\circ\text{C}$  and 2000 $^\circ\text{C}$  by analogy to sodium borosilicate glass. Thermal conductivity varies with heat treatment. The value at room temperature is 0.0015.

- (b) Calculated value, see comments preceding table of thermal and physical properties.
- (c) This is the value for Boron oxide glass between 1750°F and 3250°R.
- (d) Softening point.

#### 3.2.4 Isotope Production, Availability and Cost

##### 3.2.4.1 Production Method

Cs-134 is produced by the neutron irradiation of Cs-133, a natural occurring stable isotope of the alkali metals. The reaction is:



Production of Cs-134 is accomplished by reactor neutron bombardment of Cs-133 atoms. The rate of conversion is dependent upon the flux level of the reactor, length of time of irradiation and other factors. Table V presents data for reactor conversion of Cs-133 to Cs-134 for various times and fluxes (Ref. 7).

##### 3.2.4.2 Availability and Cost

Cs-134 has been little considered as a heat source because of its high gamma radiation and low power output. For this reason, essentially no data is available on possible production quantities and cost. Both the AEC and Oak Ridge have been contacted to obtain information in this regard, but to no avail. Since it is a material produced by the neutron irradiation of a target material (Cs-133), its availability will depend a good deal on the availability of reactor space. However, ball park numbers have been suggested by Hittman (reference 9). He estimates that based on a large scale irradiation program, the probable range for Cs-134 will be between \$1.00 and \$2.00 per curie of isotope.

TABLE V

Specific Activity of Cs-134 as a function of Irradiation Time and Neutron Fluxes.  
 (Specific Activities given under Irradiation Times are in curies per gram of Cs-133  
 target material, abundance of target material = 100%, activation cross section =  
 32.6 barns)

Neutron Flux (n/cm <sup>2</sup> sec)	Irradiation Time in Half Life Units									T <sub>max</sub> (days)	Activity at T <sub>max</sub> (curies/gram)
	0.01	0.03	0.05	0.10	0.30	0.50	1.0	3.0	5.0		
2 x 10 <sup>13</sup>	0.55	1.64	2.70	5.29	14.5	22.2	36	53.7	52.8	2806	54.1
2 x 10 <sup>14</sup>	5.45	15.9	25.7	47.9	108	139	152	72.2	30.1	658	153
2 x 10 <sup>15</sup>	49.3	118	159	190	102	43.2	4.8	0.0008	-	79	190

## REFERENCES

1. Half Lives of Cesium-137 and Cesium-134 as Measured by Mass Spectrometry. Analytical Chem. 7, 797-9, June 1963.
2. Decay of Cesium-134m UJV-1018/64, 10 p. 1964.
3. The Beta Decay of Cesium-134. Canadian Journal of Physics Vol. 42, 504-8, March 1964.
4. Private Communication with S. J. Rimshaw, ORNL, 6 December 1965.
5. Isotopic Power Data Sheets, ORNL, Cs-137 Glass, August 27, 1963.
6. Final Report, Radioisotope Evaluation Program, MNRP-MK-5002, December 23, 1964. Contract NAS 5-3879.
7. Handbook of Shielding Requirements and Radiation Characteristics of Isotopic Power Sources for Terrestrial, Marine, and Space Applications, ORNL 3576.
8. ORNL Radioisotopes Procedures Manual, F. N. Case. (Oak Ridge National Laboratory, Oak Ridge, Tennessee). ORNL 3633, June 1964.
9. The Feasibility of Producing Power from Radioisotopes. F. Hittman. Presented at Am. Nuclear Soc. Annual Meeting. Pittsburgh, Pennsylvania. June 10-14, 1957.

### 3.2.5 Annotated Bibliography

- 3.2.5.1 STUDY OF THE DECAY OF Cs-134 WITH A HIGH RESOLUTION Ge(Li) GAMMA-RAY SPECTROMETER. R. A. Brown and G. T. Ewan (Atomic Energy of Canada, Ltd., Chalk River, Ontario). Nucl. Phys., 68:325-36 June 1965

The gamma-ray spectrum of Cs-134 was studied with a 3.5 mm deep by 18 mm diameter Ge(Li) gamma-ray spectrometer. The conversion line spectrum was also studied in a high resolution beta-ray spectrometer and the K conversion coefficients of the transitions measured using a mixed source of Cs-134 and Cs-137. The energies in kev, relative intensities in per cent per disintegration, and dominant multipolarities of the gamma-transitions are: 475.26 (1.5%, E2); 563.11 (8.3%, E2); 569.24 (14.3%, M1); 604.64 (98.0%, E2); 795.80 (88.0%, E2); 801.80 (8.8%, E2); 1038.46 (1.0%, M1 + E2); 1167.65 (2.0%, E2); 1364.97 (3.4%, E2). The data are consistent with levels in Ba-134 at 0(0<sup>+</sup>), 604.6(2<sup>+</sup>), 1167.7(2<sup>+</sup>), 1400.4(4<sup>+</sup>), 1643.0(3<sup>+</sup>), and 1969.6 kev (4<sup>+</sup>). There is no evidence for previously reported levels at 1570 and 1773 kev.

- 3.2.5.2 HIGH-RESOLUTION STUDIES OF GAMMA-RAY SPECTRA USING LITHIUM-DRIFT GERMANIUM GAMMA-RAY SPECTROMETERS. G. T. Ewan and A. J. Tavendale (Atomic Energy of Canada Ltd., Chalk River, Ontario). Can. J. Phys., 42: 2286-2331 - November 1964.

The use of germanium lithium-drift p-i-n diodes as high-resolution gamma-ray spectrometers is described. With these spectrometers gamma-ray resolutions of 2.05 kev at 122 kev, 4.0 kev at 1333 kev, and 5.5 kev at 2614 kev were obtained. Using the detectors as pair spectrometers for high-energy gamma rays, a resolution of 9.8 kev on a 7.6 Mev gamma ray was obtained. The factors affecting the resolution of the detectors are discussed. Fano factors of 0.4 were observed. Efficiency curves are given for a 2.5 cm<sup>2</sup> x 3.5 mm detector and for a 5 cm<sup>2</sup> x 8 mm detector. The detectors were used to make high-resolution studies of the complex gamma spectra from sources of Cs-134, Pm-151, Gd-153, Eu-156, Gd-159, Yb-177, and Ra-226. Results are reported for the energies and intensities of the gamma rays observed in these studies.

((Also published as Canadian Atomic Energy Commission Report AECL-2079))

- 3.2.5.3 (UJV-1018/64) DECAY OF Cs-134m. J. Frana, I. Rezanka, and A. Spalek (Ceskoslovenska Akademie Ved. Ustav Jaderneho Vyzkumu, Rez). 1964, 10 p.

The decay of the isomeric state of Cs-134 was studied. The half life ( $2.93 \pm 0.05$  hours) was determined. From measurements carried out by means of a spectrometer with short lens, scintillation measurements, and chemical separations, the non-existence of the weak decay beta of this state was proved, contrary to statements found previously in literature (maximum possible intensity 0.02%, against the value of 1% found in literature). The spectrum of conversion electrons was measured by a double-focusing spectrometer, and the following transition energies were determined:  $127.3 \pm 0.3$  kev (E3) and  $138.4 \pm 0.4$  kev (M4) (K:L:M + N is 92:100:27 for the 127.3 kev transition, and 206:100:31 for the 138.4 kev transition). The K conversion coefficient of the 127 kev transition was measured, resulting in a value of  $2.55 \pm 0.4$ . The ratio of transition intensities is  $I_{138}:I_{127} = 5.7:1000$ .

- 3.2.5.4 THE PERFORMANCE OF Au-Si SURFACE BARRIER DIODE AS A BETA RAY DETECTOR. S. K. Sen (University of Manitoba, Winnipeg), Nucl. Instr. Methods, 27:74-6 (Apr. 1964).

Pulse height spectra for Cs-134 and Th (B + C + C'') were obtained. The conversion line energies were determined and compared with other determinations. The results indicate linear response of the detector from 150 to 1300 kev. The internal conversion coefficients of Cs-134 were also obtained and compared with theoretical values. (D. C. W.)

- 3.2.5.5 THE BETA DECAY OF CESIUM-134. W. Van Wijngaarden and R. D. Connor (Physics Department, University of Manitoba, Winnipeg, Manitoba). Canadian Journal of Physics, Vol. 42, 504-18, March 1964.

Magnetic spectrometer studies have revealed that the beta decay of Cs-134 proceeds via three partial spectra whose end point energies, intensities, and  $\log_{10} ft$  values are as follows:

<u>Energy</u>	<u>Intensity (%)</u>	<u><math>\log_{10} ft</math></u>
662	71	8.8
410	1	10.0
89	28	6.4



No evidence is found for beta transitions of intermediate energies. Upper limits have been placed on two previously reported beta transitions of higher energy as follows:

<u>Previously Reported Energy and Intensity</u>	<u>Present Work, Upper Limit Intensity</u>
1.453 Mev, 0.2%	0.005%
0.892 Mev, 1.5%	0.045%

These beta intensities to a revised level scheme produce a good balance with the gamma ray intensities depopulating the levels. The Q beta-value is 2.059 Mev.

Source Material	-	Cesium Chloride
Activity Concentration	-	30 curies per gram
Chemical Purity	-	vacuum sublimate onto aluminum foil at low temperature

3.2.5.6 (NP-13020 (p. 53-7)) BETA AND GAMMA RAY  
SPECTRUM OBSERVED IN THE DECAY OF Cs-134.  
P. N. Trehan (Punjab University, Chandigarh, India)

The decay of 2.19 year Cs-134 leads to the excited levels of Ba-134 by beta emission. This was extensively studied by various workers, but disparity is found in their results. An effort to clarify the situation is presented. Gamma rays from the decay of Cs-134 were measured using a NaI(Tl) crystal mounted on a photomultiplier tube and a 20-channel analyzer. A double-focusing magnetic spectrometer was used to analyze the beta spectrum of Cs-134 using a plastic phosphor as a detecting device.

3.2.5.7 INVESTIGATION OF THE DECAY OF Ba-133 and Cs-134  
BY GAMMA-GAMMA DIRECTIONAL CORRELATION  
MEASUREMENTS. F. Muennich, K. Fricke, and U.  
Wellner (Technische Hochschule, Braunschweig, Germany)  
Z. Physik, 174: 68-90 (1963) (In German)

The directional correlations of the gamma-gamma cascades following the decay of Ba-133 were measured: 355-81, 302-81, 79-81, and 276-160 kev. The results of these measurements together with results of other authors yielded the following well-defined values for the spin of

the levels of Cs-133:  $7/2+$  (ground state),  $5/2+$  (81 kev level),  $5/2+$  (160 kev level),  $3/2+$  (383 kev level), and  $1/2+$  (436 kev level). The results are discussed in the frame of the shell model. The decay of Cs-134 was investigated by measurement of the gamma-gamma directional correlations: 800-600 kev, 600-600 kev, 1.04-Mev-605 kev. A discussion of these results yielded for the mixing proportion ( ) of the gamma radiation from the transition of the second to first  $2+$  state in Ba-134 a value  $\delta = -0.50 \pm 0.05$  in contradiction to the theory of Davydov and Filippon. Spin 2 was obtained for the 1.64 Mev level of Ba-134; the parity of this state is probably negative.

3.2.5.8 LEVEL SCHEME OF BARIUM-134. S. O. Schriber, and B. G. Hogg (Allen Physics Laboratory, the University of Manitoba, Canada). Nuclear Physics 48, 647-651, 1963.

The gamma ray cascades following the decay of Cs-134 have been examined in using sum coincidence spectrometer. Compton summing peaks have been subtracted using a method developed by the authors. No evidence for the existence of levels at 1846, 1773, and 1570 kev has been found.

All source material was in liquid solution. A small drop of this solution was located at the tip of the 0.5 millimeter wall thickness, 4 millimeter outside diameter glass tube. The source to crystal distance was always 6 centimeters, and the angle between the detectors was maintained at  $135^\circ$  to avoid back scattering. Appropriate shielding was placed so that no part of one crystal could see any part of the other crystal.

3.2.5.9 DECAY SCHEME AND MATRIX ELEMENTS FOR THE TRANSITION Cs-134  $\longrightarrow$  Ba-134. H. Daniel, J. Huefner, and O. Mehling (Max-Planck-Inst. fur Kernphysik, Heidelberg). Ann Physik (7), 12:106-12 October 1963 (In German)

The decay Cs-134  $\longrightarrow$  Ba-134 was investigated with a double focusing spectrometer and a beta-gamma circular-polarization correlation setup. The asymmetry parameter (A) of the correlation between the 656 kev group and the following gamma cascade was measured to be  $A = -0.13 \pm 0.02$ , in good agreement with a value of Mann, Bloom, and Nagel. \*This (A) value implies a ratio (X) between Fermi and Gamow-Teller contributions to the decay of X =  $0.30 \pm \begin{smallmatrix} 0.04 \\ 0.03 \end{smallmatrix}$  and so a definitely non-vanishing Fermi matrix element.

The breakdown of the isotopic spin selection rule at an atomic number as large as that of Cs ( $Z = 55$ ) is not surprising. The transition is qualitatively discussed in shell model terms. Additional information is given for the disintegration scheme of Cs-134. The level scheme of Ba-134 is discussed in terms of the vibrational model.

\*Phys. Rev. 127, 2134-2137, 1962

- 3.2.5.10 HALF LIVES OF CESIUM-137 AND CESIUM-134 AS MEASURED BY MASS SPECTROMETRY. L. A. Dietz, C. F. Pachucki and G. A. Land (General Electric Company, Knolls Atomic Power Laboratories, Schenectady, New York). Analytical Chemistry 7, 797-799, June 1963

Mass spectrometric measurements of radioactive decay rates over a period of 1.5 years indicate a half life of  $30.35 \pm 0.38$  years for Cs-137 and  $2.046 \pm 0.004$  years for Cs-134\*. Precisions quoted as standard deviations are considered to be preliminary values because the experiment will be continued.

((Table 3, not shown here, presents published values for the half life of Cs-137 from 1951 to 1963. References total ten in number.)) For the cesium experiment, a batch of six chemically untreated tungsten V filaments was prepared by outgassing at about  $2000^{\circ}\text{C}$  for more than one hour in an auxiliary vacuum system. Each filament was then loaded with about  $1 \times 10^{-8}$  gram total of the radioactive cesium isotopes in the form of  $\text{Cs}_2\text{SO}_4$ . The same procedure was followed for loading filaments with the Cs-134 stock solution. ((Table 5, not shown here, presents the published values for the half life of Cs-134. These values total seven in number.))

\*((The values stated above were revised as a result of a telephone call to L. A. Dietz on 16 October 1964 to the following values.))

Cs-134 .....	$2.050 \pm 0.004$ years
Cs-137 .....	$30.0 \pm 0.38$ years

- 3.2.5.11 CIRCULAR POLARIZATION MEASUREMENTS IN THE BETA DECAY OF V-48, Co-56, Fe-59, and Cs-134. Lloyd G. Mann, Stewart D. Bloom and R. J. Nagel (Lawrence Radiation Laboratory, University of California, Livermore, California). Physical Review 127, 2134-2137, September 1962

The beta-gamma circular polarization angular correlation has been measured for V-48, Co-56, Fe-59 and Cs-134, using the Livermore apparatus described previously. Since all of these decays (with one exception) are characterized by  $J = 0$ ,  $J \neq 0$ , and  $T \approx 0$ , they possess the necessary elements for testing isospin conservation in allowed beta decay. The measured asymmetry parameters are  $-0.066 \pm 0.035$  for V-48,  $0.00 \pm 0.03$  for Co-56,  $-0.074 \pm 0.022$  for Cs-134 and  $-0.23 \pm 0.05$  and  $0.01 \pm 0.10$ , respectively, for the 462 keV and 271 keV beta branches in Fe-59. These results are consistent in all cases with a very small Fermi matrix element and therefore support the validity of isospin conservation with the doubtful exception of the 271 keV beta decay branch of Fe-59. However, in this case, the extreme difficulty of disentangling the 271 keV branch from the 462 keV branch which is pure Gamow-Teller  $\left[ \frac{3}{2} - (\text{beta}) \frac{5}{2} - (\text{gamma}) \frac{7}{2} \right]$  renders this exception rather uncertain. In the case of V-48, our result is in disagreement with that of Boehm and Wapstra who found evidence for sizable interference, but is in better agreement with the result of Daniel and Kuntze who find evidence for small interference. Our Co-56 result is in excellent agreement with the work of Daniel and Kuntze. Our result for the low energy branch of the Fe-59 decay ( $\Delta J = 0$ ) is in accord with the work of Foster and Sanders, the accuracy being, however, very poor in both measurements. With regard to the high energy branch ( $\Delta J = 1$ ) our result is lower by a factor of 2 than that of Forster and Sanders. Our Cs-134 result shows definite interference but because of the large ft value, the Fermi matrix element is still very small in keeping with the other findings at this laboratory.

((Article mentions only that cesium sources were purchased from commercial suppliers. All sources were mounted on uncoated mylar films of 1 mg/cm<sup>2</sup> by solution evaporation.))

3.2.5.12 LOW-LYING ENERGY LEVELS OF CESIUM-134. I. V. Estulin, A. S. Melioransky and L. F. Kalinkin (Research Institute of Nuclear Physics, Moscow State University, Moscow). Nuclear Physics 16, 168-74, April 1960

A study is made of the cascade gamma transitions following thermal-neutron capture in cesium nuclei. Energy and quantum characteristics of low lying excited states of Cs-134 with excitation energy up to 320 keV are determined.

((A luminescent coincidence spectrometer worked out by one of the authors of the present paper was used for measuring cascade gamma quanta. The spectrometer consists of the governing and the main channels with a NaI(Tl) crystals and a photomultiplier as gamma ray detectors.))

3.2.5.13 DECAY OF CESIUM-134. John D. French, and Max Goodrich (Louisiana State University). American Physical Society Bulletin, 4, 391, 1959

The gamma rays in the decay of Cs-134 (2.2 years half life) have been studied with a scintillation spectrometer employing a cylindrical (3 inch by 3 inch) crystal of NaI (Tl) and with a coincidence spectrometer using two such crystals in the spectrometers. The pulse spectrum in coincidence with the peak corresponding to each of the gamma rays was obtained. The data indicate that at least 15 gamma rays were present, all of which occur in the nucleus of Ba-134 following negatron decay, and none occur in the nucleus Xe-134 following positron decay. Previously unreported cross-over transitions of 1640 and 1970 kev, confirming these energy levels were observed. A decay scheme with energy levels in Ba-134 at 605, 1038, 1168, 1367, 1401, 1640, and 1970 kev were presented.

3.2.5.14 THE LEVEL SCHEME OF BARIUM-134. SOME FEATURES OF THE SODIUM IODIDE SUMMING SPECTROMETER. R. K. Girgis and R. Van Lieshout, Nuclear Physics 12, 672-688. September 1959

A detailed analysis of the gamma ray spectrum from Cs-134 was performed. The results are in agreement with previous measurements but two additional weak gamma rays of 960 and 1570 kev were found. Summing peaks of 1.17, 1.40, 1.57, 1.64, 1.77, and 1.97 Mev were observed, establishing the presence of levels of these energies in Ba-134. The gamma ray spectrum from the decay of La-134 was also examined. Only one gamma ray of 605 kev was observed, thus confirming that the first 2+ level in Ba-134 has this energy. Some features of the NaI summing spectrometer are demonstrated and discussed in an appendix to this report.

((Figure 6 of the article presents the proposed level of Ba-134.)) The energy values written on the left-hand side of the various transitions represent the gamma rays following the decay of Cs-134. The numbers in brackets represent their relative intensities in percentage

per disintegrations assuming all disintegrations to go through either the 605 or the 1170 kev transitions. The uncertain 605 kev transition de-exciting the level at 1170 kev is drawn in as a dashed line. The characteristics of the beta branches following the decay of Cs-134 in those following the decay of La-134 are shown on the graph in the following order: Energy of the Branch, Intensity and Percentages per Disintegration, and the Log ft value. These values are calculated from the various gamma ray populating and de-exciting levels.

- 3.2.5.15 GAMMA RADIATION FROM CESIUM-134. B. S. Dzhelepov, V. P. Prikhodtseva and Yu. V. Hhol'nov. Izvestia Nauk, SSSR, Vol. 23, 826-7, July 1959

((Article references the energies and relative intensities of the gamma rays from Cs-134 measured during the experiments described in the article.)) Energy of the gamma rays in kev 475, 567, (565 of 571), 606, 663, 797, 1038, 1168, 1368. Relative intensity of the gamma  $2.5 \pm 0.5$ ,  $28.0 \pm 3$ ,  $10 \pm 3$  or  $18.0 \pm 3$ , 100,  $0.5 \pm 0.3$ ,  $101 \pm 4.3$ ,  $1.16 \pm 0.04$ ,  $2.26 \pm 0.08$ ,  $3.79 \pm 0.15$ .

- 3.2.5.16 INVESTIGATION OF CESIUM-134 BY THE COINCIDENCE METHOD. Iu. A. Leleksandrov. Izvestia Akad Nauk, SSSR, 827-30, July 1958.

This investigation was undertaken for the purpose of investigation of the gamma-gamma and beta-gamma coincidences for the purpose of refining the decay scheme. Our measurements substantiate the existence of the 605-797, 605-1370, 605-570, 797-570 and 1170-802 (797) kev cascade transitions. Our results also indicate the existence of the following cascades: 605-1040 and 1170-475 kev. We also investigated the high-energy region of the Cs-134 gamma spectrum. We observed photo peaks from gamma rays of 1640, 1750, 1870, 1960, and 2040 kev energy. The intensity of all these gamma rays is of the order of  $10^{-6}$  photons per disintegration. All our experimental data on beta-gamma coincidences and the hard part of the Cs-134 gamma spectrum are consistent with the decay scheme proposed by Foster and Wiggins\*. (The relative intensities of the cascade transitions according to our evaluations are as follows: 605-797 kev, 100%. 605-570 kev, 10%. 605-1370 kev, 3%. 605-1040 kev, 3%. 797-570 kev, 10%. 1170-805 kev (797), 3%. 1170-475 kev, 2%.

\*Nuovo Cimento 2, 854, 1955

3.2.5.17 THE CROSS SECTION OF THE REACTION CESIUM-134 (N, GAMMA) CESIUM-135, AND THE HALF LIFE OF CESIUM-134. J. G. Bayly and F. Brown (Atomic Energy of Canada, Ltd., Chalk River, Ontario), and G. R. Hall and A. J. Walter (Atomic Energy Research Establishment, Harwell Didcot, Berks). Journal of Inorganic and Nuclear Chemistry 5, 259-263, 1958

The isotopic composition of cesium samples which have been subject to intense reactor neutron radiation was measured with a 6-inch radius  $60^\circ$  sector field, mass spectrometer fitted with a triple filament thermal ionization source. The cross section of the reaction Cs-134 (n, gamma) Cs-135 was calculated from the observed isotopic composition and found to be  $134 \pm 12$  barnes. By measuring the change with time of the Cs-134/(Cs-134 + Cs-135) ratio using the mass spectrometer, the half life of Cs-134 was found to be  $2.15 \pm 0.08 - 0.04$  years. In a subsidiary experiment the cross section of the reaction Cs-133 (n, gamma) Cs-134 was found by activation to be  $30.4 \pm 1.7$  barnes.

Two long irradiations were carried out. The first was approximately 10 milligrams of spectroscopically pure  $\text{Cs}_2\text{CO}_3$  in the M. T. R., Idaho for 140 days. The second was of approximately 10 milligrams of  $\text{CsNO}_3$  in the NRX reactor, Chalk River for 356 days. Two short irradiations were made, each of 16 hours, using a position of NRX similar to that used for the long irradiations. For these irradiations the cesium compounds which were of spectroscopic purity were dried at  $110^\circ\text{C}$  weighed into silica tubes which were then sealed.  $\text{CsC}_2$ , 36.8 milligrams was used in the first irradiation, and  $\text{Cs}_2\text{CO}_3$ , 32.1 milligrams in the second.

Results: The half life of Cs-134. The decay of Cs-134 is shown by the data of Table I ((Table I is not shown)). The error shown for the Cs-134 percentages are based on the standard deviation between spectrograms. The half life of Cs-134 obtained from these data is  $2.15 \pm 0.08 - 0.04$  years. The value obtained by Merritt, et al\* using a 4 pi proportional beta counter over a period of 5 years is  $2.19 \pm 0.02$  years. This latter value was used in our calculation because owing to the longer period over which decay was studied it should be more accurate. Our value of 2.15 years is a useful confirmation of Merritt's result since the mass spectrometer value is not affected by the problem of radiochemical purity.

\*Canadian Journal of Physics 35, 16, 1957

- 3.2.5.18 HALF-LIFE DETERMINATION OF SOME RADIO  
NUCLIDES. W. F. Merritt, P. J. Campion and R. C.  
Hawkings, Canadian Journal of Physics, 35, 16-20, 1957.

The half lives of the following nuclides have been determined by absolute counting techniques using a pi beta proportional counter. The period over which the observations were made is indicated by brackets. The errors quoted are the statistical counting errors. Na-22, 2.58  $\pm$  0.03 years (2 years); Ru-106, 1.02  $\pm$  0.01 years (5 years); Cs-134, 2.19  $\pm$  0.02 years (5 years); Ce-144, 285  $\pm$  2 days (4 years); Pm-147, 2.64  $\pm$  0.02 years (4 years); Tl-204, 3.56  $\pm$  0.05 years (4 years). ((A review of previous determinations is included in the article.))

- 3.2.5.19 RAPID ANALYSIS OF GAMMA EMITTERS USING A  
GAMMA RAY SCINTILLATION SPECTROMETER I.  
QUANTITATIVE ANALYSIS OF CESIUM-134 - CESIUM-  
137 MIXTURE. Fumio Aoki, Toshio Kurosawa and  
Seishi Yajima. (Chemical Society of Japan (Bulletin))  
30, 583-5 Sept. 1957

Rapid determination of individual species in a mixture radio nuclides will be required in many cases such as processing of fission products, activation analysis, and chemical, biological, or metallurgical experiments with several kinds of radio nuclides. The authors have been investigating the possibility of quantitative determination of two nuclides which cannot be resolved by the spectrometer. This report is a result of the study on Cs-134 - Cs-137 mixture.

((The chemical form of cesium was cesium chloride in aqueous solution. It was diluted to about 0.1 microcuries/milliliters with demineralized water.))

- 3.2.5.20 THE FEASIBILITY OF PRODUCING POWER FROM  
RADIOISOTOPES. F. Hittman. Presented at American  
Nuclear Society Annual Meeting. Pittsburgh, Pennsylvania.  
June 10-14, 1957
- 3.2.5.21 HALF LIVES OF Sc-46, Co-60, Zn-65, Ag-110m, Cs-134,  
and Eu-152, 194. K. W. Geiger (Division of Applied  
Physics, National Research Council, Ottawa, Canada).  
Physical Review, 105, 1539-40, March 1957.



The half lives of six gamma emitting nuclides were determined by comparison with radium standards using a lead shielded ionization chamber. The following results were obtained: Sc-46,  $83.89 \pm 0.12$  days; Co-60,  $5.24 \pm 0.03$  years; Zn-65,  $243.5 \pm 0.8$  days; Ag-110m,  $252.5 \pm 0.5$  days; Cs-134,  $2.07 \pm 0.02$  years; Eu-152,  $154, 12.2 \pm 0.2$  years. The errors quoted are twice the standard deviation calculated from a least squares analysis.

The investigated nuclides were produced by neutron radiation of the Chalk River NRX reactor. The initial activities of the samples were between 100 and 300 millicuries. Details on composition and purity are given in Table I ((presented in part below)). The gamma radiation was measured with a one-atmosphere air-filled parallel plate ionization chamber made of aluminum and shielded by a 0.6 centimeter of lead. Saturation properties were studied by the two-source method but no effects within the experimental errors of  $0.0 \pm 0.15\%$  could be found. ((Contents of Table I. Half lives of several gamma emitting nuclides)) Nuclide, Cesium-134; sample, spec-pure.  $\text{Cs}_2\text{AlF}_3$ ; decay followed over half lives, 0.8; half life,  $2.07 \pm 0.02$  years; previous measurements  $2.3 \pm 0.3$  years;  $1.7 \pm 0.1$  year. (End of Table))

((Note on Table I. Cs-134. Although the decay of Cs-134 has not been followed for a full half life, the present value is much more accurate than the figures published previously.)) ((Cited as best value in Rev. Mod. Phys. Vol. 30, #2, Pt. II, April 1958.))

### 3.2.5.22 STUDIES WITH SCINTILLATION COINCIDENCE SPECTROMETER: Cs-134. Girish Chaudra (Tata Institute of Fundamental Research, Bombay, India). Proceedings of the Indian Academy of Science A44, 194-200, 1956.

The set-up of slow fast coincidence scintillation spectrometer is described. Gamma-gamma coincidences in Cs-134 have been studied. The 604-keV gamma transition is found to be in cascade with  $460 \pm 20$ ,  $555 \pm 15$ ,  $794 \pm 15$  and  $1349 \pm 30$  keV gamma transitions. The 794-keV gamma transition is found to be in cascade with  $604 \pm 10$  and  $555 \pm 15$  keV gamma transitions. The results are consistent with the decay scheme of Cs-134 proposed by Forster\*.

\*Nuovo Cinemto, 2, 854, 1955.

3.2.5.23 HIGH ENERGY FORBIDDEN BETA RAY TRANSITIONS  
FROM Cs-134 (2.3 years), Co-60 (5.3 years), Sc-46 (84  
days), and Hg-203 (47.9 days). J. L. Wolfson,  
Canadian Journal of Physics, 34, 256-64, March 1956

Beta ray transitions of energies  $1478 \pm 6$  kev from Co-60 (5.3 years) and  $1475 \pm 6$  kev from Sc-46 (84 days) were observed with intensities of  $(1.0 \pm 0.2) 10^{-4}$  and  $(3.6 \pm 0.7) 10^{-5}$  beta rays per disintegration, respectively. Forbidden spectrum shapes were obtained for both transitions. No evidence was obtained for the beta ray transition of energy 1451 kev from Cs-134 (2.3 years) nor for the transition of energy 473 kev from Hg-203 (47.9 days) and upper limits of  $5 \times 10^{-5}$  and  $3 \times 10^{-4}$  beta rays per disintegration, respectively, are placed upon the intensities of these transitions. The log ft values are greater than 14.5,  $14.0 \pm 0.1$ ,  $13.0 \pm 0.1$  and greater than 11.3 for the transitions referred to from Cs-134, Co-60, Sc-46, and Hg-203, respectively.

The sources were all prepared by neutron irradiation in the NRX reactor. In general, the sources were prepared by evaporation of salts from nitric acid solution on backings of 2.3 milligrams/cm<sup>2</sup> aluminum foil. The sources were all approximately 2 millimeters in diameter. The Cs-134 source was about 9 milligrams/cm<sup>2</sup> thick and about 0.75 millicuries in activity.

3.2.5.24 INTERNAL CONVERSION COEFFICIENTS OF Ba-134,  
V-51, and Tl-203. Z. O'Friel and A. H. Weber  
(St. Louis University, St. Louis, Missouri). Physical  
Review 101, 1076-8. February 1956

The internal conversion coefficients of Ba-134 (Parent - Cs-134) V-51 (Parent - Cr-51) and Tl-203 (Parent - Hg-203) have been measured by the determination of the gamma ray yields from the Compton electron spectra produced in an external converter employing the analysis of R. G. Thomas and T. Lauritsen and a single magnetic lens beta ray spectrometer. The Ba-134 results involve the extension of the method to several gamma ray conversions measured simultaneously. V-51 represents the application of the method to K capture type of decay for which the direct area under curves method cannot be used. Tl-203 required the correction of the measured conversion spectrum for photoelectric conversions to obtain the pure Compton events. The values obtained for the total internal conversion coefficients are Ba-134: 569 kev gamma ray,  $(9.2 \pm 1.2) 10^{-3}$ ; 605 kev gamma ray, (5.3

$\pm 0.5) 10^{-3}$ ; 796 kev gamma ray,  $(2.4 \pm 0.3) 10^{-3}$ ; 1.367 kev gamma ray,  $(0.49 \pm 0.05) 10^{-3}$ ; V-51: 3.25 kev gamma ray,  $(1.5 \pm 0.2) 10^{-3}$ ; Tl-203: 279 kev gamma ray for K shell  $(1.5 \pm 0.1) 10^{-1}$  and  $(4.9 \pm 0.2) 10^{-1}$  for the L shell. In all cases, the precision is the standard deviation. The results are in satisfactory agreement with other experimental values and with predictions of Rose's Tables. ((All samples obtained from Oak Ridge.))

3.2.5.25 GAMMA-GAMMA DIRECTIONAL CORRELATIONS IN CESIUM-134. A. E. Everett and J. J. Glaubman (Princeton University). Physical Review 100, 955, 1955

We have found the directional correlations of the 1.37 - 0.605, 1.17 - 0.801, and 0.796 - 0.605 Mev cascades in the decay of Cs-134 to be given by:  $A_2 = 0.107, 0.095, \text{ and } 0.111, \pm 0.015$ ;  $A_4 = 0.015, 0.006, \text{ and } 0.016, \pm 0.02$ , respectively. All of these values are consistent with 4-2-0 cascades and agree with the decay scheme suggested by Cork, et al\*. In this case, the five observed gamma rays are E2, the excited state of Ba-134 are at 0.602 (2+), 1.17 (2+), 1.40 (4+), and 1.97 (4+), Mev, and the 0.563 Mev E-2 transition is between the second and first excited states, and is speeded up by a factor of 15 comparative cross over. All this is very typical to even-even nuclei in this region. However, the decay scheme of the Washington group can not be completely excluded since a 4-2-0 directional correlation can also be interpreted as due to a 3-2-0, 3-1-0, or 2-2-0 cascade, in which the first transition is mixed.

\*Phys. Rev. 90, 444-7, 1953

3.2.5.26 DECAY OF CESIUM-134. H. H. Forster, and J. S. Wiggins (Department of Physics, University of Southern California, Los Angeles, California). Nuovo Cimento, Vol. 2, No. 4, October 1955

The beta spectrum and internal conversion electrons of Cs-134 were obtained with a double thin lens beta ray spectrometer. The detector was a conventional type geiger-mueller counter with a replaceable end window. Thin nylon films, 10-50 micrograms/cm<sup>2</sup> thick were used as counter windows. The radioactive source material was cesium chloride in hydrochloric acid solution obtained from Oak Ridge. Thin uniform sources, 10-25 micrograms/cm<sup>2</sup> were deposited on nylon or tygon films by the usual evaporation methods and the films were supported by a lucite cylinder the sides of which had been cut so as to

leave only a narrow range held by four thin strips. The resolution of the instrument was varied between 1.5 and 2.3%, except in the investigation of the end point of the highest energy beta component when a resolution of 3.5% and a source up to 50 micrograms/cm<sup>2</sup> were used.

- 3.2.5.27 ANGULAR CORRELATION OF THE GAMMA RAYS OF BARIUM-134. M. G. Stewart, R. P. Scharenberg, and M. L. Wiedenbeck (Department of Physics, University of Michigan, Ann Arbor, Michigan). Physical Review, 99, 691-4, August 1955

The angular correlations have been measured between the following pairs of gamma rays from Ba-134: 605 kev - 797 kev, 1369 kev - 605 kev, and 570 kev - 605 kev. These measurements indicate that the spins associated with these gamma rays are 4, 4, 2, 0. The 605, 797 and 1368 kev gamma rays are pure quadrupole while the 570 kev gamma ray is a mixture of 94% quadrupole and 6% dipole.

- 3.2.5.28 RADIOACTIVITY OF CESIUM-134. G. Bertolini, M. Bettoni, and E. Laszarina. (I. Stieuto Di Fisica, Sperimentale del Politecin - Malino). Nuovo Cimento, Vol. 11, No. 2, 273, August 1955

The beta decay of Cs-134 and the gamma decays of Ba-134 have been investigated with an intermediate image beta spectrometer and a scintillation spectrometer. From beta-gamma coincidences, a new beta decay of 335 kev end point has been revealed and the existence of a weak beta decay on the 1350 kev level of barium has been confirmed. From beta-gamma and gamma-gamma coincidences and from internal coefficients, the relative intensity of the beta decays and the multipolarity of the gamma transitions have been investigated. The five excited states of Ba-134 are assumed having energy of 795, 1350, 1395, 1700, and 1949 kev. Their spins and parity are 2+, 2+, 3+, or 4+, 3+, or 4+, and 4+.

Cs-134, 5.6 millicuries/grams specific activity, was supplied from Harwell. It was mounted on nylon film of 0.16 mg/cm<sup>2</sup> and, in order to study carefully the beta spectrum shape, it was evaporated under vacuum on the aluminum foil 1 micromillimeter thick. To investigate the beta spectrum a source of about 1 microcurie was used. A source of about 10 microcuries was used for gamma-gamma and beta-gamma coincidences, and for internal and photoelectric conversion. ((Table 1 of this article, not reproduced, presents a summary of the work of

seven authors on the following information: beta decay energy in kev, gamma decay energy in kev and conversion coefficient.))

- 3.2.5.29 ON THE DECAY OF CESIUM-134. G. Bertolini, M. Bettoni and E. Lazzarini. Nuovo Cimento 10, 1, 746-8, April 1955

Cs-134 is known to decay by beta emission on excited states of Ba-134. Several measurements have been made on the energy of the gamma rays and on the beta ray spectrum with scintillation and magnetic spectrometers. A continuous spectrum shows a low energy component (about 90 kev) and two high energy components (about 650 kev), the in points of which differ by  $30 \div 40$  kev. We have investigated the beta decays in coincidence with gamma rays of different energies with an intermediate image beta ray spectrometer with NaI(Tl) crystal. A complete description of the beta-gamma and gamma-gamma coincidence is going to be published soon\*.

\*Nuovo Cimento, Vol. 11, No. 2, August 1955.

- 3.2.5.30 RADIOACTIVE DECAY OF CESIUM-134 AND CESIUM-134m. G. L. Keister, E. B. Lee and F. H. Schmidt (Department of Physics, University of Washington, Seattle, Washington). Physical Review 97, 451, February 1955

The radiations of Cs-134 (2.3 years) and Cs-134m (3.15 hours) have been studied in a high resolution beta ray spectrometer. The gamma rays of Cs-134 were observed by internal and external conversion, and coincidence rates were measured between the beta continuum and the internal conversion electrons of the stronger gamma rays. The beta spectrum of Cs-134 appears to consist of 4 or possibly 5 components. Nine gamma rays were found both by internal and external conversion; a tenth gamma ray appears only in internal conversion. A decay scheme is proposed which is reasonably consistent with the multipole order of each of the radiations as obtained from internal conversion data. A search was made for a ground or intermediate state beta transition from Cs-134m. No transition to the ground state was observed, (but some indication was found for a weak transition to an excited state.))

- 3.2.5.31 GAMMA RAY ENERGIES IN THE DECAY OF CESIUM-134. M. C. Yoshi, and B. V. Thosar (Tata Institute of Fundamental Research, Bombay, India). Physical Review 96, 1022-3. November 1954

The photoelectric conversion spectrum of gamma rays in the decay of Cs-134 was studied with a newly installed Siegbahn-Slatis beta ray spectrometer using a strong source and thick lead and uranium radiators to bring out weak lines. Lines corresponding to the following gamma rays were found. Expressed in kev; 467, 553, 571, 607, 794, 1027, 1164, 1368, 1401.

Figure 1 ((not shown)) shows the spectrum of the photoelectric conversion electrons of the gamma rays in lead of thickness 110 mg/cm<sup>2</sup>. Photoelectric lines were observed corresponding to the following gamma ray energies expressed in kev;  $467 \pm 15$ ,  $553 \pm 7$ ,  $571 \pm 7$ ,  $607 \pm 5$ ,  $794 \pm 3$ ,  $1027 \pm 15$ ,  $1164 \pm 10$ ,  $1368 \pm 5$ , and  $1401 \pm 15$ . This confirms the gamma ray energies reported by Schmidt and Keister\* and by Waggoner\*\*, et al, who studied the electron spectrum caused by internal conversion, and gives in addition 2 gamma rays, 1.47 Mev and the other at 1.40 Mev. A fairly consistent level scheme for barium can be drawn to explain most of the gamma ray transitions, though the gamma ray of energy 1.036 Mev has been attributed by some observers to an alternate K capture decay branch. The Radio Chemical Center, Amersham, England, which supplied the active cesium bromide gave us the quantitative spectrographic analysis of the material.

\*Phys. Rev. 86, 632, 1952

\*\*Phys. Rev. 80, 420-8, 1950

3.2.5.32 THE NUCLEAR SPIN AND MAGNETIC MOMENTS OF Na-24, K-42, RB86, Cs-131, and Cs-134. R. H. Bellamy and K. F. Smith, Cavendish Laboratories, Cambridge, Philosophical Magazine 44, 33, 1953

3.2.5.33 DECAY OF CESIUM-134 AND THE LEVEL SCHEME OF BARIUM-134. P. N. Trehan, John D. French and Max Goodrich (Louisiana State University, Baton Rouge, Louisiana). Physical Review 131, 2625-31, September 1953.

The energy (in kev) and the relative intensity of the gamma rays following the decay of Cs-134 have been measured as 475 (1.1), 563 (9), 569 (11), 605 (98), 800 (93), 1038 (1.1), 1168 (2.7), and 1367 (3.5). With the help of the various scintillation spectrometry techniques, no gamma rays of energy 960, 1570, 1640, 1770 and 1970 kev reported earlier by certain workers could be detected. In addition to the

already established 86 kev (26%) and 655 kev (71%) and point beta rays, three beta rays of end point and intensities 1453 kev (0.13%) 892 kev (0.7%) 410 kev (2.1%) were confirmed by careful measurements with the help of the beta ray spectrometer. The K-conversion coefficient of the gamma rays and the log ft value of the beta rays were obtained. The measurements established more firmly the decay scheme including the spin and parity assignments to the various levels.

Source Preparation: The Cs-134 used in our experiments was obtained as cesium chloride in hydrochloric acid from Oak Ridge National Laboratory where it had been made by (n, gamma) reaction in Cs-133. The sources used for the beta ray and the internal conversion electron studies were mounted on thin films of collodion (about 5 - 10 micrograms/cm<sup>2</sup> thickness). A drop of acid-free solution of active material was evaporated on a collodion film already treated with insulin. The sources used for the beta ray work were prepared by evaporation of a drop of active material either on aluminum foil backing or on lucite backing.

- 3.2.5.34 THE RADIOACTIVE DECAY OF CESIUM-134. OSMIUM-185, 191, 193. J. M. Cork, J. M. LeBlanc, W. H. Nester, D. W. Martin and M. K. Brice (Department of Physics, University of Michigan, Ann Arbor, Michigan.) Physical Review 90, 444-7, May 1953

From sources of cesium of high specific activity produced in the pile and studied in photographic magnetic spectrometers 19 electron conversion lines were observed. These are interpreted to show the existence of 11 gamma rays, 4 of which have not been previously reported. Certain K/L ratios are measured and the resolution of the components of the beta radiation presented. The plausible decay scheme consisting of 7 levels in the resultant Ba-134 nucleus is offered ((balance of abstract referring to osmium has been omitted)).

- 3.2.5.35 THE BETA-SPECTRUM OF 2.3 YEAR CESIUM-134. F. H. Schmidt, and G. L. Keister (University of Washington). Physical Review 86, 632, 1952

The electron radiations resulting from the decay of Cs-134 have been studied in a high resolution solenoidal spectrometer. Conversion electrons have been observed from seven gamma rays.  $561.5 \pm 1.0$ ,  $566.6 \pm 0.8$ ,  $601.2 \pm 0.5$ ,  $793.1 \pm 0.7$ ,  $1037.2 \pm 2.6$ ,  $1164.4 \pm 2.9$  and  $1365.7 \pm 3.3$  kev. The two lowest energy gammas whose ratio of K

electron intensities is about 0.5, corresponds to the single gamma ray found by the other workers. The 3 high energy lines are very weak: the measured K-1 differences do not exclude conversion to xenon but indicate that conversion is more probably occurring in barium. There is no evidence of a contaminant. The continuous spectrum shows some indication of two high energy components whose end points differ by 30-40 kev. The end point of the low energy branch (about 24%) is about 79 kev. On the basis of these energies and other data a fairly consistent scheme for Ba-134 can be constructed with levels at 793, 1359, 1394, and 1955 kev. The K internal conversion coefficients for the 793 and 601 gammas can be found with reasonable certainty. ( $2.1 \pm 0.2$  and  $5.7 \pm 0.5 \times 10^{-3}$ , respectively.) Coefficients for the 561 and 566 depend strongly upon which is associated with the low energy beta branch. The role of the 1037 gamma is very uncertain.

- 3.2.5.36 THE NUCLEAR SPIN AND MAGNETIC MOMENT OF  $^{55}\text{Cs-134}$ . V. Jaccarino, B. Bederson and H. H. Stroke (Research Laboratory of Electronics, and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts). Physical Review 87, 676-7, August 1952

The atomic beam magnetic resonance method has been applied to the measurement of the hfs interaction in the ground state of the radioactive nuclide Cs-134. A 200-millicurie sample of  $(\text{Cs-134})_2\text{CO}_3$  was prepared by neutron irradiation in the Brookhaven pile, yielding a ratio of Cs-134 to Cs-133 of 1 to 6000 in the sample. A directional oven of special design was used to produce the atomic beam of cesium, which after being ionized at the hot wire detector was analyzed with a mass spectrometer having an enrichment factor of 2000 per mass number at the mass 134 position.

- 3.2.5.37 ANGULAR CORRELATION OF THE GAMMA-RAYS OF CESIUM-134. Bertol L. Robinson and Leon Madansky (John Hopkins University, Baltimore, Maryland). Physical Review 84, 604, November 1951

A program has been initiated for the study of the correlations of cascade radiations in the decay of radioisotopes.

- 3.2.5.38 GAMMA RAYS OF Cs-134. V. S. Shpinel. Zhur. Eksptl'. i Teoret. Fiz. 21, No. 7, 853-5 (1951) July. (Letter to the Editor; in Russian)



The gamma-ray spectrum of Cs-134 has been measured with a gamma-ray spectrometer previously described by the author ((Shpinel, Zhur. Tekh. Fiz. 20, 834 (1950); NAS 5-3725)). The following gamma-ray energies (in kev) were found, the figures in parentheses being relative intensities:  $K_1$ ,  $569 \pm 5$  ( $0.35 \pm 0.09$ );  $L_1$ ,  $571 \pm 5$ ;  $K_2$ ,  $601 \pm 4$  ( $0.94 \pm 0.24$ );  $L_2$ ,  $601 \pm 7$ ;  $K_3$ ,  $794 \pm 7$  ( $1.0$ );  $L_3$ ,  $793 \pm 7$ ;  $K_4$ ,  $1024 \pm 10$ ,  $K_5$ ,  $1110 \pm 10$ ; and  $K_6$ ,  $1347 \pm 14$  ( $0.017 \pm 0.008$ ).

3.2.5.39 INTERNAL CONVERSION OF GAMMA RAYS FROM COBALT-60, CESIUM-134, AND ZINC-156. M. A. Waggoner, M. L. Moon, and A. Roberts (Department of Physics, State University of Iowa, Iowa City, Iowa). Physical Review 80, 420-8, November 1950

Using a double coil thin lens magnetic beta ray spectrometer of transmission 2.40% and line width of approximately 3% the internal conversion coefficients of the gamma rays of Co-60, Cs-134, and Zn-65 have been measured. Sufficient precision has been obtained in several cases to verify the theoretical values and to obtain unambiguous identification of the multipole character of the gamma rays. Internal conversion coefficients as small as  $10^{-5}$  can be measured with a precision of 5% ((5% error)) or better. The results obtained indicate that both of the gamma rays from the Co-60 are electric quadrupole (EQ), the 560 kev gamma rays from Cs-134 is either EQ or MD or a mixture of these. The 602 and 799 kev gamma rays from Cs-134 are both EQ, the 1.114 Mev gamma ray from Zn-65 is either EQ or MD or a mixture of these, and the 1.363 Mev gamma ray from Cs-134 may be EQ. Further data on the decay scheme of Cs-134 are given. The results show that in combination with angular correlation measurement, internal conversion data determined the angular momenta and parities of the excited nuclear states.

3.2.5.40 CROSS-OVER TRANSITIONS Ir-194, Ag-110, and Cs-134 Richard Wilson (Clarendon Laboratory, Oxford, England) Physical Review 79, 1004, September 1950.

According to the accepted decay schemes for these isotopes, cross-over transitions might be expected. A search has been made for these transitions using the photodisintegration of beryllium and deuterium as threshold detectors for a high energy gamma energy radiation. The photoneutrons were detected by Szilard-Chalmers reaction in ethyl iodide under conditions in which the detection efficiency has been measured as a function of energy. Approximate

estimates of the intensities have been made using pile activation data to get the disintegration rate of the isotope. The accuracy of these is about 50%. The results are shown in Table 1. ((Table 1 is entitled Cross-Over Transition Data and is shown in part as follows.)) Isotope; Expected energy (Mev) 1.96; expected intensity in photons per disintegration,  $10^{-5}$ ; observed intensity photons per disintegrations, less than  $10^{-4}$ . For Cs-134 the upper limit to the intensity is in good agreement with the results of Fluharty which had escaped notice until the experiment had been completed.

3.2.5.41 GAMMA-GAMMA-CORRELATION EXPERIMENTS. J. R. Beyster and M. L. Wiedenbeck. Physical Review 79, 411, July 1950.

The correlation of successive gamma rays has been reported previously for a number of radioactive substances. We have also investigated the gamma-gamma correlation for several of those activities and for a few other isotopes. Figure 1, Curve B ((not shown)) is observed function for Cs-134. It will be recalled that Cs-134 has essentially three gamma rays in cascade, the upper gamma appearing about 25% of the time. It is possible to explain the experimental data with the assumptions that the two lower transitions are quadrupole between states possessing angular momenta ( $J = 4, 2, 0$ ) and that the upper transition is quadrupole with  $J = 4, 5$ , or 6 for the uppermost state. The polarization correlation experiments and the measurements of the total absolute conversion coefficient for Cs-134 indicate that one of the lower transitions may be magnetic quadrupole ((Figure 1, not shown, presents gamma-gamma correlation function of Co-60, Cs-134, and Ag-110)).

3.2.5.42 CORRELATION IN THE DIRECTION AND POLARIZATION OF TWO SUCCESSIVE QUANTA FOR Rh-106, Co-60, and Cs-134. A. H. Williams and W. L. Wiedenbeck (Randall Laboratory of Physics, University of Michigan, Ann Arbor, Michigan). Physical Review 78, 822, June 1950

Deutsch and Metzger have reported their observation of the correlation between the polarization of one quantum in the direction of emission of the other for the two successive gamma rays of Ph-106. We have used a polarimeter of a type similar to that described by Deutsch and have verified their results. In the case of Cs-134 no polarization correlation was observed. According to the theory, correlation should be found for two quadrupole transitions, one of which is

magnetic and the other electric, only if the experimental arrangement is such that one can distinguish between which of the two quanta goes to the polarimeter or to the individual counter. In our case, this discrimination was not possible. If both transitions are electric or both magnetic, this discrimination is not necessary in order to observe a correlation. If one assumes then that both transitions are quadrupole the conclusion is that one is electric and the other is magnetic.

### 3.2.5.43 THE INTERNAL CONVERSION ELECTRONS OF SEVERAL SHORT-LIVED NEUTRON INDUCED RADIOACTIVITIES.

Richard L. Caldwell (University of Missouri, Columbia, Missouri, and Argonne National Laboratories, Chicago, Illinois). Physical Review 78, 407-11, May 1950

Accurate values of transition energies were measured for seven neutron induced radioactivities using a permanent magnet beta ray spectrograph. A special vacuum gate permitted the introduction of short-lived samples within 12 seconds after the end of bombardment without disturbing the vacuum in the spectrograph. Transition energies accurate to about 1% were determined as follows: Cesium (3 hours), 128.0 kev; Cobalt (10.7 minutes), 58.9 kev; Columbium (6.6 minutes), 41.5 kev; Dysprosium (1.3 minutes), 109.0 kev; Dysprosium (2.6 hours), 87.8 kev; Iridium (1.5 minutes), 54.7 kev; Thulium (120 days), 84.4 kev. For the five isomeric transitions approximate values of the relative intensities of the electron lines were determined, and an assignment was made of the multipole outlet of the radiation, assuming electric multiple radiation. ((The following is a partial presentation of Table 1. The balance of Table 1 deals with Cobalt, Dysprosium, and Columbium and is not presented.))

TABLE 1. ELECTRON AND TRANSITION ENERGIES

Element	Half Life (hours)	Hp (Gauss-Cm)	Electron Energy (kev)	Conversion Shell	Transition Energy (kev)
Cs*	3	1067.5	92.0	K	128.0
		1251.1	123.0	L	128.0
		1273.5	127.0	M	128.0

- 3.2.5.44 COINCIDENCE AND ABSORPTION MEASUREMENTS ON CESIUM-134, IODINE-124, GOLD-199, AND CALCIUM-43. J. Lawrence Meem, Jr., and Fred Maienschein (Physics Department, Indiana University, Bloomington, Indiana). Physical Review 76, 328-32, August 1949

The beta ray spectrum of Cs-134 was investigated by absorption and beta-gamma coincidence measurements. Beta rays of 0.60 Mev and about 90 kev were observed. ((The rest of the abstract is concerned with the other isotopes and is not reproduced.))

The disintegration scheme of Cs-134 has been investigated by Elliot and Bell\*, and by Siegbahn and Deutsch\*\* using spectrometer and coincidence techniques. The spectrum of Cs-134 has been investigated ((by the authors of this article)) by absorption and beta-gamma coincidence methods. The source was obtained from Oak Ridge and was purified by an amberlite resin ion exchange column at this University. The results are in complete agreement with the disintegration scheme proposed by Elliot and Bell\*.

\*Phys. Rev. 72, 979, 1947

\*\*Phys. Rev. 71, 483, 1947

- 3.2.5.45 AECU-517 (UAC-71) nd. 1 page. TRANSITION ENERGY OF SOME NUCLEAR ISOMERS. R. L. Caldwell, E. der Mateosian, and M. Goldhaber

Accurate values of the transition energy have been measured for a number of isomers using a permanent magnet beta ray spectrograph similar to that described by Hill in Physical Review 74, 78, 1948. As special vacuum gage permits the introduction of short-lived samples. Some results are summarized in the following table: ((table reproduced in part)) Isomer; Cesium-134; half-life, 3 hours; shells from which electrons have been observed, K, L, M; transition energy  $128.0 \pm 0.5$ .

- 3.2.5.46 THE DISINTEGRATION SCHEME OF CESIUM-134. Kai Siegbahn (Nobel Institute for Physics, Academy of Science, Stockholm). Physical Review 73, 410, February 1948

Elliot and Bell have recently published a disintegration scheme for Cs-134 which is more complete than the one proposed originally by

the present authors. Some time ago we obtained results very similar to those of Elliot and Bell\* and we confirmed their disintegration scheme in every detail. (As a result of calculations, the fraction of disintegrations leading to the 1.97 Mev level in barium is  $0.26 \pm 0.08$  in excellent agreement with the values posed by Elliot and Bell). From absorption measurements using a windowless counter, we estimate the abundance of the very soft beta ray spectrum at  $0.32 \pm 0.08$ . Our best values for the gamma ray energies are now  $0.566 \pm 0.01$  Mev,  $0.603 \pm 0.01$  Mev, and  $0.798 \pm 0.15$  Mev based on the photoelectron energies from a thin uranium radiator.

\*Phys. Rev. 72, 979, 1947

- 3.2.5.47 DISINTEGRATION SCHEME OF 1.7-YEAR CESIUM-134. L. G. Elliot and R. E. Bell (Division of Atomic Energy, National Research Council of Canada, Chalk River, Ontario). Physical Review 72, 979, 1947

The disintegration of the long-lived isomer of Cs-134 has been studied with a short magnetic lens beta ray spectrometer together with coincidence techniques. The shape of the beta ray spectrum was studied down to the low energy cut-off of the counter window (about 0.015 Mev) by use of a source of thickness approximately  $0.1 \text{ mg/cm}^2$  mounted on a mica backing about  $1.0 \text{ mg/cm}^2$ . A Fermi plot of this spectrum using the exact Fermi function of (Z, N) for  $Z = 55$  is shown in Figure 1 ((not shown)). The secondary electron spectrum obtained by photoelectric conversion of the gamma rays in a thin  $17 \text{ mg/cm}^2$  lead radiator is shown in Figure 2 ((not shown)). Three gamma rays are observed whose energies are respectively  $0.568 \pm 0.015$ ,  $0.602 \pm 0.015$ , and  $0.794 \pm 0.015$  Mev with intensities of 0.26, 1.0, and 1.0, respectively, after allowing for the energy variation of the photoelectric cross section. These results are consistent with the disintegration scheme proposed in Figure 3.

- 3.2.5.48 THE DECAY SCHEME OF CESIUM-134. Kai Siegbahn and Martin Deutsch, Massachusetts Institute of Technology, Physical Review 71, 483, 1947

The long lived isomer of Cs-134 has been studied by magnetic spectrometer and coincidence techniques with a view to its use as a radioactive standard. The great majority of the disintegrations, about 95%, proceed by the emission of beta particles of  $(0.645 \pm 0.02)$  Mev maximum energy to an excited state of Ba-134 with  $(1.36 \pm 0.03)$

Mev excitation energy, followed by the successive emission of new gamma rays of  $(0.584 \pm 0.012)$  Mev and  $(0.776 \pm 0.015)$  Mev, respectively. In a few per cent of the disintegrations a gamma ray of  $(1.35 \pm 0.03)$  Mev is emitted. This is probably due to single quantum de-excitation of the same excited state.

### 3.3 CESIUM-137

#### 3.3.1 Nuclear Characteristics

3.3.1.1 Atomic Number 55

3.3.1.2 Atomic Weight 137

3.3.1.3 Half Life

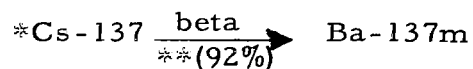
Cs-137,  $30.0 \pm 0.08$  years

Ref. 1

Ba-137,  $158 \pm 5$  seconds

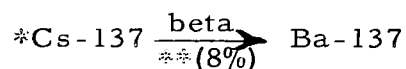
Ref. 2

#### 3.3.1.4 Energy Levels and Decay Scheme

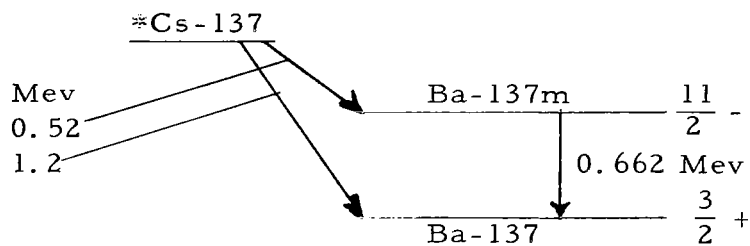


Beta<sub>max</sub> = 0.52 Mev

Gamma = 662 kev



Beta<sub>max</sub> = 1.2 Mev



\* Reference 3

\*\* Reference 4

### 3.3.1.5 Other Sources of Radiation

The 0.662 Mev gamma emitted by this isotope overshadows any bremsstrahlung produced by the 0.514 Mev beta or the low yield (8%) 1.175 Mev beta. Reference 5 suggests the following gamma radiation properties to be used for shielding purposes.

<u>Gamma Radiation Rate</u> <u>(photons/watt-sec)</u>	<u>Photon Energy</u> <u>(Mev)</u>
$6.39 \times 10^{12}$	0.662

The isotopic purity of Cs-137 in the fuel compound is approximately 35%. The rest of the fraction is composed of other cesium isotopes that are inert or have very long half lives). However, trace amounts of Cs-134 produced during the fission process by  $(n, \gamma)$  reactions can be found in the Cs-137. The amount of Cs-134 in the Cs-137 product will vary according to the age of the waste being processed and may be as much as 5% of the Cs-137 activity. (Ref. 6) However, the amount of Cs-134 activity decreases by about a factor of 2.0 each 2.1 years after reactor irradiation time.

### 3.3.2 Chemical Characteristics

#### 3.3.2.1 Fuel Forms

A summary of some cesium compounds and their physical properties which have been reported in the literature are presented in Table VI (Ref. 7). Because of the excessive chemical activity and high water solubility of cesium and most of its compounds, much effort has been devoted to the development of an inert cesium compound suitable for isotopic heat source use. The compound developed is the cesium "poly-glass", borosilicate glass in which the normal alkali, sodium, is replaced by cesium. The glass development has been carried out independently by two laboratories which report maximum cesium contents by weight at 40% (Ref. 7) and 45% (Ref. 8).

#### 3.3.2.2 Material Compatibility

As pointed out above, the low melting points and high chemical activity characteristic of cesium compounds could cause corrosion of the isotope container. However, the poly-glass eliminates any compatibility problems. By containing the cesium



in the amorphous glass matrix, it becomes inert and insoluble. The softening point of the glass is higher than that for most cesium compounds, tending to further eliminate any containment problem. In addition, the temperature of the glass must be raised well above the softening point before separation of the components can occur.

TABLE VI  
Specific Power and Power Densities for a Number  
of Cs-137 Compounds

Compound	Mol. wt.	Weight % metal	Density (g/cm <sup>3</sup> )	Specific Power (w/g)	Power Density (w/cm <sup>3</sup> )
Cs	132.91	100	1.873	0.154	0.289
CsCl	168.37	78.94	3.97	0.122	0.484
Cs <sub>2</sub> SO <sub>4</sub>	361.89	73.45	4.243	0.113	0.479
CsF	151.91	87.49	3.586	0.134	0.481
CsO · $\frac{32}{3}$ WO <sub>3</sub>	274.38	15.50	6.15	0.0239	0.147
Cs <sub>2</sub> U <sub>2</sub> O <sub>7</sub>	853.88	31.13	5.2	0.0481	0.250
Cs <sub>2</sub> TiO <sub>3</sub>	361.72	73.49	3.4	0.113	0.384
CsNbO <sub>3</sub>	273.82	48.54	3.8	0.0749	0.285
CsTaO <sub>3</sub>	361.86	36.73	5.5	0.0567	0.312
Cs <sub>2</sub> O · Al <sub>2</sub> O <sub>3</sub> · 2SiO <sub>2</sub>	503.95	52.75	3.1	0.0814	0.252

### 3.3.2.3 Effects of Impurities on Nuclear Characteristics

The normal Cs-137 fuel form has associated a high fraction of other cesium isotopes which is considered as inert. Reference 9 gives an isotopic purity of only 35 per cent. However, since the other cesium fraction is inert, these isotopes act only as diluents and do not affect the Cs-137 nuclear characteristics. References 5 and 6 indicate that impurities of Cs-134 may exist in small quantities (1 to 5%). This must be considered in most cases in shielding calculations.

### 3.3.3 Thermal and Physical Properties of Cs-137

The thermal and physical properties of Cs-137 glass compound are found in Table VII.

TABLE VII

## Thermal and Physical Properties of Cs-137 Glass

Thermal Conductivity	$\frac{\text{cal Cm}}{\text{sec } ^\circ\text{C Cm}^2}$	0.0025 (a)	Ref. 10
Specific Power	$\frac{\text{watts}}{\text{gram}}$	0.0814 (b)	Ref. 7
Thermal Output	$\frac{\text{watts}}{\text{K curie}}$	4.84	Ref. 3
Heat Capacity	$\frac{\text{cal}}{\text{gm } ^\circ\text{C}}$	0.44 (c)	Ref. 12
Weight Density	$\frac{\text{gm}}{\text{cm}^3}$	3.2	Ref. 5
		3.1	Ref. 3
Melting Point	$^\circ\text{C}$	1275 (d)	Ref. 3
Specific Activity	$\frac{\text{K curies}}{\text{gm}}$	0.087	Ref. 7

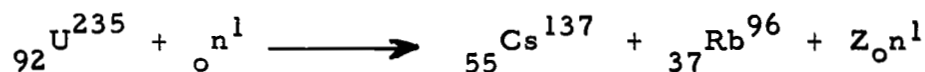
Key to Thermal and Physical Properties of Cs-137

- (a) This value holds between  $1000^\circ\text{C}$  and  $2000^\circ\text{C}$  by analogy to sodium borasilicate glass. Thermal conductivity varies with heat treatment. The value at room temperature is  $\sim .0015$ .
- (b) Computed value using a density of  $3.2 \text{ gm/cm}^3$  for the cesium glass, 16 curies Cs-137 per gm cesium glass, half life of 30 years, and 0.7882 Mev per disintegration resulting from the emission of the .52 Mev beta 92% of the time with an average energy of .159 Mev, a 1.2 Mev beta 8% of the time with an average energy of 0.403 Mev and a .662 Mev gamma 92% of the time.
- (c) This is the value for boron oxide glass between  $1750^\circ\text{R}$  and  $3250^\circ\text{R}$ .
- (d) Softening point.

### 3.3.4 Isotope Production Availability and Cost

#### 3.3.4.1 Production Method

Cs-137 is produced by fission of the heavy elements Thorium, Uranium, and Plutonium, and is obtained from the chemical processing of spent reactor fuel elements



After the separation of the gross fission product mixture from the solution containing the dissolved fuel elements the cesium isotopes are separated out by standard chemical techniques.

The cesium fraction contains the isotopes Cs-133, Cs-135, Cs-136, and Cs-137 (reference 5). After normal fuel element "cooling" times, the Cs-136 content has decayed to less than 0.1% of the cesium fraction. However, Cs-133 which is inert, and Cs-135 whose half life ( $2.1 \times 10^6$  years) is so long that it might be considered inert, represent sizable fractions of the cesium content. Thirty days after removal from the reactor, the proportions of 133, 135, and 137 are approximately equal. (Cs-135 content depends on the reactor flux.)

#### 3.3.4.2 Availability and Cost

The cesium production levels for the years 1964-1980 are tabulated below. These values were obtained from the Division of Isotope Development, USAEC, and represent the latest available from their offices.

	FISCAL YEAR							
	<u>1964</u>	<u>1965</u>	<u>1966</u>	<u>1967</u>	<u>1968</u>	<u>1969</u>	<u>1970</u>	<u>1980</u>
Mc/year	3.5	3.5	3.5	3.5	10	10	10	10
Kw(t)/year	16.8	16.8	16.8	16.8	48	48	48	48

As with the other fission product isotopes, it is expected that by 1970 commercial power capabilities will have increased the availability of Cs-137 to 62 kw(t)/year and by 1980 to as much as 410 kw(t)/year (Reference 10).

Present costs of this isotope are given by Reference 13 as \$0.75/curie and projected to \$0.50/curie.

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2. On the Cesium-137 Disintegration Scheme. Physical Review 75, 197, Jan. 1949.
3. Personal Communication with S. J. Rimshaw, ORNL. 5 December 1965.
4. The Total Internal Conversion Coefficient of the 0.662 Mev Transition of Ba-137 and Branching Ratio in the Decay of Cs-137. J. S. Merritt, G. Taylor, S. C. Misca. Physics in Canada 20:37-38 (1964).
5. Handbook of Shielding Requirements and Radiation Characteristics of Isotopic Power Sources for Terrestrial, Marine, and Space Applications, ORNL-3576.
6. Oak Ridge National Laboratory Catalog on Radio and Stable Isotopes, TID-18870.
7. Power Densities of a Number of Source Compounds, S. J. Rimshaw (Oak Ridge National Laboratory, Oak Ridge, Tennessee). ORNL-3741.
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13. Radionuclide Power for Space, Nucleonics, Vol. 21, No. 3, March 1963.

### 3.3.5 Annotated Bibliography

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- 3.3.5.2 DECAY OF CESIUM-137 DETERMINED BY ABSOLUTE COUNTING METHODS. Janet S. Merritt and J. G. V. Taylor (Atomic Energy of Canada Ltd., Chalk River, Ontario). Anal. Chem., 37: 351-4 (March 1965)

Precise decay data for Cs-137 were determined by absolute counting techniques aided by a rapid method for the carrier-free separation of Ba-137m. The following values for the decay parameters were obtained: half life of Ba-137m,  $2.554 \pm 0.003$  minutes; total internal conversion coefficient of the 662-keV transition,  $0.1100 \pm 0.0011$ ; K-conversion coefficient,  $0.0894 \pm 0.0010$ ; branching ratio for the decay of Cs-137 to Ba-137m,  $0.952 \pm 0.010$ ; ratio of 662 keV gamma emission to Cs-137 disintegration rate,  $0.857 \pm 0.009$ . The relative uncertainty of  $\sim \pm 1\%$  obtained for the latter ratio should make possible the absolute assay of Cs-137 by gamma spectrometry to  $\pm 2\%$  or better.

- 3.3.5.3 Refer to 3.1.5.2. HALF-LIVES OF Sr-90 AND Cs-137. K. F. Flynn, L. E. Glendenin, A. L. Harkness, and E. P. Steinberg (Argonne National Laboratories, Illinois). J. Inorg. Nucl. Chem., 27: 21-3 (January 1965).
- 3.3.5.4 ORNL-3750. ANALYTICAL CHEMISTRY DIVISION ANNUAL PROGRESS REPORT FOR PERIOD ENDING NOVEMBER 15, 1964. (Oak Ridge National Laboratory, Tennessee). Jan. 1965. Contract W-7405-eng-26. 144p

((An IAEA Cs-137 standard intercalibration was done. New half-life values for As-76, Cs-137, La-140 and U-232 were measured.))

- 3.3.5.5 SECOND FORBIDDEN BETA DECAY AND THE GROUND STATE TRANSITIONS OF Cl-36 AND Cs-137. P. Lipnik (Center de Physique Nucleaire, Heverle-Louvain, Belgium). p. 331-5 of "Selected Topics in Nuclear Spectroscopy." New York, John Wiley and Sons, 1964.

Formulas for the spectrum shape factor and the longitudinal polarization of electrons were derived in the Konopinski-Uhlenbeck approximation. The ground-state beta transitions of Cs-137 were analyzed by the shape factor relation and a defined parameter.

- 3.3.5.6 DEMONSTRATION OF A PARENT-DAUGHTER RADIO-ACTIVE EQUILIBRIUM USING Cs-137 - Ba-137m.  
Gregory R. Choppin and Carson L. Nealy (Florida State University, Tallahassee). J. Chem. Educ., 41: 598-600 (Nov. 1964).

It is possible to show the relation between radioactive half life and both the rate of decay and of growth of a radioactive daughter with the Cs-137 - Ba-137m system. It also has the advantage of a short-lived daughter separable by simple techniques without requirement of an activation source. Furthermore, since gamma emission is associated only with the Ba-137m, its behavior can be followed directly by scintillation counting with a NaI(Tl) crystal in the presence of Cs-137.

- 3.3.5.7 (SAN-366-1) THE CESIUM-137 POWER PROGRAM.  
Final Report. (Royal Industries, Inc., Pasadena, California). May 1964. Contract AT(04-3)-366. 242p.

Activities in a program to design a compact and lightweight thermoelectric generator, power by Cs-137 to safely provide uninterrupted power, in the five watt (e) range, to an undersea seismic station are described. The Cs-137 thermoelectric generator was not capable of delivering 5 w. The other design objectives met during the research are summarized.

- 3.3.5.8 ISOTOPIC POWER DATA SHEETS, Compiled by S. J. Rimshaw, Oak Ridge National Laboratory, Oak Ridge, Tennessee. For presentation at Industry Information Meeting on Isotopic Power Development and Applications, Washington, D. C., May 18-19, 1964

#### Cs-137 Glass

Source Material	Cesium Borosilicate glass
Half-Life	Cs-137 - $30 \pm 0.3$ years (Ba-137m - 2.6 min)

Decay and Radiation Properties	Cs-137 $\longrightarrow$ Ba-137m		
	Beta - Cs-137	Gamma	Other
	0.51 (92%) 1.17 (8%)	none	none
	Beta - Ba-137m	Gamma - Ba-137m	
	none	0.662	IT
Isotopic Composition	43.4% Cs-133, 20.1% Cs-135, and 36.5% Cs-137		
Activity Concentration	16 curies/g of glass		
Radiochemical Purity	> 95%. The only significant radiochemical impurity is Cs-134. The amount of Cs-134 in the Cs-137 product will vary according to the age of the waste being processed; it may be as much as 5% of the Cs-137 activity but in currently available feed material is < 3%.		
Chemical Purity	> 98% cesium, < 2% Rb, small quantities of Na, K, SiO <sub>2</sub> introduced in reagents.		
Specific Power	0.0774 watts per gram of glass or 4.84 watts/kilocurie		
Thermal Energy	207 curies per thermal watt		
Density	3.1 g/cm <sup>3</sup>		
Power Density	0.24 watts/cm <sup>3</sup> based on 16 curies/g of glass		
Thermal Conductivity	Estimated to be $1-2 \times 10^{-3}$ cal/sec·cm·°C at room temperature and $2.5-3 \times 10^{-3}$ cal/sec·cm·°C between 1000°C and 2000°C by analogy to sodium borosilicate glasses		
Coefficient of Expansion	Estimated to be $1-2 \times 10^{-5}$ °C <sup>-1</sup> (a factor of ~ 4 less than ordinary glass)		
Softening Point	~ 1275°C		

**Mechanical Strength**                      The Cs-137 glass has exhibited brittleness. No definitive measurements of mechanical strength have been made

**Thermal and Radiation Stability**                      The glass devitrifies if held at 1000°C for a few hours

Radiation Attenuation	Dose Rate, rads/hr at 100 cm	Shielding in Centimeters of Uranium Required for a Cs-137 Source Strength of (Source contains 1% Cs-134 and 99% Cs-137)		
		100 w	1000 w	10,000 w
	100	1.5	3.4	4.7
	10	3.6	5.5	7.2
	1	6.0	8.1	9.8
	0.1	8.4	10.8	12.6

**Gas Evolution Due to Radioactive Decay Processes**                      None

**Leach Rate**                       $\sim 0.2 \text{ mg/cm}^2 \cdot \text{day}$  in  $\text{H}_2\text{O}$  at 30°C

**Vapor Pressure**                      Volatility of Cs at 1200°C is  $\sim 3.5 \text{ mg/cm}^2 \cdot \text{hr}$  ( $\sim 12 \text{ mc of Cs-137/cm}^2 \cdot \text{hr}$ )

**Resistance to Thermal Shock**                      Poor

**Burnup Characteristics**                      Does not oxidize; has ablative characteristics of glass

**Capsule Compatibility**                      Excellent with usual capsule materials such as stainless steel and Hastelloy "C".

### 3.3.5.9      GAMMA-RAY ANGULAR CORRELATION ATTENUATION FACTORS.      B. Herskind and Y. Yoshizawa (Universitet, Copenhagen).      Nucl. Instr. Methods, 27: 104-8 (Apr. 1964)

Gamma attenuation factors for angular correlations and angular distributions were computed from total absorption coefficients for a 76 x



76 mm NaI(Tl) crystal. The calculations cover the range of distances between a source and a crystal surface from 1 to 20 cm and the range of energies from 0.05 to 4.0 Mev. In order to determine attenuation factors for the photopeak alone, measurements were performed for gamma rays of 0.060 Mev (Am-241), 0.142 Mev (Ce-141), 0.32 Mev (Cr-51), 0.662 Mev (Cs-137), 1.332 Mev (Co-60), and 2.754 Mev (Na-24) within the same range of distances. The attenuation factors for the photopeak were computed by using the experimental results. The experimental factors are compared to the results calculated from the total absorption coefficients and to the results of the Monte Carlo calculation given by Yates.

- 3.3.5.10 IN-TIME INTEGRATED FISSION RATE STUDIES IN URANIUM USING ALPHA OR GAMMA SPECTROMETRY. A. Guillon, S. May, S. Sapena, and R. Sauvagnac, Bull. Inform. Sci. Tech. (Paris), No. 81, 35-7 (Mar. 1964) (In French)

Some methods for measuring burn-up of fissile material are reviewed. Alpha spectrometry after irradiation (measurement of U-235 disappearing), which is accurate only for high integrated fluxes, is discussed. Measurements of the ratio of fission products to uranium are also considered. Some examples are given for irradiations made under different conditions. The fission products studied were Cs-137, Ce-144, and Ba-140. A chemical separation is described for each of these radionuclides.

- 3.3.5.11 THE STRUCTURE OF THE GAMMA-FIELD OF A CESIUM-137 SOURCE. V. A. Vorob'ev. At. Energ USSR, 16:69-70 Jan. 1964 (In Russian)

The angular spectrum function of scattered gamma radiation from an infinite, isotropic plane Cs-137 source, having an  $E_0 = 0.661$  Mev in an air-equivalent medium, was investigated by the Monte Carlo method at distances corresponding to 0.5, 1, and 2 mean free paths (MFP) from it. The surface activity was assumed to be 1 quantum/cm<sup>2</sup> sec. At 0.5 MFP from the source the chief contribution of the scattered radiation was noted at a body angle of about 75°; at 1 MFP the maximum was located at 60°; and at 2 MFP it was found at 0°. At the shortest distance, carrying out the observations under 0°, in the direction of normal, a peak was noted at the 0.3 Mev region in the energy spectrum; this was due to the first-generation gamma quanta emitted at about 70°. The integrated spectrum of the plane source was similar to the

integrated spectra of point sources in a homogeneous medium. The buildup factors calculated agreed well with the values reported by H. Goldstein.

3. 3. 5. 12 IRRADIATION SOURCES FOR THE NON-DESTRUCTIVE TESTING OF METALS. E. Becker, Technik (Berlin), 18: 793-9 (Dec. 1963). (Rev. Metal Lit., 21; No. 5, March 1964) (In German)

Physical properties of common irradiation sources used in the testing of metals, such as Co-60, Cs-137, Ir-192, and Tm-170 isotopes, and their application in the testing of steel were investigated. The defect detectability was determined and optimum wall thickness ranges for the individual isotopes were investigated.

3. 3. 5. 13 CALCULATION OF THE SPECTRAL-ANGULAR DISTRIBUTION OF SCATTERED GAMMA QUANTA FROM A MONODIRECTIONAL CESIUM-137 SOURCE IN IRON. L. P. Kimel<sup>1</sup>, A. M. Panchenko, and V. P. Terrent'ev. At. Energ. USSR, 15: 328-31 (Oct. 1963) (In Russian)

The Monte Carlo method was used for calculating the spectral and angular distribution of the scattered energy of gamma quanta, emitted with an initial energy of 0.661 Mev by a Cs-137 point source placed in an infinite Fe medium. The 5420 measurements were carried out by determining the path of the quanta up to the first interaction, determining the type of the interaction, and calculating the energy of the scattered gamma quantum by the Compton effect; at the same time the direction of the flight path after scattering was also determined. The measurements were repeated until the quantum disappeared as the result of the photoeffect or because of transition to an energy level below 10 kev. Angular integration of the scattered radiation spectra showed that the energy of the quanta is limited within 0.13 to 0.26 Mev. The data obtained made it possible to calculate the buildup factor  $B_E$  of a flat, monodirectional source.

3. 3. 5. 14 Refer to 3.2.5.10. HALF LIVES OF CESIUM-137 AND CESIUM-134 AS MEASURED BY MASS SPECTROSCOPY. Anal. Chem. 7, 797-99, June 1963

3. 3. 5. 15 THE SHAPES OF THE SCINTILLATION GAMMA-RAY SPECTRA AND MEASURING CONDITIONS. Shinji Okano (Inst. of Physical and Chemical Research, Tokyo). Radioisotopes (Tokyo), 12: 261-5 (May 1963) (In Japanese)

Scintillation gamma-ray spectra of various nuclides are examined. It is shown that the shapes of the spectra are varied, even for the same nuclide, by the geometries and dimensions of the gamma-ray sources and the phosphors and their relative positions. Phosphors of NaI(Tl) with varying geometries are used. The gamma-ray sources examined are Na-22, Mn-54, Co-60, Zn-65, and Cs-137. The counts vs wave-height curves are shown. (From NSA of Japan)

- 3.3.5.16 MASS-SPECTROMETRIC DETERMINATION OF MICRO-QUANTITIES OF CESIUM. I. V. Burovina, V. P. Nesterov, and D. G. Fleishman. Radiokhimiya, 5: 272-6 (1963) (In Russian)

Mass-spectrometric method was used for determining Cs-133 in biological objects. The half life of Cs-137 was determined as  $30.1 \pm 0.7$  years. Data are given on Cs-133 content in a human brain and muscle.

- 3.3.5.17 THE MASS SPECTROMETRIC DETERMINATION OF THE CESIUM-137 HALF LIFE. B. F. Rider, J. P. Peterson, Jr., and C. P. Ruiz. (General Electric Company, Atomic Power Equipment Department, Vallecitos Atomic Laboratory, Pleasanton, California). Nuclear Science and Engineering 15, 284-7 March 1963

The half-life of Cs-137 has been measured by observing the rate of growth of stable Ba-137 from a known quantity of Cs-137 by the use of isotope dilution mass spectrometry. The half life thus obtained is  $29.2 \pm 0.3$  years.

Sample preparation. A 200 millicurie batch of fission product Cs-137 in 5 milliliters of one mole HCL was passed through a Dowex 50 column. The column was washed with an additional 15 milliliters of one mole HCL. A small fraction of this eluate was mass analyzed and the remainder was added to a polyethylene bottle containing accurately weighed amounts of standardized Cs-133 and enriched Ba-138 solution. A number of Cs-137 atoms in this master solution was determined to be a standard deviation of 0.68% from 183 mass scans from ten filaments. (The value of  $29.2 \pm 0.3$  years is different to a small extent from a previous determination by mass spectrometry\*.) The difference from the previous mass spectrometric value can be accounted for primarily by a difference in the measurement of the slope of the Ba-137 growth line. In this work this slope was obtained

with improved accuracy by making many observations over a longer period of time, and by eliminating the need for chemical loss correction.

\*Farrar, et al, Canadian Journal of Chemistry, 39 page 681, March 1961.

- 3.3.5.18 THE BETA SPECTRUM OF Cs-137. H. Daniel and H. Schmitt (Max-Planck-Institut für Kernphysik, Heidelberg). Z. Physik, 168: 292-7 (1962) (In German)

The beta spectrum of Cs-137 was measured with an iron-free spectrometer. The energy of the soft component was found to be  $E_0 = 514 \pm 2$  kev. The parameter  $k$  of the hard component shape factor  $C_{2VA}^{(2)}(W) = q^2 + kp^2$  was determined to be  $k = 0.015 \pm 0.004$ . The beta intensities amount to 6.5% (hard component) and 93.5% (soft component). The  $K$  conversion coefficient of the isomeric transition was measured to be  $\alpha_k = 0.093 \pm 0.003$ , in excellent agreement with Sliv's theoretical value.

- 3.3.5.19 THE HALF-LIFE OF CESIUM-137. D. G. Fleishman, I. V. Burovina, and V. P. Nesterov, Atomnaya Energiya, Vol. 13, No. 6, 592-3, December 1962

The half life of a radioactive isotope can be determined by various approaches, by measuring the absolute activity of a known number of its atoms in terms of the decrease in the amount of the radioisotope under study as time progresses, or in terms of the rate of accumulation of radioactive daughters. In the present note, the rate of decay of the Cs-137 in the sample is reported as a result of measuring the beta activity in a liquid scintillator in the amount of Cs-137 as found by the isotope dilution method. To measure the beta activity, a certain quantity of an aqueous solution of Cs-137 nitrate was introduced into a liquid scintillator. Approximately 0.3 milliliters standard solution was added to the scintillator constituting in total 1.5 of the total volume of the scintillator and depressing its light yield only negligibly. The fairly high light yield of the scintillator and the use of the FEU-1B photomultiplier tube, with excellent integrated sensitivity and a low tube noise level, made possible an efficiency of the order of 93% in recording Cs-137 betas

The value of the half-life of Cs-137 found from the above data is  $30.1 \pm 0.7$  years. This value agrees within the limits of the error with the results reported by other authors in the recent literature ( $29 \pm 1$  year\*,  $30.4 \pm 0.4$  years\*\*).

\*Atomnaya Energiya, Vol. 10, No. 6, 622-3, June 1961

\*\*Canadian Journal Chemistry 39, 681-3, March 1961

- 3.3.5.20 HALF LIFE OF CESIUM-137. B. F. Rider, J. P. Peterson, Jr., and C. P. Ruiz (GE-APED, Pleasanton). Transactions of the American Nuclear Society 5, 1, 195-6, June 1962

The half-life of Cs-137 has been determined from the rate of formation of daughter Ba-137 in a measured amount of Cs-137 by isotopic dilution mass spectrometry. Only one such measurement;  $30.4 \pm 0.4$  years has been reported\*. Other measurements based on radioactivity measurements include  $26.6 \pm 0.4$ ,  $30.0 \pm 0.4$ , 28.6,  $28.4 \pm 1.4$ ,  $32.6 \pm 1.6$ , 27, and  $29 \pm 1$  year. Because of the uncertainty of this value and its importance in nuclear fuel burn-up analysis by the Cs-137 to uranium ratio method a re-investigation was undertaken using the mass spectrometer method. The Cs-133 solution was a primary standard prepared by weight from spectrographically pure CsCl. The Ba-138 was standardized by isotopic dilution with a primary standard prepared by weight from spectrographically pure CsCl. The Ba-138 was standardized by isotopic dilution with a primary standard prepared by weight from analyzed CP natural  $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$ . By isotopic dilution the initial number of Cs-137 and Ba-138 atoms in the flask were known to a standard deviation of 0.56% and 0.43%, respectively. Two solid emission mass spectrometers and a two-stage magnetic analyzer using an electron multiplier detector and pulse counting technique and a single stage magnetic analyzer with a Faraday caged detector and vibrating reed electrometer were used to determine the mass ratios.

((From this experiment)) the half life was determined to be  $29.15 \pm 0.25$  years at one standard deviation. This value is consistent with the average of the values referenced and has a smaller uncertainty than previous measurements.

\*Canadian Journal Chemistry 39, 681-3. March 1961.

- 3.3.5.21 MEASUREMENTS OF INTERNAL CONVERSION COEFFICIENTS. Solve Hultberg, Daniel J. Horen, and Jack M. Hollander (Lawrence Radiation Laboratory, University of California, Berkeley, California). Nuclear Physics, 28, 471-7. 1961.

Internal conversion coefficients of M1, M4, and E2, transitions from the Y-87, Cs-137, Au-198, Ir-192, decays have been studied by using the internal-external conversion (IEC) method. For unhindered transitions of the  $2+ \longrightarrow 0+$  type we have found  $(\text{Alpha}_2)_{317} = 0.054 \pm 0.003$  for the 317 kev transition from Ir-192 and  $(\text{Alpha}_2)_{412} = 0.028 \pm 0.0015$  for the 412 kev transition from Au-198, the theoretical values being 0.054 and 0.030, respectively.

- 3.3.5.22 ANALYSIS OF GAMMA RAY SCINTILLATION SPECTRA BY THE METHOD OF LEAST SQUARES. L. Salmon (Physics Division, UKAEA Research Group, Atomic Energy Research Establishment, Harwell, Didcot, Berks). Nuclear Instruments and Methods 14, 193-199, December 1961

The method of least squares has been applied to the analysis of gamma ray scintillation spectra, the calculations being performed on a Ferranti "Mercury" computer. The method is superior to the usual graphical methods of quantitative interpretation. Analytical results are obtained rapidly, the accuracy being consistent with calculated precision. The method is particularly suitable for routine analysis at low counting rates.

- 3.3.5.23 DECAY HALF-LIFE OF CESIUM-137. M. P. Glazunov, A. I. Grivkova, B. A. Zaitsev, and V. A. Kiseley, Atomnaya, Energiya, Vol. 10, No. 6, 622-3, June 1961

The Cs-137 precipitation used in the present study was separated from a solution of uranium fission products by the ferro-cyanide method which is based on the ability of cesium to enter into a highly insoluble compound with nickel ferro-cyanide. The separated preparation was a solution of cesium chloride. An assessment of the radio chemical purity of the cesium shows that gamma-radiation due to radioactive impurities was less than 0.01% of the total activity. This was affirmed by mass spectrometric and beta-gamma spectrometric techniques. ((The value for the decay life of Cs-137 was determined by these authors to be  $29 \pm 1$  year.))

- 3.3.5.24 CESIUM-137 AND ITS GAMMA-RADIATIONS IN TELE-RADIO THERAPY. R. Thoraesus, Acta-Radiologica, 55, 385-95, May 1961

A review of the characteristics of cesium radioactivity including partial comparison with that of cobalt is given. The measurement of cesium gamma-radiation by means of an extensively calibrated sub-standard is described and the experimental results particularly the half-value layers of some materials are presented. Certain clinically important properties of the radiation, the radiation protection using radioactive cesium, and the result of determination of the peak energy of the roentgen radiation equivalent in penetration to the cesium gamma-radiation are discussed.

- 3.3.5.25 HALF-LIFE OF CESIUM-137. H. Farrar, A. K. Dasgupta and R. H. Tomlinson. Canadian Journal of Chemistry 39, 681-3, March 1961

The half-life of Cs-137 was found to be  $30.4 \pm 0.4$  years by determining the amount Ba-137 produced in various times from a known number of Cs-137 atoms. The number of barium and cesium atoms were determined from isotope dilution data obtained with a mass spectrometer.

About 0.5 milligrams of fission product cesium was used for this experiment amounting to about 20 millicuries of activity in generating about  $1 \times 10^{-8}$  grams of Ba-137 per day. Measurements of the amount of Ba-137 formed in 8 different periods have been made. The period in each case was the time between similar parts of consecutive cesium elutions. From these data ((contained in the article)) the half-life was calculated to be  $30.4 \pm 0.4$  years. The errors in the half-life has been estimated from the assigned errors in each of the measured quantities.

- 3.3.5.26 A LARGE VOLUME FOUR PI LIQUID SCINTILLATION DETECTOR. B. G. Dunavant and J. E. Christian (Bionucleonics Laboratory, Purdue University, LaFayette, Indiana). International Journal of Applied Radiation and Isotopes 8, 223-7, October 1960

This detector has a scintillator volume of one cubic foot and sample chamber  $4\frac{1}{3}$  inches in diameter and 8" long. Four 5" photo-multiplier tubes give a photocathode coverage of about 10% of the detector's wall area. Good light collection efficiency results in the counter being relatively insensitive to source position in the sample chamber. The detector is housed in an iron shield providing 5 inches of shielding. Such detectors as the one described should find wide application particularly in biological studies and clinical diagnosis. ((Cs-137 was used as a calibration standard for the detector described in this article.))

- 3.3.5.27 INTERACTION OF BETA DECAY AND SECOND FORBIDDEN SPECTRUM OF CESIUM-137. Toshio Katoh, Shuichiro Yamasaki and Yasukazu Yoshizawa. (Department of Physics, Faculty of Science, Osaka University, Osaka, Japan.) Nuclear Physics 17, 548-62, July 1960

We discussed the allowed as well as non-unique first forbidden beta-spectra and in particular the second forbidden spectra of Cs-137, taking into account parity non-conservation. Our analysis can be summarized in the following way: From the consideration of Fierz terms in allowed as well as forbidden transitions, we can treat the two combinations STP and VA separately. Though a linear combination of the five interactions is possible no interference term between STP and VA exist. In the discussion of non-unique first forbidden transitions the combination VA is more favorable than STP. From the second forbidden spectrum of Cs-137 we see that  $\text{Re}(C_x C_y + C_x' C_y') / 1 C_y l^2 \simeq 1 C_y' l^2 + 1 C_x' l^2$  where x and y mean s and f or v and a.

- 3.3.5.28 THE DESIGN AND APPLICATION OF A DEMOUNTABLE FOUR PI GEIGER MEULLER COUNTER. L. K. Burton, and N. G. Trott (Physics Department, Institute of Cancer Research, Royal Cancer Hospital, London) International Journal of Applied Radiation and Isotopes 8, 20-8, May 1960.

An improved four Pi GM counter has been designed from which the cathode can be easily removed leaving the anode wire intact and still under tension. Counters of this type have been used successfully for several years in the measurement of radioactive nuclide standards and results of intercomparisons of standards with other laboratories are presented. The counters have also been used in conjunction with gamma ionization measurements in determining the specific gamma position (k factor) of Fe-59, Cs-137 and Ir-192. ((Applications of these counters in biological and medical work are described.))

- 3.3.5.29 ON THE DETERMINATION OF ABSOLUTE INTERNAL CONVERSION COEFFICIENTS BY THE COMPARISON OF CONVERSION LINES AND PHOTO LINES. AN APPLICATION TO THE 662-KEV TRANSITION IN THE DECAY OF CESIUM-137. Solve Hultberg and Rune Stockendal, Arkiv for Fysik, Vol. 14, No. 36, 565-77, 1959.



The method for the determination of absolute values of internal conversion coefficients by the beta spectrometric comparison of conversion line and photo line intensities has been studied. The deduction of gamma ray intensities from measured photo lines is treated and it is shown that experimentally determined photoelectric distributions can be used in the theoretical formulae to a good approximation. The instrument used was a flat double focusing beta ray spectrometer and general expressions are given for a source converter assembly of cylindrical symmetry about the spectrometer axis. An application was made to the 662-kev, 4M transition in Ba-137 and the conversion coefficient was found to be  $0.093 \pm 0.006$ . A  $2.19 \pm 0.02 \text{ mg/cm}^2$  uranium converter was used.

The activity ((used in the subject experiment)) consisted of 1 milli-curies of Cs-137 in one normal nitric acid and was obtained from the Radiochemical Center, Amersham. From this solution two radioactive sources were prepared; a strong one on a platinum backing by drop-wise evaporation and a weak one by vacuum evaporation. The sources were circular with a diameter of 6 millimeters. The weak source was intended for the internal conversion measurements and the strong one for recording photoelectrons from a uranium converter for the determination of the gamma ray intensity.

3.3.5.30 CONVERSION OF AMPLITUDE DISTRIBUTIONS INTO ENERGY SPECTRA. Yu A. Kazanskii, Pribori i Tekh, Eksptl, 4, 32-4, July - August 1959

The instrumental spectrum of pulse amplitudes and the efficiency of a single crystal scintillation gamma-spectrometer with a CsI(Tl) crystal were investigated. Using the data obtained, a number matrix was constructed by means of which using the method of successive subtractions, the amplitude pulse distributions were converted into energy spectra. For certain energy values of gamma radiation the efficiency,  $E_{pi}$ , of the spectrometer was determined by means of Cs-137, Cs-134, and Co-60 gamma radiation sources of known activity. The sources were placed at a certain distance from the crystal and a number of pulses in the photopeak were counted. Knowing the number of gamma quanta of given energy which were emitted by the source in unit time and by calculating the solid angle of the columnator it was possible to determine efficiency values for  $E = 0.661$ ,  $0.80$ , and  $1.33$  and  $1.36$  Mev which are shown by the dotted line in Figure 4. ((Figure 4 is not shown.))

- 3.3.5.31 BETA AND GAMMA RAYS SPECTROSCOPY OF CESIUM-137. Yasukazu Yoshizawa (Department of Physics, Faculty of Science, Osaka University, Osaka, Japan). Nuclear Physics 5, 122-140, Jan. 1958.

Beta rays and internal conversion electrons from Cs-137 have been studied using a 2-directional focusing beta ray spectrometer (mean radius = 40 centimeters). The K shell conversion coefficient  $\alpha_K$  of 661 kev gamma transition has been determined to be  $0.0976 \pm 0.0051$  and K/L/M to be  $(5.66 \pm 0.04)/1/(0.260 \pm 0.003)$  which are in good agreement with Rose's calculated values for the M4 transition. The maximum energy of the lower component of the beta rays has been found to be  $514 \pm 2$  kev. The curie plot, when corrected by the factor of the unique first forbidden transition, shows a straight line. The spectrum of the higher component was interpreted by the second forbidden correction factor with a linearity combination of scalar and tensor interaction. In an attempt to determine the sign of the ratio of scalar to tensor interaction constants,  $G_S/G_T$ , the spectrum could be only explained by the negative sign, assuming that the single particle model is a good approximation for the nuclear matrix element.

Cs-137 as cesium-chloride was deposited on an aluminum foil of 1.8 mg/cm<sup>2</sup> thickness. The average thickness of the sample was about 0.2 mg/cm<sup>2</sup> and an acetic acid solution of insulin was used to make the source uniform. The beta ray and internal conversion spectra are shown in Fig. 1 where a strong conversion line is exhibited.

- 3.3.5.32 Refer to 3.2.5.19. RAPID ANALYSIS OF GAMMA EMITTERS USING A GAMMA-RAY SCINTILLATION SPECTROMETER. Chem. Soc. of Japan (Bull) 30, 583-5. Sept. 1957.
- 3.3.5.33 PERFORMANCE OF A BETA SCINTILLATION SPECTROMETER SPECTRA AND CONVERSION COEFFICIENTS OF CESIUM-137 AND BISMUTH-207. R. A. Ricca (Instituut Voor Kernphysisch, Onberzuek, Amsterdam, Nederland, Physica, 23, 693-703. July 1957)

The performance of a beta scintillation spectrometer (anthracene crystal) in nuclear spectroscopy has been investigated. A method of calibration of this spectrometer, its resolution and its response to different energies are reported. The nuclides Cs-137, Au-198, Bi-207, which have well known conversion lines, were used for the

calibration. The function  $(\Delta E)^2/E$  was plotted against the energy and found to be linear in the energy region considered. The spectra of P-32, Co-60, Au-198, and Cs-137 were analyzed. The total conversion coefficient of the 0.662 Mev gamma ray in the decay of Cs-137 was computed and found to be  $0.114 \pm 0.022$ . The K conversion coefficients of the two stronger gamma rays in the decay of Bi-207 were determined with the help of the measured gamma scintillation spectrum and found to be 0.019 (0.57 Mev transition) and 0.114 (1.06 Mev transition).

We have tried to determine the total conversion coefficients of the 0.662 Mev gamma ray following the decay of Cs-137 and the decay coefficients of the two stronger gamma rays in the decay of Bi-207 with the help of the scintillation spectrometer. Cs-137 (Figure 7) shows the beta and conversion spectrum of Cs-137 ((Fig. 7 not shown)). The total conversion coefficient  $\alpha_{TOT}$  can be expressed as follows:  $\alpha_{TOT} = x/(1-x)$ ;  $x = N_e/N_{BETA} = S_E \int_{BETA}^{\infty} E^{-1} dE$ . Where  $S_E$  and  $S_{BETA}$  are the areas under the conversion peak in the corresponding data spectrum. (Corrected for back scattering by extrapolation of the curie plot.)  $\int_{BETA}^{\infty} E^{-1} dE = E_{BETA}^{-1}$  we found  $\alpha_{TOT} = 0.114 \pm 0.222$ .

- 3.3.5.34 BETA INTERACTION IN THE DECAY OF CESIUM-137. Toshio Katoh, Masao Nozawa Yasukazu Yoshizawa (Department of Physics, Faculty of Science, Osaka University, Osaka) and Yujiro Koh (Faculty of Science Engineering, Osaka City University, Osaka). Journal of the Phys. Soc. of Japan 12, 738, June 1957

Recently it has been said that the ratio of  $|G_s|/|G_t|$  in beta interaction is close to one. Its sign, however, has not been determined although some attempts have been done. The present paper concerns the trial for this problem investigating the spectrum of the higher components of the beta rays from Cs-137. A two-directional focusing beta ray spectrometer of mushroom type with the mean radius of 40 centimeters was used. The resolution of the spectrometer was 0.17 to 0.5%. It could be concluded from the measurements of P-32 and Y-90 spectra that the distortion due to the spectrometer was negligible. The thickness of the source was about  $0.2 \text{ mg/cm}^2$ .

- 3.3.5.35 THE HALF LIFE OF CESIUM-137. F. Brown, G. R. Hall, and A. J. Walter (Atomic Research Establishment, Harwell, Didcot, Berks). Journal of Inorganic and Nuclear Chemistry 1, 241-7, Oct. 1955

The beta decay half life of Cs-137 has been obtained from a specific activity measurement. The disintegration rate was measured in a 4 pi type proportional counter. The number of atoms of Cs-137 was measured by mass spectrometry using the isotopic dilution method. The result is  $30.0 \pm 0.3 - 0.4$  years. No allowance is made in this error for uncertainties in the decay scheme used.

((This paper describes how the half life has been obtained by means of a specific activity determination.)) The Cs-137, a cesium-sulphate solution was obtained from the Radiochemical Center, Amersham, Bucks. This cesium was of fission product origin and therefore besides Cs-137 it contained Cs-133 (stable), Cs-136  $3(10)^6$  years, and a small quantity of Cs-134 (2.3 years). The radiochemical purity was such that not less than 99.9% of the activity was due to Cs-134, Cs-137, and Ba-137m (the daughter of Cs-137). Cs-135 contributed to the mass but hardly at all to the activity. A master solution of this material was prepared containing about 10 micrograms per milliliter of cesium and made slightly acid with sulfuric acid. From this solution a number of dilutions were prepared using water containing various concentrations of natural cesium as carrier.

3.3.5.36 HALF-LIFE OF CESIUM-137. D. M. Wiles and R. H. Tomlinson (Department of Chemistry, Hamilton College, McMaster University, Hamilton, Canada). Physical Review 99, 188, July 1955

The half-life of Cs-137 was found to be  $26.6 \pm 0.4$  years by observing the disintegration rate of a known number of atoms\*. The disintegration rate was measured with a 4 pi proportional counter and the number of atoms was determined from isotope dilution data obtained with a mass spectrometer.

A solution of carrier free Cs-137 was prepared from fission products by ion exchange techniques. Disintegration rate per unit volume of this solution was determined with a 4 pi proportional counter and the number of atoms of Cs-137 by means of isotope dilution with a mass spectrometer. (The value of 26.6 years is considerably lower than the currently expected value of  $33 \pm 2$  years, calculated from comparison of the mass spectrometric ratios of Cs-133/Cs-137, in fission products differing up to 5 years in age.)

\*((Presently accepted value = 30.0 years.))

- 3.3.5.37 INVESTIGATION OF THE NUCLEAR ISOMERISM OF Zn-69\*, Se-79\*, Se-81\*, Rn-103\*, and Ba-137\*. G. M. Drabkin, B. I. Orlov, and L. E. Rusinov, Izvestia Akad Nauk, SSSR 12, 19, 324-37, May-June 1955

The radioactive isotopes Cs-137 decays by beta emission to Ba-137,  $T = 33$  years. In 98% of a total number of disintegrations the beta decay of Cs-137 leads to the formation of the metastable Ba-137 nuclei which has a half life of 2.6 minutes. The electron spectrum of Cs-137 and Ba-137 obtained by us is shown in Fig. 9. The continuous beta spectrum and end point energy of  $540 \pm 10$  kev belongs to Cs-137. The K, L, and M lines are associated with the isomeric transitions of Ba-137\* of energy  $661 \pm 2$  kev. The experimental value of  $K/L = 6.0 \pm 0.1$  is in agreement with the value of 6 for M4 transitions from the curve of Goldhaber and Scharf. The decay scheme for Cs-137 is pictured in Fig. 9. The state of the stable Ba-137 nucleus is  $d_{3/2}$ ; the metastable state of Ba-137\* is  $h_{11/2}$ .

- 3.3.5.38 NUCLEAR MATRIX ELEMENT FOR BETA DECAY AND THE RATIO OF COUPLING CONSTANTS. Jun-Ichi, Fujita (Department of Physics, Faculty of Science, University of Tokyo). Progress of Theoretical Physics, 13, 3, 260-4, March 1955

The nuclear matrix elements for the beta decay in the second and third forbidden transitions are derived on Bohr's collective model. The results are used for determining the ratio of the scalar and the tensor coupling constants in the analysis of the beta decay of Rb-87, Cs-137, Tc-99, and Fe-59 in terms of a linear combination of the scalar and the tensor Fermi interactions.

- 3.3.5.39 ON THE CONVERSION SPECTRA OF THE ELEMENTS Ba-137 AND Ba-134. Julien Verhaeghe and Jean Demuynck. Compt. rend. 239, 1374-6 (1954) Nov. 22 (In French) ((No attempt was made to translate this article))

- 3.3.5.40 CONVERSION COEFFICIENT OF THE ISOMERIC GAMMA RAY IN THE DECAY OF BARIUM-137. A. H. Wapstra, Arkiv fur Fysik, 7, 3, 275-7, 1954

From beta spectrometer measurements on Cs-137, the K conversion coefficient of the 662 kev transition in Ba-137m was found to be 0.093

$\pm 0.006$  (M4 transition) and the K/L ratio  $5.8 \pm 0.3$ . The curie plot is straight down to 75 kev if a special correction factor is used.

Our active sample was prepared from the same carrier free Cs-137 used for determining the gamma energy and was spread with insulin on an  $0.15 \text{ mg/cm}^2$  aluminum backing.

- 3.3.5.41 INTERNAL CONSTANT EFFECT IN BARIUM-137.  
Larry Spruce and G. Goertzel (Physics Department,  
Washington Square College, New York University, New  
York, New York). Physical Review 93, 642, Feb. 1954

Continuous gamma rays accompanying internal conversion were recently detected for the first time. The process bears the same relationship to the Compton effect as internal pair production does to pair production, and will be referred to as the internal Compton effect. The ratio of the total number of gamma rays between 50 and 200 kev to the number of internally converted electrons was found to be in crude agreement with the ratio predicted by the only theory available, the semi-classical calculations of Wang Chang and Falkoff but the angular distribution differed from the theoretical prediction.

- 3.3.5.42 INTERNAL CONVERSION COEFFICIENT OF BARIUM-137. Toshio Azuma (Physics Department, Naniwa University, Sakai, Osaka, Japan). Journal of the Physical Society of Japan, 9, 1-3, Jan-Feb 1954

((The)) internal conversion coefficient of Ba-137 has been investigated with a double coil magnetic lens beta ray spectrometer. The relative intensities of the internal conversion electron of the 663 kev gamma ray for K and L, L and M, energy levels have been estimated as  $4.62 \pm 0.18$  and  $15.0 \pm 0.8$ , respectively. The value of K/L is in accord with the previous experimental values, and that of L/M is presented as a new value.

- 3.3.5.43 Refer to 3.1.5.18. FISSION YIELDS OF STABLE AND LONG-LIVED ISOTOPES OF CESIUM, RUBIDIUM, AND STRONTIUM IN NUCLEAR SHELL STRUCTURE. D. R. Wilgs, B. W. Smith, R. Horsley, and H. G. Thode. Canadian Journal of Physics, 31, 419, 1953

- 3.3.5.44 BETA SPECTRA OF Fe-59, Rb-87, Tc-99, Cs-137 AND THE COUPLING CONSTANTS OF SCALAR AND TENSER INTERACTIONS IN BETA DECAY. Masato Morita, Jun-Ichi, Fujita, and Masarni Yamada (Department of Physics Faculty of Science, University of Tokyo). Progress in Theoretical Physics, Vol. 10, No. 6, 630-40 December 1953

The beta spectra of Fe-59, Rb-87, Tc-99, and Cs-137 are investigated with the linear combination of scalar and tensor interactions in the Fermi-theory of beta decay, and it is inferred that the relative sign between two coupling constants of scalar and tensor interactions is minus.

- 3.3.5.45 THE MEASUREMENT AND INTERPRETATION OF THE K AUGER INTENSITIES OF Sr-113, Cs-137, and Au-198. C. D. Broyles, D. A. Thomas, and S. K. Haynes (Vanderbilt University, Nashville, Tennessee). Physical Review 89, 715-724 February 1953

Auger electron intensity measurements have been made on Cs-137, Au-198, and Sr-113, with a magnetic lens spectrometer which is briefly described. K auger yields for barium and mercury of  $0.130 \pm 0.007$  and  $0.056 \pm 0.008$ , respectively, were recorded. The K auger electrons were resolved into three groups. K-LL, K-LX, and K-XY, according as 2, 1, or 0, no L electrons were involved. The ratios of the intensities of the latter two to the K-LL were found to be for In,  $0.417 \pm 0.016$  and  $0.076 \pm 0.008$ , for ba,  $0.40 \pm 0.04$  and  $0.08 \pm 0.02$ . The measured ratio of K-LX to K-LL for mercury was  $0.71 \pm 0.15$ . The relationship of these results to theory is discussed. A summary of measurements of K fluorescence yield and K auger line intensity ratios is given. The auger spectrum from Au-198 indicates that K capture to Pt-198 cannot occur in more than 0.5% of the disintegrations. A crude estimate of the L to K capture ratio for Sr-113 indicates that it is not abnormally high.

- 3.3.5.46 A PRECISION MEASUREMENT OF THE CESIUM-137 GAMMA LINE. G. Lindstrom, K. Siegbahn, and A. H. Wapstra. (Nobel Institute for Physics, Stockholm, Sweden.) Proceedings of the Physical Society, (London) B66, 54-9, Jan. 1953

The recent establishment of a set of standard lines for precision work in beta ray spectroscopy is reviewed. In this work use was made of a semicircular spectrometer adjusted for high resolution. The proton method for measuring the magnetic field and a large double focusing spectrometer. In this way absolute measurements of the lines were made as well as a number of different relative measurements. The general consistency is very satisfactory indicating a precision of the standard lines of one or two parts in  $10^4$ . So far the following lines have been measured. The F, I, L, and K lines of Th(b, c''), the 411 kev line of Au-198, the annihilation radiation, the 2.75 Mev line of Na-21, the 662 kev line of Cs-137. In the present paper the measurements of the Cs-137 are described. The line has been studied as a conversional line and as a photo line emanating from a uranium converter. The combined results of the two spectrometers yield the following value of the energy of the Cs-137 line.  $E_{\text{gamma}} = 661.65 \pm 0.15$  kev.  $H_p$  value of the conversion line is  $3381.28 \pm 0.50$  gauss centimeters.

In order to get a uniform distribution of the cesium activity the insulin method of Langer (1949) was used. The activity was deposited on an aluminum foil with thickness  $0.2 \text{ mg/cm}^2$  on an area of  $4 \times 18 \text{ mm}^2$ . One may suspect a certain concentration of the activity at the edges of the rectangular area. A slit  $2 \text{ mm} \times 13 \text{ mm}$  placed on the aluminum foil masked off the dangerous regions. The same slit was used for the active Th source which is very uniform, being prepared in an activation vessel using a Rd Th sample.

3.3.5.47    PRECISION MEASUREMENTS OF NUCLEAR GAMMA  
RAY WAVELENGTHS OF Ir-192, Ta-182, RaTh, W-187,  
Cs-137, Au-198 AND ANNIHILATION RADIATION.  
David E. Muller, Harry C. Hoyt, David J. Klein, and  
Jesse W. M. DuMond (California Institute of Technology,  
Pasadena, California). Physical Review 88, 775-93,  
Nov. 1952

Measurements of the gamma rays from Cs-137, Au-198, and the annihilation radiation from Cu-64 were carried out after the calibration described above. The wavelength and energy of the single gamma ray following the decay of Cs-137 has been determined from the average of six measurements. The result in values are  $\lambda = 18.737 \pm 0.004$  milliangstroms;  $E = 661.60 \pm 0.14$  kev.



3. 3. 5. 48 NYO-869 STUDIES OF INTERNAL CONVERSION ELECTRONS FROM BARIUM-137. W. Kinney, B. Hildebrand. Progress Report AEC Contract by G. N. Glascoe, H. Landon, W. A. McKinley (Department of Physics, Rensselaer Polytechnic Institute, Troy, New York. Pg. 34, October 1952

(Under Table of Contents):

The L. Lamb, N. Reutherford, type field monitor now operates as an integral part of the beta ray spectrometer. A series of investigations on the internal conversion lines of Ba-137 was initiated with three principal purposes in mind.

1. Determination of which of the L. subshells is the principal contributor to the L conversion line and a similar study of the M line.
2. A measurement of the ratio  $\alpha_K/\alpha_L$ . The ratio of the K to L internal conversion coefficients.
3. A more precise determination of the Ba-137 gamma ray energy. The sources used in this study were two centimeters by one millimeter of area, density approximately 50 micrograms/cm<sup>2</sup> on aluminized backing of 100 micrograms/cm<sup>2</sup>.

3. 3. 5. 49 INTERNAL CONVERSION IN Pr-144, In-144, Ba-137, and Cd-110. W. C. Kelly, Physical Review 85, 101-3, Jan. 1952.

Beta ray spectrometer measurements have been made of the internal conversion ratio  $\alpha_K/\alpha_L$  for four nuclear transitions. Values obtained are  $5.3 \pm 0.1$  for the 132 kev transition of Pr-144;  $1.30 \pm 0.05$ , 192 kev, In-144;  $4.57 \pm 0.05$ , 662 kev Ba-137; and  $14 \pm 2$ , 656 kev, Cd-110, tentative assignments of multipolarity are given.

Three of the four radioactive materials were available at reasonably high specific activity. Cs-137 a parent of Ba-137 was obtained from Oak Ridge as CsCl at an activity of 1.05 millicuries/milliliter and with total solids not exceeding 2.4 mg/ml.

- 3.3.5.50 RADIOACTIVE DECAY OF Cs-137. M. A. Waggoner  
(Department of Physics, State University of Iowa, Iowa  
City, Iowa). Physical Review 82, 906-9, June 1951

The spins of the ground states of Cs-137 and Ba-137 are known to be  $7/2$  and  $3/2$ , respectively. The shape of the 518-kev beta-ray spectrum has been measured and found to correspond to the correction factor  $G' = (W_0 - W)^2 + A(W^2 - 1)$  in agreement with previous work. This is the shape predicted by the Fermi theory of beta-ray decay and the Gamow-Teller selection rules for a transition for which  $I = \pm 2$  and there is a parity change. The internal conversion coefficient of the 663-kev gamma-ray has been measured and found to be 0.097, which is in agreement with the theoretical value for magnetic  $2^4$ -pole radiation.

- 3.3.5.51 THE L X-RAY SPECTRA FROM RADIOACTIVITY DECAY OF TRANSURANIUM ELEMENTS. G. W. Barton, Jr., H. P. Robinson, and I. Perlman (Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California). Physical Review 81, 208-13, 1951 ((Also published as AECD-2494))

A bent crystal X-ray spectrometer is described and some results are given on the analysis of L series X-rays produced in radioactive decay processes of transuranium elements. There is generally good agreement between measured energy values of L series lines and those predicted by the Mosely relationship. The relative intensities of the various lines produced in this case from gamma ray internal conversion are compared with those from uranium excited by electron bombardment and values reported of internal conversion excited X-rays in the region of lead.

Plutonium X-rays from decay of Curium-242. The isotope Cm-242 is an alpha particle emitter with 162 days half life prepared for the present study by the neutron irradiation of the 475 year Am-241. The alpha decay of Cm-242 includes fine structure in which roughly 20% of disintegrations go to the excited state of Pu-238, about 50 kev above the ground state. The accompanying gamma ray transition is largely internally converted in the L shell and the X-rays measured in the study are those resulting from the refilling of these L orbit vacancies.

- 3.3.5.52 MEASUREMENT OF CESIUM-137, AND COBALT-60  
GAMMA RADIATION WITH A PRESSURE IONIZATION  
CHAMBER. G. R. Grove, Journal of Research,  
National Bureau of Standards, 48, 147-51, February 1951

The characteristics of the pressure ionization chamber at the National Bureau of Standards have been studied for the gamma-radiation from a Cs-137 source. (0.6614 Mev) and from a Co-60 source (1.1715 and 1.3316 Mev). The dosage rates produced in air by these sources were measured with this chamber with an accuracy of about 2%. Dosage rates obtained within extrapolation chamber and a Victoreen thimble chamber agreed with the values measured by the pressure chamber within the limits of the experimental errors.

- 3.3.5.53 INTERNAL CONVERSION OF GAMMA RAY FROM Cs-137.  
M. A. Waggoner (Department of Physics, State University  
of Iowa, Iowa City, Iowa). Physical Review 80, 489, 1950

The present work yields (1) a value of 0.625 Mev for the energy of the peak of the K internal conversion line, (2) a value of 0.518 Mev for the end point of the low energy beta group, and (3) a spectrum shape for the 0.518 Mev beta group which gives a straight Fermi plot when corrected by the correction factor  $G'$  and thus indicates that this transition involves a spin change of two with a change of parity. All of these results are in agreement with those of Osaba\*.

\*Phys. Rev. 76, 345, 1949

- 3.3.5.54 A PRECISE DETERMINATION OF THE ENERGY OF THE  
CESIUM-137 GAMMA RADIATION. Lawrence M. Langer  
and R. Douglas Moffat (Indiana University, Bloomington,  
Indiana). Physical Review 78, 74-75, 1950

In an attempt to establish Cs-137 as a calibration standard, the energy of the internal conversion electrons has been carefully determined by direct comparison with that of the 0.4112 Mev gamma ray of Au-198. A composite source of Au-198 and Cs-137 was prepared on a 0.00025 inch aluminum backing. The source was 0.25 centimeters wide and 2.5 centimeters high, and was spread with the aid of insulin. The source thickness was estimated to be 0.03 mg/cm<sup>2</sup> of Au-198 and 0.1 mg/cm<sup>2</sup> of Cs-137. ((The gamma energy in Mev for Cs-137 was found to be  $0.6614 \pm 0.0007$ ))

3.3.5.55 THE BETA SPECTRA OF Cs-137, Y-91, Pm-147, Ru-106, Sm-151, P-32, and Tm-170. Harold M. Agnew. Physical Review 77, 655-60, March 1950

The beta spectra of Cs-137, Y-91, Pm-147, Ru-106, Sm-151, P-32, and Tm-170 have been investigated using a large double magnetic lens spectrometer. The investigation of these materials was carried out with sources whose thicknesses were less than  $0.1 \text{ mg/cm}^2$ . The sources were mounted on thin foils in such a manner that they did not accumulate charge. Cs-137 and Y-91 were found to have distinctly non-linear Fermi plots. Ru-106, Pm-147 and Sm-151 have linear Fermi plots, and the Fermi plot of Tm-170 is slightly curved. Auger electron have been found associated with the Ba-137 decay product of Cs-137. The end points of the continuous beta ray spectra of those materials have been determined as follows: Low energy group of Cs-137,  $0.323 \pm 0.004 \text{ Mev}$ , -----, Pm-147,  $0.229 \pm 0.001 \text{ Mev}$ .

((Those parts of the abstract not dealing with Cs-137 or Pm-147 have been omitted.))

The sources were mounted on either zapon or LC600 films weighing less than  $0.01 \text{ mg/cm}^2$ .

3.3.5.56 THE RESPONSE OF THE ANTHRACENE SCINTILLATION COUNTER TO MONO-ENERGETIC ELECTRONS. John I. Hopkins (Oak Ridge National Laboratory, Oak Ridge, Tennessee). Physical Review 77, 406-7, Feb. 1950.

Approximately 50 microcuries of Cs-137 evaporated on a thin formvar film served as a source of electrons.

3.3.5.57 ATOMIC BEAM MAGNETIC RESONANCE EXPERIMENTS WITH RADIOACTIVE ELEMENTS Na-22, K-40, Cs-135 and Cs-137. Luther Davis, Jr., Darragh E. Nagle, and Jerrold R. Zacharias (Department of Physics and Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts). Physical Review 76, 1068-75, Oct. 1949

Modifications of the atomic beam magnetic resonance method for the determination of nuclear spins and moments by observation of the hyperfine structure of atomic ground states are described, which make it possible to work with very small quantities in samples with low

concentration. These modifications include the analysis of the beam by means of a mass spectrometer while performing the resonance experiment. A new source of beams of atomic alkali metals is described.

### 3.3.5.58 BETA SPECTRA OF FORBIDDEN TRANSITIONS.

Lawrence M. Langer and H. Clay Price, Jr. (Department of Physics, Indiana University, Bloomington, Indiana). Physical Review 76, 641-6, Sept. 1949

The beta spectra of the forbidden transitions Y-91, Sr-89, Y-90, Cs-137, RaE, Au-198, Re-186, and P-32 have been measured with thin sources and high resolution. Forbidden type spectra are found for the first five, Au-198, Re-186, and P-32 yield spectra with the allowed shape. Evidence is advanced for the reliability of the nuclear shell structure model and for the validity of GT selection rules. End points found are Y-91,  $1.537 \pm 0.007$  Mev; Sr-89,  $1.463 \pm 0.005$  Mev; Y-90,  $2.180 \pm 0.007$  Mev; Cs-137,  $0.51 \pm 0.01$  Mev; Au-198,  $0.965 \pm 0.005$  Mev; Re-186,  $1.063 \pm 0.006$ ; and P-32,  $1.689 \pm 0.01$  Mev.

The data on the Cs-137 distribution were obtained with several sources, the thinnest of which had an average thickness of  $0.03 \text{ mg/cm}^2$  and the thickest was  $0.2 \text{ mg/cm}^2$ .

### 3.3.5.59 THE FORBIDDEN TRANSITION OF YTTRIUM-91 AND CESIUM-137. J. Osoba (Department of Physics, Washington University, St. Louis, Missouri). Physical Review, Vol. 76, 345-9, August 1949

Yttrium-91 decays by the emission of a single beta particle with a maximum energy of 1.54 Mev, while Cs-137 decays in two ways. (1) Beta decay (maximum energy = 0.518 Mev), to Ba-137 followed by a gamma transition to the ground state and (2) Beta decay (maximum energy = 1.2 Mev) directly to the ground state. Probably not more than 5% of the Cs-137 nuclei decay directly to the ground state. Conventional (allowed transition) curie plots of Y-91 in the low energy group of beta particles of Cs-137 display curvature which is concave toward the energy axis at high energies in concave upward at low energies. It is assumed that the Gamow-Teller selection rules govern the beta process, the Fermi-function,  $F(Z, W)$ , for allowed spectra must be multiplied by a factor  $G = (W^2 - 1) + (W_0 - W)^2$  in a beta transition for which there is a change of parity, and for which the spin change is 2 units. If these conventional curie plots of Y-91 and

Cs-137 are modified by the factor  $G$ , the resulting plots are approximately straight lines. This indicates that the beta decay of Y-91 and the low energy beta decay of Cs-137 involve a spin change of 2 units and a parity change. These results contribute evidence for the validity of the Fermi theory of beta decay in the Gamow-Teller selection rules. The K internal conversion coefficient for the 0.663 Mev gamma from the decay of Cs-137 was found to be 0.081 while the ratio of the K conversion electrons to the L conversion electrons was 5.0. The gamma radiation seems to be either magnetic  $2^+$  pole or electric  $2^+$  pole.

Radioactive sources were prepared by first defining the source area with insulin on a thin Zapon film about  $0.03 \text{ mg/cm}^2$  thick. The active material in solution was then applied to this area and fast dried under an infrared lamp. ((The author feels it would be useless to give a figure for the source thickness since the active deposit seemed to be in small crystals.)) Previous work done at this laboratory indicated that Cs-137 decays by simple beta emission to a metastable state, Ba-137 which then decays (half life = 158 seconds) to the ground state of Ba-137 by the emission of a 0.663 Mev gamma ray. The present investigation confirms the result obtained by Mitchell and Peacock\* that the decay of cesium occurs in 2 ways ((as described in abstract)). As will be shown probably not more than 5% of the Cs-137 nuclei decay directly to the ground state.

\*Physical Review 75, 1272, 1949

3.3.5.60 DISINTEGRATION OF CESIUM-137. Charles L. Peacock and Alan C. G. Mitchell (Indiana University, Bloomington, Indiana). Physical Review 75, 1272-4, April 1949

Decay by emission of a single group of beta rays with an end point of 0.550 Mev to a metastable state of Ba-137 (156 seconds) has been reported for Cs-137 (33 years) from this state an internally converted gamma ray of energy 0.663 Mev is emitted. A simple decay scheme was suggested from these results. Mitchell and Peacock measured the ratio  $N_K/N_L$  for the internally converted electrons as well as the internal conversion coefficient  $\alpha$   $K_L$ . The measurement of these quantities as well as the known half-life for the metastable state indicate that the transition Ba-137 (metastable)  $\longrightarrow$  Ba-137 is electric  $2^+$  pole. They also pointed out certain difficulties with the proposed decay scheme. In the present experiment the beta ray spectrum of Cs-137 has been measured using  $180^\circ$  type magnetic

spectrometer of about 1% resolving power. The spectrum consists of 2 groups. The main group having an energy of 0.521 Mev and relative abundance 95% and a much weaker group having an energy of approximately 1.2 Mev and a relative abundance 5%.

The source used in this portion of the work had a surface density of not greater than  $0.1 \text{ mg/cm}^2$  mounted on Zapon of  $0.01 \text{ mg/cm}^2$ . A thicker source was used to investigate the higher energy beta rays since this group is only 5% abundant. The energy of the gamma ray causing the internal conversion electron has been measured and found to be  $0.669 \pm 0.005$  Mev. In addition, a line of very low intensity appears at 25 kev and can be ascribed to auger electrons arising as a result of an internally converted gamma ray. The gamma ray and the two beta ray groups appear to form a consistent energy level scheme.

3.3.5.61 ON THE Cs-137 DISINTEGRATION SCHEME. Alan C. G. Mitchell and Charles L. Peacock (Indiana University, Bloomington, Indiana). Physical Review 75, 197, Jan. 1949

The beta and gamma ray spectrum of Cs-137, 33 years, has been measured by Townsend, Owen, Cleland and Hughes\*, who find that the beta ray spectrum is simple with an end point at 0.550 Mev. In addition, there is only one gamma ray of energy of 0.663 Mev which is internally converted. Later the same authors showed that there were no coincidences between the beta and gamma rays and proved that the beta disintegration leads to a metastable state of Ba-137 which has a half life of  $158 \pm 5$  seconds. The gamma ray is emitted from this metastable state.

The K and L internal conversion lines of the 0.663 Mev gamma ray have been resolved, and the ratio  $N_K/N_L$  has been determined by several investigators. The best value for this ratio is  $4.8 \pm 0.3$ . In addition the internal conversion coefficient  $\alpha_K$  has been measured and is 11.8%. ((The balance of the article is concerned with spin in magnetic moments and is not reproduced here.))

\*Phys. Rev. 74, 499, 1948.

3.3.5.62 THE DISINTEGRATION OF Cs-137. J. Townsend, Marshall Cleland, and A. L. Hughes (Department of Physics, Washington University, St. Louis, Missouri). Physical Review 74, 499, Aug. 1948

In a previous letter\* it was reported that Cs-137 (33 years), has a simple beta spectrum and an end point energy of 0.550 Mev and a gamma ray of 0.663 Mev energy partially internally converted (12%). Subsequent coincidence counting experiments revealed no beta-gamma coincidences. However, coincidences were found between conversional electrons and the associated X-rays. A critical absorption experiment to determine the X-ray energy was performed by placing aqueous solutions containing Sn, Sb, Te, and I, between the source and one of the counters. The X-rays were found to be characteristic of barium indicating that the gamma rays are emitted from the excited barium nuclei which follow the beta decay of cesium. The long half life of the Cs-137 and excited Ba-137 nuclei indicated that large spin charges are involved in both beta and gamma transitions. The spin of the ground state of Ba-137 is known to be 3/2.

\*Phys. Rev. 74, 99-100, 1948

3.3.5.63 BETA AND GAMMA SPECTRA OF Cs-137. J. Townsend, G. E. Owen, Marshall Cleland, and A. L. Hughes (Washington University, St. Louis, Missouri). Physical Review 74, 99-100, July 1948

An investigation in this laboratory on the absorption of gamma rays called for the use of radioactive isotopes emitting monoenergetic gamma rays. Among those used was Cs-137, a fission product of high specific activity, produced by the Clinton Laboratories at Oak Ridge, Tennessee. ((The subject investigation showed that.)) The radiation from Cs-137 consists of a simple beta ray spectrum with an end point at 0.550 Mev energy and a single gamma ray of 0.663 Mev energy, 12% of the gamma ray being internally converted. The monoenergetic nature of the gamma ray combined with the long half life of Cs-137, 33 years, suggests its use as a gamma ray standard. Coincident studies are being made to secure information as to the decay scheme of this isotope.

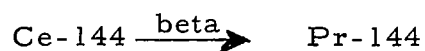


### 3.4 CERIUM-144, PRASEODYNIUM-144

#### 3.4.1 Nuclear Characteristics

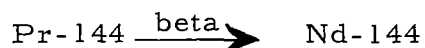
3.4.1.1	Atomic Number	Ce-144 58	Pr-144 59
3.4.1.2	Atomic Weight	144	144
3.4.1.3	Half Life	285 days Ref. 1	17 minutes Ref. 2

#### 3.4.1.4 Energy Levels and Decay Schemes



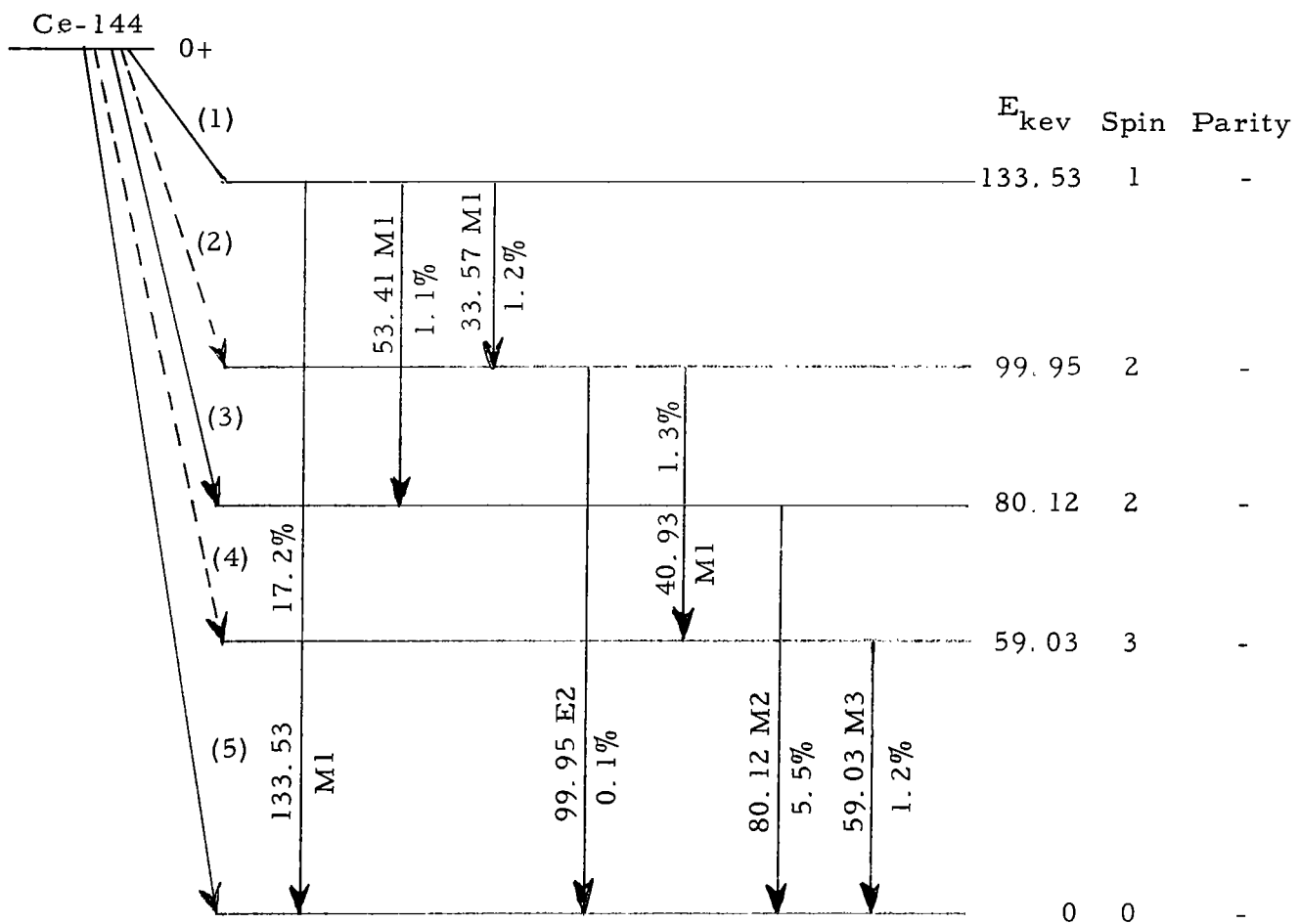
Beta	0.320 Mev (76 ± 2)%	
	0.240 Mev (4.5 ± 0.5)%	Ref. 3
	0.190 Mev (19.5 ± 1.0)%	Ref. 4

Gamma	133.53 Kev	
	99.95 Kev	
	80.12 Kev	Ref. 3
	59.03 Kev	
	53.41 Kev	Ref. 4
	40.95 Kev	
	33.57 Kev	

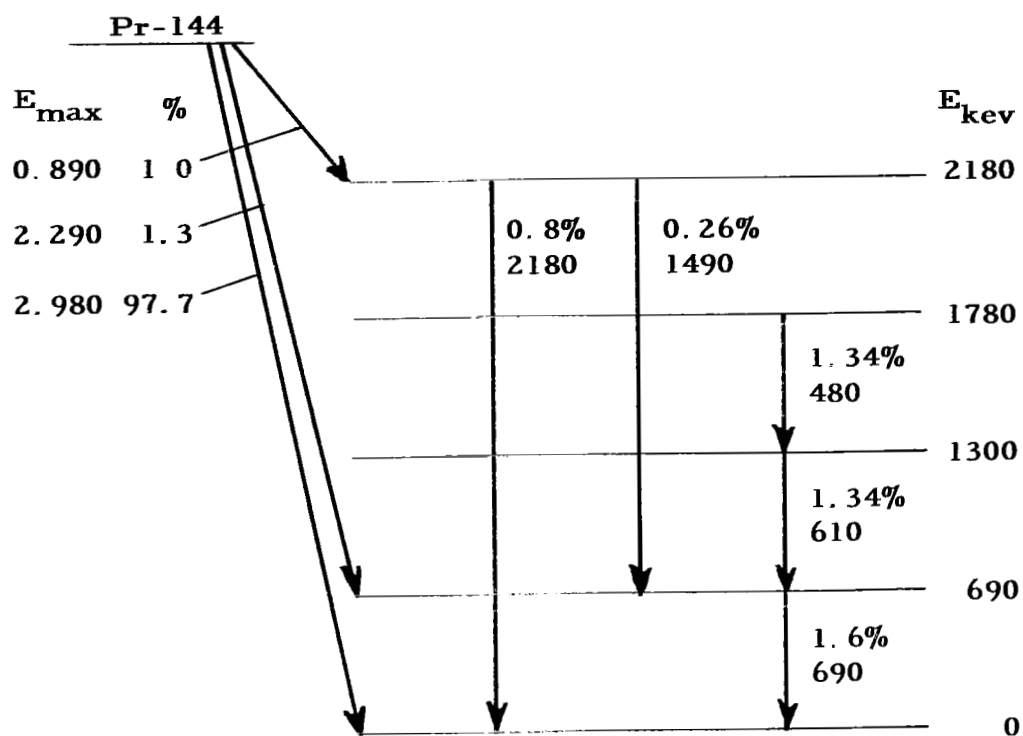


Beta	2.980 Mev (97.7)%	Ref. 4
	2.290 Mev (1.3)%	
	0.800 Mev (1.0)%	

Gamma	2.180 Kev	
	1.490 Kev	Ref. 4
	0.690 Kev	
	0.610 Kev	
	0.480 Kev	



	$E_{\beta\text{max}}$	%	$\log ft$
(1)	190	$19.5 \pm 1.0$	7.2
(2)	-	$< 0.3$	$> 9.2$
(3)	240	$4.5 \pm 0.5$	8.2
(4)	-	$< 0.4$	$> 9.2$
(5)	320	$76.0 \pm 2$	7.4



Reference 4

### 3. 4. 1. 5 Other Sources of Radiation

Ce-144 is in secular equilibrium with Pr-144 which decays with a 2.98 Mev beta particle. This results in the production of bremsstrahlung which although small in comparison to the gamma rays due to isotope decay, are still considered in shielding calculations. Table VIII presents these values. They were calculated by the shielding group at the Martin Company, and were published previously in reference 5.

TABLE VIII

#### Bremsstrahlung from Praseodymium-144

<u>Energy (Mev)</u>	<u>Abundance (%)</u>
0 - 0.15	2.26
0.15 - 0.45	0.57
0.45 - 0.75	0.24
0.75 - 1.05	0.12
1.05 - 1.35	$5.5 \times 10^{-2}$
1.35 - 1.65	$2.4 \times 10^{-2}$
1.65 - 1.95	$9.4 \times 10^{-3}$
1.95 - 2.25	$3.0 \times 10^{-3}$
2.25 - 2.55	$6.6 \times 10^{-5}$
2.55 - 2.85	$7.6 \times 10^{-5}$

Table IX presents gamma emission rates as determined by Oak Ridge to be used for shielding calculations. They include gammas due to decay of both cerium and praseodymium and bremsstrahlung produced by the latter (reference 6).

TABLE IX

#### Gamma Radiation Associated with Ce-144 - Pr-144

<u>Photon Energy (Mev)</u>	<u>Photons/watt-sec</u>
0.7	$7.484 \times 10^{10}$
1.5	$1.17 \times 10^{10}$
2.2	$3.74 \times 10^{10}$

TABLE IX (continued)

Photon Energy (Mev)	Photons/watt-sec
0.35	$8.204 \times 10^{11}$
0.95	$6.15 \times 10^{10}$
1.55	$9.644 \times 10^9$
2.15	$9.644 \times 10^8$
2.60	$2.00 \times 10^7$

### 3.4.2 Chemical Characteristics

#### 3.4.2.1 Fuel Forms

A summary of the physical characteristics of some of the more important cerium compounds are tabulated in Table X (reference 7). In general, all compounds with the exception of the halides and borides will oxidize readily at elevated temperatures. Cerium metal ignites in air at 150-180°C and burns in halogen vapors above 200°C.  $\text{Ce}_2\text{O}_3$  ignites in air at 200°C and goes to  $\text{CeO}_2$ . The carbide has been reported as being pyrophoric at room temperature. The halides exhibit the disadvantage of high vapor pressure ( $\text{CeF}_3$  boils at 2300°C) and melts too low for use in thermionic systems. It can be seen that the only choice of isotope fuel form is between the oxide and the borides. Since the oxide has both higher melting point and power density, it is considered the most desirable as the fuel compound for use in power conversion systems.

TABLE X

Specific Activity and Power Density for a Number  
of Cerium Source Compounds

Compound	Mol. Wt.	Wt. % Metal	Density, g/cm <sup>3</sup>	Melting at °C	Specific Power watts/g of compound	Power density watts/cm <sup>3</sup> of compound
Ce	140.12	100	6.7		3.48	23.3
$\text{Ce}_2\text{O}_3$	328.24	85.38	6.9	1690	2.97	20.5
$\text{CeO}_2$	172.12	81.41	7.3	2600	2.83	20.7
$\text{CeB}_6$	205.05	68.34	4.80	2190	2.38	11.4

TABLE X (continued)

Compound	Mol. Wt.	Wt. % Metal	Density, g/cm <sup>3</sup>	Melting at °C	Specific power watts/g of compound	Power density watts/cm <sup>3</sup> of compound
CeB <sub>4</sub>	183.41	76.41	5.74		2.66	15.3
Ce <sub>2</sub> C <sub>3</sub>	316.29	88.61	6.97		3.08	21.5
CeC <sub>2</sub>	164.15	85.37	5.56	1000	2.97	16.5
CeN	154.14	90.91	8.09	2000	3.16	25.6
β-CeSi <sub>2</sub>	196.31	71.38	5.45		2.48	13.5
CeP	171.11	81.89	5.56		2.85	15.8
CeS	172.20	81.38	5.94	2500	2.83	16.8
Ce <sub>2</sub> O <sub>2</sub> S	344.33	81.39	6.01		2.83	17.0
Ce <sub>3</sub> S <sub>4</sub>	548.65	76.62	5.67		2.67	15.1
Ce <sub>2</sub> S <sub>3</sub>	376.46	74.45	5.19		2.59	13.4
CeS <sub>2</sub>	204.26	68.60	5.07		2.39	12.1

#### 3.4.2.2 Material Compatibility

Ceric oxide is not chemically compatible with refractory metals at high temperatures. However, stainless steel and the super alloys show no attack by CeO<sub>2</sub> at temperatures in excess of 2000°F. The results of such compatibility studies were discussed in detail in Section 3.6 on plutonium.

In addition to chemical reaction with the oxide, the decay from cerium to praseodymium and finally to neodymium may result in valence changes, and some melting of intermediate products. A praseodymium dioxide corresponding to the ceric oxide is known although melting point data does not seem to be available. Since the half-life of Pr-144 is very short, the amount of this oxide in the fuel pellet at any time will be extremely small. The end product neodymium oxide, Nd<sub>2</sub>O<sub>3</sub>, melts at 2280°C (reference 8). However, the lower valence might result in some free oxygen in the system which will corrode the refractory metal containers. The use of a rhodium liner has been proposed to prevent contact between the cerium oxide fuel form and the refractory metals. This inert material should prevent any corrosive chemical reaction.

### 3. 4. 2. 3 Effects of Impurities on Nuclear Characteristics

The isotopic purity obtainable in a Ce-144 fuel compound is very low. It is given as 18% by reference 9. This is due primarily because of dilution by other stable isotopes of cerium. Thirty days after removal from the reactor, the composition of cerium isotopes produced by irradiation of U-235 is (reference 2)

Ce-140	35%	(stable)
Ce-141	5%	(33 day half life)
Ce-142	33%	(stable)
Ce-144	27%	(238 day half life)

It can be seen that the Ce-144 content is already quite small, and because of its fairly short half-life, its concentration with respect to the stable isotopes of cerium will continue to diminish at an appreciable rate.

The Ce-141 is readily decayed out and will not affect the nuclear characteristics of the aged Ce-144 compound. In fact no impurities are considered when determining the radiation levels from this isotope primarily because the abundant gammas generated by both cerium and praseodymium-144 overshadow any other possible contaminants.

### 3. 4. 3 Thermal and Physical Properties of Ce-144

The thermal and physical properties of Ce-144 and  $\text{Ce}_2\text{O}_3$  are found in Table XI.

TABLE XI

#### Physical Properties

		<u>Ce</u>	<u><math>\text{Ce}_2\text{O}_3</math></u>
Thermal Conductivity	$\frac{\text{cal cm}}{\text{sec } ^\circ\text{C cm}^2}$	0.0141(a) Ref. 10	0.0092(b) Ref. 11
Specific Power	$\frac{\text{watt}}{\text{gm}}$	26.0(c, d) Ref. 12	4.02(e)

TABLE XI (continued)

		<u>Ce</u>	<u>Ce<sub>2</sub>O<sub>3</sub></u>
Thermal Output	$\frac{\text{watts}}{\text{kilocurie}}$	8.15 (c) Ref. 13	8.15 (c) Ref. 13
Heat Capacity	$\frac{\text{cal}}{\text{gm } ^\circ\text{C}}$	0.048 Ref. 10	1.46 (c) Ref. 14
Heat of Fusion	$\frac{\text{cal}}{\text{mole}}$	105.0 (a) Ref. 10	--
Weight Density	$\frac{\text{gm}}{\text{cm}^3}$	6.9 Ref. 10	6.06 (f) Ref. 6
Melting Point	$^\circ\text{C}$	1690.0 Ref. 11	2600.0 (g) Ref. 15
Boiling Point	$^\circ\text{K}$	--	--
Specific Activity	$\frac{\text{dis/min}}{\text{microgram Ce-144}}$	$7.05 \times 10^9$ (c) Ref. 13	$7.05 \times 10^9$ (c) Ref. 13

KEY

- (a) This is the value for Te-127 (Tellurium) and has been used because of the similarities in the physical properties of Tellurium and Cerium.
- (b) Value at 495°C for CeO<sub>2</sub>. Other values are .0112 at 92°C, .0105 at 280°C, .0080 at 757°C, .005 at 2000°C, Ref. 13.
- (c) Calculated values.
- (d) This is the value for pure Ce-144.
- (e) The power density in watts/gm for Ce<sub>2</sub>O<sub>3</sub> with a purity of Ce-144 of 18%.
- (f) Effective density of compound.



- (g) Melting point of  $\text{CeO}_2$  but because of the strong physical similarities of the two compounds this value has been suggested.

The properties for the  $\text{CeO}_2$  fuel form were not included because of the strong physical and thermal similarities between the two fuel forms.

Because of the fairly short half life of Ce-144, its concentration with the stable isotopes of cerium will vary significantly, depending on the age of the material since formation in the fission process. The theoretical density of the fuel forms are difficult to achieve hence the power densities and other physical properties vary depending on the values used.

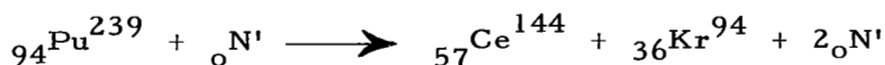
As a result of the above mentioned problems and because the values found in the literature are not always adequately defined a consistent set of values for the specific power, power density and specific activity were calculated using the following assumptions:

1. The Cerium and Praseodymium are in equilibrium, i. e. , for every disintegration of Ce-144 there will be a disintegration of Pr-144.
2. The other isotopes of cerium are sufficiently "aged" so that they do not contribute.
3. The source purity of Ce-144 is 18%.
4. The density of the  $\text{Ce}_2\text{O}_3$  fuel form is  $6.06 \text{ gm/cm}^3$ .

#### 3.4.4 Isotope Production, Availability and Cost

##### 3.4.4.1 Production Method

Cerium is produced in quantity by fission of heavy elements in nuclear reactors. A typical fission reaction is:



At the time of fuel recovery the Ce-144; together with other cerium isotopes, is separated from the spent fuel elements. Cerium is separated from the gross fission product mixtures with the other rare earth elements. The cerium fraction can be separated from the other rare earths either by ion exchange and solvent extraction or selective precipitation, all processes being carried out at closely controlled ph.

The cerium fraction will contain the isotopes 140, 141, 142 and 144. The 140 and 143 isotopes are inert and contribute nothing to the heat output of the isotope. Ce-141 has a half-life of 33 days and will decay rapidly. It is thus desirable to hold the isotope for some time before processing to permit the majority of this short lived isotope to decay away. The Xe-144 isotope has a half-life of about one second, so that all Ce-144 resulting from its decay can be considered as appearing instantaneously upon fission.

#### 3.4.4.2 Availability and Cost

The planned production capacity for Ce-144 as shown below was taken from published data from Division of Isotope Development, USAEC.

This data originally published in 1963 has been altered to reflect the latest estimates from DID.

	FISCAL YEAR							
	<u>1964</u>	<u>1965</u>	<u>1966</u>	<u>1967</u>	<u>1968</u>	<u>1969</u>	<u>1970</u>	<u>1980</u>
Mc	3.5	3.5	3.5	3.5	50	100	100	100
Kw(t)	27.7	27.7	27.7	27.7	350	790	790	790

Private power reactor production is expected to increase the cerium availability considerably above those shown in the Table above. In 1970 the total quantity of Ce-144 is expected to be 1330 Kw(t) and in 1980 this will rise to 4200 Kw(t) (Reference 16).

Present costs of Ce-144 are \$0.50/curie but are expected to decrease to as low as \$0.04/curie.

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3. Beta-Gamma Coincidence Counting for the Calibration of Nuclides with Complex Decay Schemes, Int. Journal Applied Rad. and Isotopes, 12-13, 137-42, March 1962.
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3. 4. 5      Annotated Bibliography

3. 4. 5. 1      Refer to 3. 1. 5. 4.    LARGE SOURCES OF BETA RADIATION FOR CHEMICAL PROCESSING.    Isotopes Radiation Technology, 2: 158-69 (Winter 1964-65).
3. 4. 5. 2      ORNL-P-447.    DETERMINATION OF NUCLIDE CONCENTRATIONS IN SOLUTIONS CONTAINING LOW LEVELS OF RADIOACTIVITY BY LEAST-SQUARES RESOLUTION OF THE GAMMA-RAY SPECTRA..    E. Schonfeld, A. H. Kibbey, and W. Davis, Jr. (Oak Ridge National Laboratory, Tennessee). (Sept. 3, 1964). Contract W-7405-eng-26.    42p (CONF-721-2)

From 8th Conference on Analytical Chemistry in Nuclear Technology, Gatlinburg, Tennessee, Oct. 1964.

((Ce-144, among other isotopes, was used to demonstrate the capability of gamma ray spectrometers to determine the radionuclide content in solutions.    No properties of Ce-144 are discussed.))

3. 4. 5. 3      Refer to 3. 10. 12.    RSG-A-1.    PRELIMINARY REPORT ON PUBLIC HEALTH HAZARD ANALYSES OF SPACE ISOTOPIC POWER SOURCES.    J. J. Fitzgerald, Gordon L. Brownell, and N. Irving Sax - Jack J. Gabay, ed. (New York, State Department of Health, Albany).    Mar. 1, 1964.    Contract SAPH 76262.    87 p.
3. 4. 5. 4      IN-TIME INTEGRATED FISSION RATE STUDIES IN URANIUM USING ALPHA OR GAMMA SPECTROMETRY.    A. Guillon, S. May, S. Sapena, and R. Sauvagnac.    Bull. Inform.Sci.Tech. (Paris), No. 81, 35-7 (Mar. 1964). (In French)

Some methods for measuring burnup of fissile material are reviewed. Alpha spectrometry after irradiation (measurement of U-235 disappearing), which is accurate only for high integrated fluxes, is discussed. Measurements of the ratio of fission products to uranium are also considered. Some examples are given for irradiations made under different conditions. The fission products studied were Cs-137, Ce-144, and Ba-140. A chemical separation is described for each of these radionuclides.

- 3.4.5.5 KFK-199. BESTIMMUNG DER SPEZIFISCHEN AKTIVITÄT EINIGER RADIONUKLIDE. (Determination of the Specific Activity of a Radionuclide). H. Ruf and H. Muenzel (Kernforschungszentrum, Institut für Radiochemie, Karlsruhe, Germany). Feb. 1964. 45p.

A method is described for determining the yield and specific activity of the fission products Ce-144, Sr-89, Sr-90, Tc-99, Zr-95, Nb-95, Ru-106, Ru-103, I-131, and Cs-137. The yield determination is based on the method of gravimetric analysis with the use of paper filters. The conditions for the necessary weight constant of the paper filters were worked out. Determination of the absolute decay rate in the measurement preparation depended on the determination of the operation stages of the counter by means of activation, which was standardized by the 4- $\pi$  method.

- 3.4.5.6 Refer to 3.1.5.3. A RADIOMETRIC METHOD FOR THE ISOTOPIC ANALYSIS OF MIXTURES OF Ce-141 AND Ce-144, Sr-89 AND Sr-90 IN SAMPLES OF LOW TOTAL ACTIVITY. S. V. Belov and N. V. Sidorov. Zh. Analit. Khim., 19: 1049-52 (1964). (In Russian)

- 3.4.5.7 DECAY OF Ce-144. Takeki Iwashita, Takashi Inamura, Yoshio Ikemoto and Seizaburo Kageyama (Department of Physics, Tokyo University of Education, Tokyo, Japan). Journal of the Physical Society of Japan, Vol. 18, No. 10, Oct. 1963.

Level structure of Pr-144 was investigated by gamma-gamma coincidence measurements and the spins of the levels were determined using the gamma-gamma directional correlation method. The 166 keV level proposed by Freeman has been confirmed. A new 92 keV gamma ray coincidence with the 41 keV gamma ray has been found. A decay scheme containing the new level is proposed. Interpretations of the levels are discussed in terms of the shell model.

The Ce-144 sample used in the present work was obtained from the Oak Ridge National Laboratory in the form of  $\text{CeCl}_3$  in HCl solution. A portion of this liquid containing about 0.02 milligrams of Ce-144 was placed in a plastic holder and was set at the center of a goniometer 6 centimeters apart from the face of each scintillator.

3. 4. 5. 8      SPIN OF THE Pr-144 GROUND STATE.    BETA DECAY MEASUREMENTS ON Ce-144 and Pr-144.    E. Creutz, L. A. Deraedt, J. P. Deutsch, L. Grenacs, D. Siddique. (University of Louvain, Nuclear Physics Center, Heverle - Louvain, Belgium.)    Physics Letters 6, 329-31, Oct. 1, 1963

Carrier free Ce-144 was used as source material in Cerium-chloride form, and the counting rate in the beta channel was carefully kept low to avoid spurious coincidences.    A 400 channel RIDL analyzer was incorporated in the layout to display the coincidence beta spectrum. ((The conclusion drawn by the authors of this article is that it is possible to assign uniquely the spin value 0 to the ground state of Pr-144. ))

3. 4. 5. 9      THE SPINS OF Pr-144 FROM BETA-GAMMA CORRELATION MEASUREMENTS.    W. Collin, H. Daniel, S. Margulies, O. Mehling, P. Schmidlin, H. Schmitt, and K. S. Subudhi. (Max Planck Institute for Physics, Heidelberg, Germany).    Physics Letters 5329-31, August 1, 1963.

((This article is devoted to an experiment and discussion of the  $0^- \longrightarrow 0^+$  transition of Pr-144. ))

3. 4. 5. 10     EVALUATION OF ISOTOPE THERMIONIC GENERATORS.    Robert J. Harvey, Thomas C. Robinson, Joseph J. Fitzgerald, and Gordon Brownell.    Transactions of the American Nuclear Society,    6, 88-91, June 1963.

This article is quite general in nature.    It describes power output of various candidate fuel elements and analysis mission times and cost per thermowatt for polonium-210, Cm-242, Ce-144, Cm-244, and Pu-238.

3. 4. 5. 11     EXPECTED ALPHA-DECAY DATA OF THE RARE EARTH NUCLIDES ON THE BASIS OF DIFFERENT SYSTEMATICS.    T. Fenyés (Inst. of Nuclear Research, Hungarian Academy of Sciences, Debrecen) and Z. Boedy. At. Kozlemen., 5: Suppl. to No. 3-4 (Paper 6) 17 p. (1963)

The alpha-decay data of rare earth nuclides are summarized.    The presently existing experimentally measured alpha- and beta-decay energies allow the computation of the decay energy of some 30 further

nuclides. By plotting the alpha-decay energy vs. atomic number, the expected alpha decay energy of nuclides, the alpha-activity of which has not yet been detected experimentally, was estimated. The estimation of the alpha-decay energies for the above nuclides on the basis of the available mass data as well as p and n separation energies was also performed. The expected alpha-partial half-lives were also estimated from the gained alpha-decay energies using a semi-empirical relation expressing explicitly the dependence of the half-life on the atomic number. Finally, some questions about the possibility of experimental detection of the alpha-activities not yet measured are discussed.

3. 4. 5. 12 BETA-SPECTRA OF Ce-144. Masatosi Fujishiro and Tosio Azuma. Ann. Rept. Radiation Center, Osaka Prefect., 4: 86-9 (1963)

The beta spectra of Ce-144 were studied. The results obtained were five beta groups of  $320 \pm 5$ ,  $260 \pm 4$ ,  $240 \pm 4$ ,  $185 \pm 4$ , and  $130 \pm 4$  kev. The experimental arrangement used is described.

3. 4. 5. 13 A PARAMETRIC STUDY OF DIRECT NUCLEAR ELECTROGENERATOR CELLS USING A BETA EMITTING SOURCE. Allan J. Cohen and Charles A. Low, Jr. (Lewis Research Center, Cleveland). New York, American Institute of Aeronautics and Astronautics, 1963, Preprint 63048B, 15p. (CONF-10-57)

From Electric Propulsion Conference, Colorado Springs, Colorado, March 1963.

A design analysis is completed for direct nuclear electrogenerator cells using the beta emitting radioisotope Ce-144. The physical phenomena that complicate the design are the beta particle energy spectrum, the energy losses in the fuel and support foil, and the relativistic motion of the particle in the electric field. In addition to these physical restrictions, the various cell parameters such as radius ratio, fuel layer thickness, support foil thickness, and collector thickness are interrelated in such a complex manner that the IBM 7090 digital computer is required to find the characteristics of the cells. Cylindrical and spherical geometries are studied to determine the minimum weight per unit power. The main details of the mathematical operations performed by the computer are outlined along with information pertinent to the input requirements and results



of the program. The simple calculations necessary to complete the description of the specific cells are indicated. The cell characteristics that result from these procedures are the operating voltage, current and power per unit area, the temperatures of the emitter and collector, and the weight per unit power. From these characteristics, the cell parameters that yield the optimum feasible design can be logically determined by simple analytical methods, which are described. As a result of these studies, a few designs are presented that can have weight/power ratios of 0.3 to 0.5 kg/kw. The study shows that within reasonable construction and physical limits a simple, efficient, and lightweight direct nuclear electrogenerator can be designed for space power uses.

3. 4. 5. 14 A SCINTILLATION SPECTROMETER FOR HIGH ENERGY BETA DECAYS. J. G. Cramer, Jr., B. J. Farmer, and C. M. Class. (Rice University, Houston, Texas) Nuclear Instruments and Methods. 16, 289-300, August 1962

A scintillation spectrometer has been developed for application to beta decays having end point energies of 3 Mev or more. The spectrometer uses a large plastic scintillator on which the betas are incident from an external source. The beam of betas is restricted to a narrow cone to insure that virtually the total energy is absorbed within the scintillator. Spectra are recorded with a multi-channel pulse height analyzer. The pulse height scale is calibrated in terms of energy by means of the end points of known beta spectra in the edges of Compton Recoil electron distribution from gamma rays. The relationship between pulse height and energy is found to be linear up to 13 Mev. End point energies of spectra can be assigned to an accuracy of 1-2% on the basis of the calibration. The response of the spectrometer to monoenergetic electrons for the energies below 3 Mev is found to be essentially Gaussian. Spectra of the beta decays from Ce-144 - Pr-144, Al-28 F-20 and B-12 compare favorably with those obtained by magnetic analysis.

((This article describes a scintillation spectrometer whose purpose is to measure high energy beta rays. Ce-144 is used as an illustrative example.))

3. 4. 5. 15 STUDY OF CERIUM-144 DECAY SCHEME BY COINCIDENCE METHOD. N. V. Forafontov, V. S. Shpinel and Ts B. Vasilev. (Nuclear Research Institute, Moscow State University, USSR) Nuclear Physics 35, 260-72, June 1962

The Ce-144 decay is studied by means of a two lens beta spectrometer and a luminescent beta (gamma) spectrometer. A decay scheme containing a new level, 147 kev, is constructed on the basis of beta-e<sup>-</sup>, e<sup>-</sup>-e<sup>-</sup> and gamma - e<sup>-</sup> coincidence measurements.

3. 4. 5. 16 APPLICATION OF COINCIDENCE METHOD IN STUDIES OF Ce-144 DECAY SCHEME. Ts. B. Vasilev, N. V. Forafontov, and V. S. Shpinel. Vestn. Mosk. Univ., Ser. III, No. 3, 3-16 (May-June 1962) (In Russian)

The Ce-144 decay scheme was studied using a beta spectrometer and a beta (gamma) luminescence spectrometer in coincidence. The results obtained for beta-e, e-e, and e-gamma coincidences are in agreement with decay schemes found in published literature. An additional level at 147 kev was introduced into the scheme.

3. 4. 5. 17 ORNL-TM-198. ISOTOPES DIVISION QUARTERLY REPORT, OCTOBER-DECEMBER 1961. J. H. Gillette (Oak Ridge National Laboratory, Tennessee). June 5, 1962. Contract W-7405-eng-26. 39p.

Various source preparations and the preparation of cesium borosilicate glass are discussed. ((Several other topics were discussed in this report. However, they have no bearing on the subject of this document.))

3. 4. 5. 18 4 PI BETA GAMMA COINCIDENCE COUNTING FOR THE CALIBRATION OF NUCLIDES WITH COMPLEX DECAY SCHEMES. H. Houtermans and M. Miguel (International Atomic Energy Agency, Vienna, Austria) International Journal of Applied Radiation and Isotopes 12-13, 137-42, March 1962

The 4 pi beta gamma coincidence calibration is normally used only for nuclides with decay schemes which are both reasonably well known and fairly simple. In this paper it is shown that by a systematic variation of the mean beta efficiency  $a_m$  (the ratio of the coincidence to the gamma counts), the error caused by the self-absorption of the beta rays can (in 4 beta-pi counting) be eliminated by an extrapolation method. This is possible even for very complex and uncertain decay schemes, provided that at least one beta gamma cascade is known. The results of a calibration of a solution of Ce-144 plus Pr-144 are reported as an example.

An extrapolation method is described ((in this article)) for a calibration of a Ce-144 plus Pr-144 solution. Figure 6 shows the rather complex disintegration schemes for these nuclides. For the measurements a flat pill box type 4 pi flow counter was used for the beta rays and a 1-3/4 x 2" NaI crystal for the gamma rays. The sources were the residue from drops dried on a plastic film (about 15 ug/cm<sup>2</sup>) with gold evaporated on both sides (15 micrograms/<sup>2</sup> cm each). Different beta self-absorption was obtained by the variation of the source area as determined by the surface covered by the drop of wetting agent or by the variation of the contents of inactive carrier. Two different conditions of pulse height discrimination were used for the gamma rays.

3. 4. 5. 19 NP-11622. THE TRANSVERSE POLARIZATION OF INTERNAL CONVERSION ELECTRONS EMITTED FOLLOWING BETA DECAY (thesis). Technical Report No. 35. J. Bernard Blake (Illinois University, Urbana). March 1962. Contract ONR 1834 (05). 46p.

The transverse polarization of conversion electrons following beta decay was measured for eight different decays. Decays were studied for the following isotopes: Hg-203, Pa-233, Au-198, Au-199, Ce-141, Ce-144, Tm-170, and I-131. Electron polarization was measured by Mott scattering. Results are presented in tables. Corrections made to the raw data are described.

((A portion of this thesis was published as Government Report NP-11737 in 1960. It carried the same title.))

3. 4. 5. 20 CONSIDERATION OF SELF-ABSORPTION AND SELF-SCATTERING MEASURING THE ABSOLUTE ACTIVITY OF THICK LAYER SPECIMENS. G. B. Belov, V. M. Erokhin, L. M. Anokhina and Yu. V. Solov'ev. Priroda i Tekhnika Eksperimenta 56-62, Sept-Oct 1961.

The results obtained in determining  $u = f(E_{\max})$  that have been published in the literature are generalized. Dependence given by  $u = 0.01523 E_{\max}^{-1.514}$  have been derived. A nomograph for determining the correction for self-absorption is proposed. Self-absorption curves for a 4 pi solid angle of the Y-90 and Y-91 radioisotopes were plotted. Calibration curves for determining the corrections for self-absorption and self-scattering in measuring the activities of the Sr-89, Sr-90 and Y-90, Y-91, Ce-141, and Ce-144 —→ Pr-144 radioisotopes by means of a MST-17 end type counter were plotted.

3. 4. 5. 21 NUMBER OF PHOTONS PER DECAY OF Ce-144 and Eu-155.  
A. N. Silant'ev and B. S. Kuznetsov. Izvestia Akad Nauk.  
25, 1186-7, Sept. 1961

The number of photons per decay of Ce-144 and Eu-155 were measured using a 4 pi beta gamma coincidence procedure. The Ce and Eu were purified chromatographically. In order to check the purity of the specimens additional measurements were carried out after the passage of four months. The results of the repeat measurements agreed with those of the initial ones. The following numbers of Ce-144 were determined. Energy of the gamma in kev 36, 80, 134. Number of photons per cent, respectively, 8.2, 1.6, 9.7.

3. 4. 5. 22 REACTOR FUEL PROCESSING. Technical Progress  
Review, Vol. 4, No. 3. Stephen Lawroski, ed. (Argonne  
National Laboratories, Illinois). July 1961. 67p.

Information is given regarding commercial aspects of fuel processing, including data on the production and properties of Po-210, Cm-242, Pu-238, Ce-144, Pm-147, Cs-137, and Sr-90 radionuclide heat sources. A critical incident at Idaho Chemical Processing Plant is described. The chemical explosion characteristics of TBP-uranyl nitrate-nitric acid systems are investigated. The preparation of fuels for processing is studied, with particular attention to chemical dejacketing and dissolution procedures. Improvements are described in solvent extraction, ion exchange, volatility, fused salt, and pyrometallurgical fuel processing methods. Means for controlling corrosion in these processes are discussed. Waste disposal systems and operations are described, including low-level liquid waste handling, reduction to solids, final disposal, and separation of specific isotopes. The production and recovery of U and Th and their compounds are described. One hundred and seventy-eight references.

3. 4. 5. 23 THE DECAY OF CERIUM-144. J. N. Geiger, R. L.  
Graham and G. T. Ewan. (Atomic Energy of Canada, Ltd.,  
Chalk River, Canada.) Nuclear Physics 28, 387-406, 1961.

The results of conversion electron gamma and gamma-gamma coincidence studies of the Ce-144 decay are presented in this paper. The gamma ray spectra studied include those coincident with the  $L_1$  33.57,  $L_2$  40.93, K 80.12,  $L_2$  80.12, K 133.53 and  $L_1$  133.53 kev conversion lines with gamma 80.12, gamma 133.53 and the Pr K, X-rays. A detailed search of the conversion electron spectrum for the

K line of a 66 kev transition is reported. This search gave a negative result. The intensity of such a line is less than 5% of that of the  $L_1$  33.57 conversion line. These new experiments provide both qualitative and quantitative support for the Ce-144 disintegration scheme previously reported by us, which was deduced from a high resolution investigation of the Ce-144 internal conversion electron spectrum. The results do not support the various modifications to this scheme which have been recently proposed by four experimental groups. The conversion electron gamma coincidence results limit the intensity of a beta feed to the 99.95 kev level in Pr-144 to less than 0.3% for disintegration.

The Chalk River precision ray beta spectrometer was used for both the conversion line study and the conversion electron gamma coincidence investigation reported in this paper. The source used for this investigation was the weaker of those prepared and used in our earlier studies and is two years old. The spectrometer was operated at instrument momentum resolution settings in the range of 0.1 to 0.3% in the present work. A cylindrical side window proportional counter operating with a continuous flow methane filling was used as the beta detector.

- 3.4.5.24 IMPROVING THE ACCURACY OF DOSIMETRIC CHARACTERISTICS OF Ce-144, Pr-144, and Kr-85.  
V. V. Ovechkin and D. V. Ovechkin. Sb. Rabot po Nekotorym Voprosam Dozimetrii i Radiometrii Ionizir. Izluchenii, No. 2, 50-1 (1961)

Results are given of calculation of gamma constants for Ce-144, Pr-144, and Kr-85, made on the basis of more precise experimental data on the corresponding gamma-quanta yields. The gamma constant was determined by the formula:  $K_{\text{gamma}} = 1.54 \times 10^5 \sum h\nu_i \cdot \text{gamma}_i \cdot n_i$  r/hr-mc, where  $h\nu_i$  is the gamma-quantum energy in Mev;  $\text{gamma}_i$  is the coefficient of true gamma absorption in air in  $\text{cm}^{-1}$ ;  $n_i$  is the yield of gamma quanta of a given energy per decay. The accuracy of determination of gamma-quanta yield of Ce-144 and Pr-144 with energies of 80, 134, 700, 1490, and 2180 kev was improved by using a face-end beta-counter and a scintillation gamma spectrometer. (Ref. Zh., Fiz. 1963)

- 3.4.5.25 PHYSICAL BASIS FOR DOSIMETRY OF BETA RAYS.  
K. K. Aglintzev and V. P. Kassatkin, p.1375-80 of "IXth International Congress of Radiology, Vol. 2." B. Rajewsky, ed. Stuttgart, Georg Thieme Verlag, 1961. (In German)

For determination of the mean beta dose, the number of beta particles and their energy distribution must be known. The effective electron spectra of S-35, W-185, Tl-204, Y-91, Sr-90 + Y-90, P-32, and Ce-144 + Pr-144 at various depths of a tissue-equivalent substance were measured and graphed. The contribution to the dose of electrons of various energies in the field of S-35, Tl-204, and Y-91 sources was determined. The dose dependence of a beta particle on the maximum energy of the beta spectrum was measured. The results permit the determination of dose magnitudes at various depths of the substance irradiated with a fixed number of beta particles of a given isotope.

((The original text of this article appeared in Med. Radiol. 5 #1, 52-8 Jan. 1960 and was published in Russian.))

3. 4. 5. 26 (PR-P-48) PHYSICS DIVISION PROGRESS REPORT FOR OCTOBER 1, 1960-DECEMBER 31, 1960. (Atomic Energy of Canada Ltd., Chalk River Project, Chalk River, Ontario). 80p. (AECL-1196)

The validity of the Ce-144 decay scheme deduced from conversion line studies was confirmed. Preliminary experiments were carried out to assess the feasibility of precise gamma-energy measurements using the external conversion process. ((This is a partial abstract. Several other topics that are not concerned with this report are discussed.))

3. 4. 5. 27 (MND-P-3009-1) SNAP PROGRAMS. Quarterly Progress Report No. 1 (for) October 22-December 31, 1959. (Martin Co., Nuclear Div., Baltimore). Nov. 1960. Contract AT(30-3)-217. 287p.

Work was done on the development of low-power thermionic generators which included prototype development, heat-transfer studies and tests, vacuum tests in prototype shells, cathode and sapphire creep tests, a parametric study, the development of molybdenum fuel capsules, heliarc welding of molybdenum fuel capsules, hazards studies on Ce-144 thermionic units, fabrication of a containment cask for Ce-144 units, and fueling molybdenum capsules with Ce-144 pellets.

3. 4. 5. 28 (PR-P-47) PHYSICS DIVISION PROGRESS REPORT, JULY 1, 1960 TO SEPTEMBER 30, 1960. (Atomic Energy of Canada, Ltd., Chalk River Project, Chalk River, Ontario.) 75p. (AECL-1142)

Conversion line spectra were examined for a large number of isotopes in order to see whether the K-line widths could be predicted.

Relative E2 transition probabilities from the 2+ and 3+ gamma-vibrational levels in Gd-154, Gd-156, and Dy-160 are discussed. The results of studies of the conversion line spectra of Ce-143, Ce-144, and Ba-140 are given.

3. 4. 5. 29    BALANCED ION CHAMBER MEASUREMENT OF HALF LIVES OF SEVERAL RADIOISOTOPES.    H. T. Easterday and R. L. Smith (University of Oregon, Eugene, Oregon). Nuclear Physics 20, 155-8, 1960

Half lives of six gamma emitters have been measured using a balanced ion chamber arrangement with one chamber driven by a radium source. Data analyzed by least squares yielded the following results ((among others, Cs-134,  $2.05 \pm 0.05$  years, Ce-144,  $277 \pm 4$  days)). ((This article is primarily a description of the balanced ion chamber and its use. As a proof of its capability several half lives of isotopes were determined, among these were Ce-144 and Cs-134.))

3. 4. 5. 30    INVESTIGATION OF BETA RADIATION OF Nb-95 and Ce-144 BY THE METHOD OF ABSORPTION IN AIR. N. E. Tsvetaeva and L. A. Rozenfeld, Journal of Experimental and Theoretical Physics USSR, 38, 641-642, Feb. 1960

The authors of this article investigated the beta radiation of Ce-144 by the method of absorption in air. By means of absorption in an aluminum foil, the beta radiation of Pr-144 was eliminated. Using this, the absorption in aluminum for Pr-144 to zero thickness of absorber was extrapolated in accordance with the variation of the absorption curve for Ru-106 - Rh-106.

3. 4. 5. 31    THE DECAY OF Ce-144.    M. J. Freeman (Bedford College, London). Proceedings of the Physical Society of London. 74, 449-56. Oct. 1959

The beta gamma and internal conversion spectra of Ce-144 have been studied with a magnetic spectrometer, scintillation counter, in coincidence techniques. The internal conversion coefficients of the 80 kev and 133 kev gamma rays were measured. Four partial beta spectra were observed and new evidence has been found for a level at 166 kev in the Pr-144 nucleus. No evidence was found for the 95 kev and 145 kev gamma rays in the decay.

A 5 millimeter diameter disc source was prepared by vacuum distillation of cerium-oxide onto a  $250 \text{ ug/cm}^2$  aluminum backing strengthened by a  $50 \text{ ug/cm}^2$  nylon film. Figure 6, The Proposed Decay Scheme of Ce-144, is presented on page 25A of the lab notebook.

3. 4. 5. 32    CONCERNING A 100-KEV TRANSITION IN THE SPECTRUM OF Ce-144.    A. V. Gnedich, L. N. Kryukova, and V. V. Murav'eva (Institute of Nuclear Physics, Moscow State University).    J. Exptl. Theoret. Phys. (USSR) 36, 329, Jan. 1959

Although the radiation from Ce-144 has been investigated by many authors there is still no established Ce-144, Pr-144 decay scheme. We investigated the conversion spectrum of Ce-144 with the aid of a spiral beta spectrometer having a resolution of 0.25 per cent. ((A diagram shown in this article gives the 57.7 kev conversion line.)) The shape of this line and the fact that its half widths (0.63%) is greater than the single lines of this spectrum make it obvious that this is a complex line. ((Graphical resolution of this line yielded two components with energies 57.76 and 57.45 kev. Also resolved was the  $l_i - 100$  line with  $E_e = 92.83 \text{ kev}$ .)

Absorption in air at less than atmospheric pressure was also carried out in accordance with the course of the absorption curve in air for Ru-106 - Rh-106. As can be seen the value of the air coefficient of Ce-144 = to 3.35 differs greatly from the assumption that only a single beta radiation component with energy about 0.3 Mev exists in Ce-144 as stated by several authors.

3. 4. 5. 33    DECAY OF Ce-144 AND Pr-144.    R. J. Hickok, W. A. McKinley (Rensselaer Polytechnic Institute, Troy, New York).    Physical Review 109, 113-16, January 1, 1958

The beta and gamma spectra from the decay of Ce-144 and Pr-144 have been investigated by means of a magnetic lens beta spectrometer and single channel scintillation spectrometer. Three beta spectra with end point energies in relative intensities of 184 kev (30%), 243 kev (3%), and 320 kev (63%) were observed for Ce-144. Gamma rays and internal conversion lines corresponding to 54, 80, and 134 kev transitions were also detected. Internal conversion coefficients indicate an M1 multipolarity for the 80 and 134 kev transitions. Pr-144 yields three partial beta spectra with end point energies and relative intensities of 0.90 Mev (2%), 2.45 Mev (3%) and 3.15 Mev (95%).



Corresponding gamma rays were also detected. Beta gamma and gamma-gamma coincidence measurements substantiate the proposed decay scheme.

3. 4. 5. 34    GAMMA-GAMMA ANGULAR CORRELATION AND INTENSITY MEASUREMENTS ON LOW ENERGY GAMMA RAYS IN THE DECAY OF Ce-144.    W. Zuk and S. Gustafsson

The decay of Ce-144 has been investigated using a gamma scintillation spectrometer and an automatic gamma-gamma directional correlation apparatus. The relative intensities of the following gamma rays of Pr-144, 134, and 80 kev, and the 36 kev X-ray have been measured giving the following result -  $100:33.4 \pm 1.0:107.0 \pm 2$ . The result of the 80 kev gamma ray is in agreement with Hickok, et al, and in disagreement with Geiger, et al. A measurement has been done of the gamma-gamma angular correlation of the 54-80 kev cascade in Pr-144. Our measurement confirms the (1-) spin assignment of Geiger, et al, for the 134 kev level. In Nd-144 has been measured the angular correlation of the 1.49 - 0.69 Mev cascade. The result is in very good agreement with the previous measurement by Steffan.

Only very much diluted liquid sources of  $\text{CeCl}_3$  in HCl have been used. The cylindrical lucite source container has an inner diameter of approximately 2 millimeters and a wall thickness of 0.8 millimeters. The source height range from 12 to 16 millimeters and the source strength was generally a few microcuries.

3. 4. 5. 35    LEVEL SCHEME OF Pr-144.    A. K. Sengupta, R. Bhattacharyya, J. Lahiri and P. N. Mukherjee (Saha Institute of Nuclear Research, Calcutta, India).

The level scheme of Pr-144 following beta decay of Ce-144 has been studied with a Siegbahn-Slatis beta ray spectrometer and scintillation spectrometers. Three beta groups have been observed with end point energies and relative intensities as 184 kev (26%), 240 kev (8%), and 312 kev (65%). Corresponding gamma rays of energies 53 kev, 81 kev, and 134 kev, have also been detected. Internal conversion lines of these transitions, along with those of some other gamma lines of energies 33 kev, 38 kev, 42 kev, 59 kev, and 95 kev have been observed. R/L ratio and half time measurement of the 134 kev gamma line indicates an M1 multipolarity for this transition. Coincidence studies have also been made and a tentative decay scheme is proposed.

The source was obtained carrier free in the form of cerium-chloride in HCl solution from the Radio Chemical Center, Amersham. Sources were prepared by evaporation of the solution on a thin mylar foil ( $0.5 \text{ mg/cm}^2$ ). The beta spectrum was taken with a Siegbahn-Slatis beta ray spectrometer with a momentum resolution of 1.9% at a transmission of 5%. The detector was a GM counter with a window of mylar foil which transmitted electrons upon an energy of about 20 kev. A proposed Ce-144 decay scheme is presented as Figure 5 in this article.

3.5 PROMETHIUM-147

3.5.1 Nuclear Characteristics

3.5.1.1 Atomic Number 61

3.5.1.2 Atomic Weight 147

3.5.1.3 Half Life

2.620 ± 0.005 years

Reference 1

3.5.1.4 Energy Levels and Decay Scheme

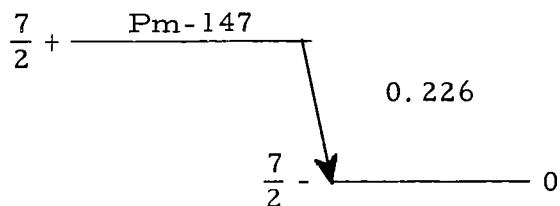
Pm-147  $\xrightarrow{\text{beta}}$  Sm-147

Beta 0.226 Mev (100%)

Reference 2

No Gamma

Reference 3



Reference 4

### 3.5.1.5 Other Sources of Radiation

There is only a small quantity of gamma radiation associated with the decay of Pm-147, and the bremsstrahlung energy will be less than the 0.226 Mev beta emitted during the decay. Thus, if pure Pm-147 could be obtained, practically no shielding would be required since the structural materials and self-absorption in the source would probably be sufficient to reduce dose rates to acceptable levels. However, small amounts of Pm-146, Pm-148, and Pm-148m are also formed during the fissioning process, and these cannot be separated. The amount of Pm-146 formed is small, and the effective energy of the gamma radiation released during its disintegration is about the same as that of Po-210. Pm-148 and Pm-148m on the other hand, contain many more gamma rays and would require heavy shielding if freshly prepared Pm-147 were used. Fortunately, Pm-148 and Pm-148m have short half lives (42 days and 5 days) compared to Pm-147 and the effect of these contaminants disappears after two to three years. The half life of Pm-147 is long enough to allow storage for the period of about 3 years without too great a loss of promethium. Table XII presents the decay data of these isotopes (Reference 5).

Table XII (Ref. 3)

#### Promethium Decay Data

Nuclide	Half-life	Beta Radiation		Gamma Radiation	
		Max. E. Mev	Abundance %	E. Mev.	Abundance %
Pm-147	2.62 years	0.225	100	-	-
Pm-146	4.4 years*	0.78	35	0.45	65
				0.75	65
Pm-148	42 days	0.47	40	0.10	8
		0.57	15	0.19	5
		0.64	11	0.29	12
		0.76	19	0.41	16
		1.07	5	0.43	11
		1.20	3	0.55	92
		2.00	1	0.63	90
		2.50	6	0.73	36
				0.91	19
				1.01	20
				1.46	2

\*Nuclear Physics 48, 555-60, 1963

### 3.5.2 Chemical Characteristics

#### 3.5.2.1 Fuel Forms

A summary of the melting points and specific powers for various promethium compounds are listed in Table XIII (Refs. 6, 7). To date, only the oxide has been seriously considered for use as a heat source in power conversion systems.

Table XIII (Refs. 6, 7)

#### Promethium Compounds

Compound	Melting Point (°C)	Specific Power (w/gm)
Pm metal	1080	0.333
Pm <sub>2</sub> O <sub>3</sub>	2270	0.286
PmF <sub>3</sub>	1340	0.240
PmCl <sub>3</sub>	1010	0.193
PmBr <sub>3</sub>	950	0.127
PmI <sub>3</sub>	1070	0.093

#### 3.5.2.2 Material Compatibility

Studies have been conducted at the Battelle-Northwest Laboratories in Richland, Washington on the compatibility of promethium oxide with various container materials (Ref. 6). Due to a lack of sufficient quantities of promethium, the studies were conducted with the oxides of samarium and neodymium which have atomic numbers both one above and below that of promethium.

Table XIV presents an analysis of the results of those tests.

Table XIV

	<u>Molten Metal</u>	<u>Solid Metal</u>	<u>Oxide</u>
Time, Days	23	30	42
Temp, °C	1100	600	1100
Tantalum	excellent	excellent	excellent
Molybdenum	-	-	-
Zirconium	-	excellent	-
Titanium	fair	good	poor
Platinum	poor	-	good
Hastelloy C	poor	fair	good
Hastelloy N	poor	-	good
Hastelloy X	poor	-	good
Haynes-25	poor	-	fair
Stainless Steel 310	poor	fair	-
Inco-600	poor	poor	-
Rene-41	poor	poor to fair	-
Carpenter-20	poor	-	-
TZM	-	excellent	-
Naval Brass	-	fair	-

Excellent - No attack observed  
 Good - less than 1 mil penetration  
 Fair - 1-10 mil penetration  
 Poor - greater than 10 mil penetration

### 3.5.2.3 Effects of Impurities on Nuclear Characteristics

The chemical purity of Pm-147 is given as 99% by reference 8, the principal impurity being the samarium daughter which grows in at a rate of 2.2% per month. The most probable radioactive impurities in the Pm-147 product are Eu-155 (a fission product), Eu-152 and 154 which are produced by neutron activation of fission-produced stable europium isotopes (Ref. 9), and Am-241. Radiochemical purity of 1000 d/m total alpha and  $10^{-11}$  curies of Eu-154 per curie of Pm-147 was achieved in the production of 13,000 curies of Pm-147 (Ref. 10).

The only major contaminant that affects the nuclear characteristics of promethium are the isotopes Pm-146 and Pm-148m. Traces of these two gamma emitters in the Pm-147 fuel compound require the use of considerably more shielding than would be necessary if pure Pm-147 could be produced.

### 3. 5. 3 Thermal and Physical Properties of Pm-147

The thermal and physical properties of Pm-147 are found in Table XV. The thermal and physical properties of Pm<sub>2</sub>O<sub>3</sub> are also presented.

Table XV

#### Thermal and Physical Properties of Pm and Pm<sub>2</sub>O<sub>3</sub>

		<u>Pm</u>	<u>Pm<sub>2</sub>O<sub>3</sub></u>
Thermal Conductivity	$\frac{\text{cal cm}}{\text{sec } ^\circ\text{C cm}^2}$	0.031(a) Ref. 11	(b) Ref. 12
Specific Power	$\frac{\text{watts}}{\text{gram}}$	0.3330 Ref. 6	0.286 Ref. 6
Thermal Output	$\frac{\text{watts}}{\text{K curie}}$	0.35(c) Ref. 13	0.35(c) Ref. 13
Heat Capacity	$\frac{\text{cal}}{\text{gm } ^\circ\text{C}}$	(d) Ref. 14	(e) Ref. 15
Heat of Fusion	$\frac{\text{cal}}{\text{mole}}$	180 Ref. 16	-
Weight Density	$\frac{\text{gm}}{\text{cm}^3}$	7.3 Ref. 16	7.3 Ref. 16
Melting Point	$^\circ\text{C}$	1080 Ref. 16	2270 Ref. 16
Boiling Point	$^\circ\text{C}$	3300 Ref. 16	-
Specific Activity	$\frac{\text{K curies}}{\text{gm Pm-147}}$	0.928(c)	0.928(c)

(a) At 27°C

(b)  $0.0045 + (1.414 \times 10^{-8})(1020 - T)^2$  for samarium,  
T = °K.

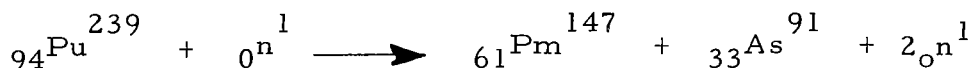
- (c) Calculated using 2.67 years as half life, 0.059 Mev/disintegration as the average energy per disintegration, a purity of 95% and no other radioactive elements present, and a density of 6.72 gm/cm<sup>3</sup>.
- (d)  $(4.0 + 0.00925 T - 2.9 \times 10^{-9} T^3)$  for promethium  
T = °K.
- (e)  $(28.99 + 0.00576 T - 415,900/T^2)$  for neodymium  
T = °K.

The Pm-147 is relatively pure with respect to its radioactivity since only about 1% Pm-148 (half life of 42 days) will be present four months after discharge from a reactor. With a cooling time of two years, the Pm-146 (half life of 4.4 years) content will be  $2.5 \times 10^{-5}\%$  of the Pm-147 activity (Ref. 6). The theoretical density of Pm<sub>2</sub>O<sub>3</sub> is 7.3 g/cc. Practical production density is about 7.0 g/cm<sup>3</sup>, or 95% of theoretical maximum.

### 3.5.4 Isotope Production, Availability and Cost

#### 3.5.4.1 Production Method

Pm-147 is produced by the fission of heavy elements in nuclear reactors. A typical fission reaction is:



Pm-147 is chemically separated from spent fuel elements. After separation of the gross fission product mixture from the solution containing the dissolved fuel elements, the promethium isotopes are separated out by standard chemical techniques.

#### 3.5.4.2 Availability and Cost

The following table presents the prospective production level of Pm-147 as published by the Division of Isotope Development, USAEC, in January of 1963 and brought up to date with recent personnel contacts:



# FISCAL YEAR

	<u>1964</u>	<u>1965</u>	<u>1966</u>	<u>1967</u>	<u>1968</u>	<u>1969</u>	<u>1970</u>	<u>1980</u>
Mc	0.5	0.5	0.5	0.5	15	30	30	30
Kw(t)	0.18	0.18	0.18	0.18	5.5	11	11	11

The above values, which are based primarily upon government production facilities, can be expected to be increased considerably during the 1970-1980 time period through private power production. In 1970 the quantity of promethium is expected to be  $25 \frac{\text{Kw(t)}}{\text{year}}$  and by 1980 this quantity will rise to  $111 \frac{\text{Kw(t)}}{\text{year}}$  (Ref. 17).

The cost of promethium compares favorably with other fission product isotopes. Present costs are approximately \$0.20 curie and are expected to decrease in 1968 (Ref. 7).

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7. Personal communication with H. H. Van Tuyl 21 September 1965.
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### 3. 5. 5      Annotated Bibliography

3. 5. 5. 1      Refer to 3. 3. 5. 1. (ORNL-3802) REVIEW OF RADIO-ISOTOPES PROGRAM, 1964. J. H. Gillette - Ingrid B. Lane, comp. (Oak Ridge National Laboratory, Tennessee). May 1965. Contract W-7465-eng-26. 71p.
3. 5. 5. 2      CALORIMETRIC DETERMINATION OF THE MEAN BETA-ENERGY AND HALF-LIFE OF PROMETHIUM-147. E. J. Wheelwright, D. M. Fleming, and F. P. Roberts (General Electric Co., Richland, Washington). J. Phys. Chem., 69: 1220-3 (April 1965)

The mean beta energy of the Pm-147 decay was calorimetrically measured at periodic intervals extending over a 1-year period. Two independently prepared 1000-C sources of highly purified  $\text{Pm}_2\text{O}_3$  were examined. The results obtained were self-consistent and in excellent agreement with measurements made on the same sources by an independent scientist using a second calorimeter. Values of  $0.3330 \pm 0.0005$  w/g of Pm-147 and  $2.620 \pm 0.005$  years for the beta-decay energy and the half life, respectively, are believed to be very reliable values.

((This article was published in almost identical form as government report #HW-SA-3698, May 20, 1964))

3. 5. 5. 3      THERMAL POWER OF PROMETHIUM-147. J. C. Posey, R. S. Pressly, and J. H. Gillette (Oak Ridge National Laboratory, Tennessee). Nucl. Sci. Eng., 21: 403-6 (March 1965). (ORNL-P-61)

The thermal power of Pm-147 was determined to be  $0.414 \pm 0.005$  w/kC, using calorimetry and  $4\pi$  beta proportional counting. This value corresponds to an average beta energy of  $0.070 \pm 0.001$  Mev.

3. 5. 5. 4      BNWC-45. PROMETHIUM ISOTOPIC POWER DATA SHEETS. H. H. Van Tuyl (Chemistry Department, Battelle, Northwest, Richland, Washington). 15 March 1964

The Division of Isotopes Development of the AEC has assigned to the Pacific Northwest Laboratories the responsibility for developing technology for preparing and using promethium as a radioisotopic fuel

in space, marine, and terrestrial application. Technology has been developed (and demonstrated on a  $2 \times 10^4$  curie scale) to permit large scale preparation of highly purified Pm. The half life and specific heat generation rate were measured. Shielding requirements for Pm have been studied, and the existence of Pm-146 in production and power fuels was discovered and its consequences investigated. Source preparation and encapsulation are being investigated, along with studies on compatibility with container materials and re-entry considerations.

((This report goes into detail on Pm nuclear properties, production technology, container materials compatibility, and physical and chemical properties. Much of the information presented in the preceding Pm-147 section was taken directly from this document.))

- 3.5.5.5 (BMI-1709) FABRICATION STUDIES ON Pm-147 BETA-VOLTAIC BATTERIES. Russell H. Barnes, Robert L. Ritzman, John F. Kircher, and Duane N. Sunderman (Battelle Memorial Inst., Columbus, Ohio). January 25, 1965. Contract W-7405-eng-92. 43p.

Techniques for preparing betavoltaic batteries consisting of a thin layer of Pm-147 in the form of hydrated  $\text{Pm}_2\text{O}_3$  sandwiched between two standard n-on-p silicon solar cells and encapsulated in a 13/16-in-diameter by 7/8-in-long Type 304 stainless steel case were studied. The Pm-147 was purified by ion exchange, using alpha-hydroxy isobutyric acid as the elution agent. Low-intensity light sources were used to select the solar cells. Batteries containing 2, 4, and 8 curies of Pm-147 were prepared. Power outputs from the 2-curie units averaged about 3 uw, with the maximum output for any one battery being 5.7 uw. The 4-curie units averaged 8 uw, with a maximum of 9.3 uw for one battery. The single 8-curie battery prepared produced 10.7 uw. It was concluded from the study that major increases in battery performance should come from use of solar cells designed for low-power operation with a Pm-147 beta source.

- 3.5.5.6 Refer to 3.1.5.4. LARGE SOURCES OF BETA RADIATION FOR CHEMICAL PROCESSING. Isotopes Radiation Technol., 2: 158-69 (Winter 1964-65)

- 3.5.5.7 (HW-81746 (p. 2, 40-1) THE SPECIFIC POWER AND THE HALF-LIFE OF Pm-147. D. M. Fleming (General Electric Co., Hanford Atomic Products Operation, Richland, Washington)

The specific power of Pm-147 was found to be  $0.330 \pm 0.0007$  w/g. A preliminary value for its half life is 2.68 year.

- 3.5.5.8 MEASUREMENT OF MOTT ASYMMETRY AT 100 kev, OF LONGITUDINAL ELECTRON POLARIZATION OF Co-60 AND Pm-147 FROM 40 to 210 kev AND OF A LARGE SOURCE DEPOLARIZATION UNEXPECTED BY THEORY. V. Eckardt, A. Ladage, and U. V. Moellendorff (Universitat, Hamburg). Phys. Letters, 13: 53-5 (Nov. 1, 1964)

The Mott asymmetry function  $S(\theta, Z, E)$  can be measured by the double scattering of beta particles with known polarization. For electron energies in the range of 100 kev there are discrepancies between theoretical calculations and experimental results. The polarization of electrons from Co-60 and Pm-147 sources were measured with an energy resolution of  $\pm 2.5$  kev in the range from 40 to 210 kev. A double focusing permanent bending magnet transmits only electrons with energies of  $100 \pm 2.5$  kev. The method has the advantage that the scattering at the gold target occurs for all accepted beta energies at the same energy of 100 kev.

- 3.5.5.9 THE EXTERNAL BREMSSTRAHLUNG PRODUCED IN THIN FOILS OF Pm-147. Vincent C. Truscello, Joseph Silverman. Paper presented at the Symposium of Low Energy X and Gamma Sources and Applications. October 20, 21 1964, Chicago, Illinois. Sponsored by the Atomic Energy Commission.

A NaI(Tl) scintillation spectrometer was used in conjunction with a 512-channel pulse height analyzer to study the spectral distribution of the external bremsstrahlung produced by Pm-147 betas ( $E_{\max} = 223$  kev) in thin foil targets.

A thin source of Pm-147 (mass thickness of  $0.3 \text{ mg/cm}^2$ ) was prepared by depositing 23 microcuries of this radionuclide on a sheet of aluminized mylar ( $0.6 \text{ mg/cm}^2$ ). The bremsstrahlung was produced by thin foil targets ranging in thickness from 1 to  $70 \text{ mg/cm}^2$  and in atomic numbers from 13 to 73. The bremsstrahlung was found to have a linear dependence on the atomic number of the target material as predicted by theory. It was further determined that the experimental data could be represented by the empirical relations:

Photon yield =  $3.7 \times 10^{-5}$  Z photons/beta particle  
Photon intensity =  $1.8 \times 10^{-6}$  Z Mev/beta particle

- 3.5.5.10 (MLM-1204) MOUND LABORATORY PROGRESS REPORT FOR JUNE 1964. J. F. Eichelberger, G. R. Grove, and L. V. Jones (Mound Laboratory, Miamisburg, Ohio). Sept. 18, 1964. Contract AT(33-1)-Gen-53. 23p.

The half-life of promethium-147, which was prepared at the Hanford Laboratories, was  $2.6244 \pm 0.006$  years with an estimated absolute error limit of 0.0006 years. The power and average beta energy for promethium were determined to be 0.33267 watts per gram and 60.54 kev, respectively.

- 3.5.5.11 (DPSPU-64-30-20) DETERMINATION OF PROMETHIUM-147 IN ENVIRONMENTAL SAMPLES. R. W. Taylor (Du Pont de Nemours (E.I.) & Co. Savannah River Plant, Aiken, S.C.). Sept. 1964. Contract AT(07-2)-1. 10p. (CONF-727-3)

From 10th Annual Bioassay and Analytical Chemistry Meeting, Cincinnati, Oct. 1964.

A procedure employing a combination of simple analytical techniques was developed to determine Pm-147 in environmental samples. Lanthanides were initially precipitated from solution as hydroxides. Rare earths that interfere with beta counting of Pm-147 and other contaminants were removed by ion exchange, solvent extraction, and precipitation. Pm-147 was finally co-precipitated with neodymium oxalate, fired to the oxide, and counted in a gas-flow proportional counter. Recovery was  $71\% \pm 9\%$  (relative standard deviation). Decontamination factors for fission products usually found in environmental samples were 10,000.

- 3.5.5.12 THE MEASUREMENT OF AQUEOUS SOLUTIONS OF METALLIC RADIOISOTOPES BY LIQUID SCINTILLATION METHODS. A. Dyer, J. M. Fawcett, and D. U. Potts (Royal Coll. of Advanced Tech., Salford, Lancaster, England). Intern. J. Appl. Radiation Isotopes, 15: 377-80 June 1964

Determination of Ag-110m, Na-22, Pm-147, and Tl-204 in aqueous solutions by scintillation methods is described.

3. 5. 5. 13 ISOTOPIC POWER DATA SHEETS. S. J. Rimshaw (Oak Ridge National Laboratory, Oak Ridge, Tennessee). For presentation at the Industry Information Meeting on Isotopic Power Development and Applications, Washington, D. C., May 18-19, 1964

Source Material	$\text{Pm}_2\text{O}_3$
Half-Life	$\text{Pm-147} \rightarrow 2.67$ years
Decay and Radiation Properties	<div> <div> <math>\text{Pm-147}</math> </div> <div> <math>\text{Sm-147}</math> (stable) </div> </div> <div> <div>Beta</div> <div>Gamma</div> </div> <div> <div>0.225 Mev (100%)</div> <div>none</div> </div>
Isotopic Composition	100% $\text{Pm-147}$
Radiochemical Purity	About 1% $\text{Pm-148}$ ( $T_{1/2} = 42$ days) will be present 4 months after discharge from a reactor. With a cooling time of two years, the $\text{Pm-146}$ ( $T_{1/2} = 1.94$ years) content will be $5 \times 10^{-5}\%$ of the $\text{Pm-147}$ activity, and the $\text{Pm-148}$ activity will be less than the $\text{Pm-146}$ activity.
Chemical Purity	95% $\text{Pm-147}$ principal impurity is Nd at last separation time. Purity can approach 99%, but will decrease as stable $\text{Sm-147}$ daughter grows in.
Specific Power	0.41 watts/kilocurie of $\text{Pm-147}$ or 0.324 watt/gram of pure $\text{Pm}_2\text{O}_3$ .
Thermal Energy	2440 curies per thermal watt.
Density	Theoretical density is 7.3. Practical production density is 90% of the theoretical density, or $6.6 \text{ g/cm}^{-3}$ .
Power Density	$2.03 \text{ watts/cm}^{-3}$ based on 95% $\text{Pm}_2\text{O}_3$ and a density of $6.6 \text{ g/cm}^{-3}$ .
Thermal Conductivity	Values for typical rare earth oxides are about $0.006 \text{ cal/sec} \cdot \text{cm} \cdot ^\circ\text{C}$ at $150^\circ\text{C}$



Specific Heat	0.0944 cal/g from 0° to 1000°C for Sm <sub>2</sub> O <sub>3</sub> .
Coefficient of Expansion	$10.8 \times 10^{-6}/^{\circ}\text{C}$ from 30° to 740°C for Sm <sub>2</sub> O <sub>3</sub> .
Melting Point	2350°C for Sm <sub>2</sub> O <sub>3</sub> .
Mechanical Strength	For Sm <sub>2</sub> O <sub>3</sub> : Modulus of Rupture = 2000 lb/in <sup>2</sup> at room temperature. Modulus of elasticity (sonic) = $26.5 \times 10^{-6}$ lb/in <sup>2</sup> at room temperature.
Thermal and Radiation Stability	Fabricated sources have exhibited good stability for several years.
Radiation Attenuation	Shielding required is small for Pm-147. Shielding requirements are established by the Pm-146 and Pm-148 content. Refer to "Shielding Requirements for Promethium Sources," HW-77375, and "Radiation Characteristics and Shielding Requirements of Isotopic Power Sources for Space Missions," ORNL-TM-591 (Rev.). Also, refer to "Handbook of Shielding Requirements and Radiation Characteristics of Isotopic Power Sources for Terrestrial, Marine and Space Applications," ORNL-3576.
Gas Evolution Due to Radioactive Decay Processes	None
Leach Rate	No data available. Most fired rare earth oxides hydrate and disperse in boiling water.
Vapor Pressure	No data available.
Resistance to Thermal Shock	No data available.
Burnup Characteristics	Dispersibility poor.
Capsule Compatibility	Excellent with most metals.

- 3.5.5.14 ON THE MEAN BETA DISINTEGRATION ENERGIES OF Pr-143, S-35, AND Pm-147. Vaeinoe Hovi, Lasse Niemelae, and Jorma Varonen (Univ. of Turku, Finland). Ann. Acad. Sci. Fennicae, Ser. A VI, 150: 1-12 (1964).

By using an isothermal microcalorimetric method and radioactive beta sources, the mean beta disintegration energies of Pr-143 and S-35 were measured at liquid nitrogen temperature. The activity of these sources was known with a great accuracy. The measured values of the mean beta disintegration energy, 310 and 48.8 keV for Pr-143 and S-35, respectively, are in very good agreement with those obtained from Fermi's theory and from experimental beta spectra. The mean beta disintegration energies of S-35 and Pm-147 were also determined by means of a scintillation spectrometer in which an anthracene split-crystal was used.

- 3.5.5.15 Refer to 3.10.12. (RSG-A-1) PRELIMINARY REPORT ON PUBLIC HEALTH HAZARD ANALYSES OF SPACE ISOTOPIC POWER SOURCES. J. J. Fitzgerald, Gordon L. Brownell, and N. Irving Sax - Jack J. Gabay, ed (New York. State Dept. of Health, Albany). Mar. 1, 1964. Contract SAPH 76262. 87p.

- 3.5.5.16 CONSTRUCTION OF A PROMETHIUM-147 ATOMIC BATTERY. H. Flicker, J. J. Loferski and T. S. Elleman, IEEE Transactions on Electron Devices, January 1964

The feasibility of constructing an atomic battery by combining the beta emitting radioisotope Pm-147 and a semiconductor containing a PN junction has been studied, both theoretically and experimentally. Properties of cells made by diffusing a P type impurity into an N type GaAs P impurity into N type silicon (n/p) are compared. The problems associated with the presence of an alpha emitting impurity Am-241 in the Pm-147 and with the fabrication of a suitable Pm-147 source are considered. Experiments with prototype batteries composed of Pm-147 and N/P silicon cells have lead to the conclusion that such batteries would have a half life of 1.6 years, somewhat shorter than that of Pm-147 (2.6 years). Such a battery could generate a few milliwatts of power from a volume of about 2 cubic inches including shielding. ((Table 1, properties of Pm-147 is presented as follows:))

$E_{\max}$  =  $230 \times 10^3$  ev, max energy of beta particles

$E_{\beta \text{ av}}$  =  $73 \times 10^3$  ev, average energy of beta particles

$t_H$  = 2.6 years, half life

G = 980 curies/gm, specific activity

$I_{\max}$  =  $1.1 \times 10^{-8}$  amps/cm<sup>2</sup>, self absorption limited electron current which would be emitted from a surface of a semi-infinite layer.

$ZE_{\beta \text{ av}}I_{\max}$  =  $1.7 \times 10^{-3}$  watts/cm<sup>2</sup>, maximum power emitted from both faces of a thick planar source.

$\alpha_o$  =  $1300 \text{ cm}^{-1}$ , self linear absorption coefficient of Pm-147 beta rays.

$\alpha_o/\rho$  =  $150 \text{ cm}^2/\text{gm}$ , mass absorption coefficient of Pm-147 beta rays.

3.5.5.17 CEA-2355. ETUDE DU SCHEMA DE DESINTEGRATION ET DES MOMENTS ANGULAIRES DES NIVEAU EXCITED DU Pm-147 PAR DES MESURES DE SPECTROGRAPHIE ET DE CORRELATIONS ANGULAIRES (these). (Study of the Disintegration Process and of the Angular Moments of The Excited Levels of Pm-147 Using Spectrographic and Angular Correlation Measurements (thesis). Claude Philis France. (Commissariat a l'Energie Atomique. Centre d'Etudes Nucleaires, Saclay). 1963, 87p. (In French)

The energies and relative intensities of fifteen gamma lines were determined by a direct spectrographic study. A previously unreported level at  $77 \pm 2$  kev was identified. A coincidence study confirmed the positions of the excited levels of Pm-147. The 77-kev line was placed between the 409 and 490-kev levels. Angular correlation measurements permitted the assignment of angular moments of 5/2 or 7/2 to the 685-kev level. The associated mixing coefficients were also obtained.

3.5.5.18 EXCITED STATES OF Pm-147. E. Spring (Department of Physics, University of Helsinki, Finland). Physics Letters, 7, 3, 218-9, November 1963.

The present work relates to continued study of Pm-147 and has as ((its)) aim a systematic study of singles and coincidence spectrum, and fixing the spins for a number of the excited states by means of angular correlation measurements. In this investigation, use was made of a source of Nd-147 in solution obtained from Harwell. The source had the form of a cylinder 2 mm in diameter and 5 mm in length. All spectra were measured 4-6 weeks after radiation to avoid disturbances arising from activities of shorter life. Several single spectra were measured and analyzed. The pulse height spectra of gamma rays in coincidence with the following gate pulses were recorded: 85-110 kev, 110-135 kev, 135-160 kev, 170-190 kev, 190-230 kev, 240-280 kev, 380-440 kev, 440-480 kev, 530-570 kev, 580-620 kev, and 670-710 kev. The decay scheme ((for Nd-147)) proposed in Figure 1 (not shown) is based on these measurements. The relative intensities are expressed as percentages of the intensity of the 531 kev gamma transmission. The errors are  $\pm 10\%$  of the absolute intensity for the strong transitions but increase to  $\pm 50\%$  for the weak ones.

3.5.5.19 RAI-202. THE DEVELOPMENT OF LARGE BETA RADIATION SOURCES AND THEIR APPLICATIONS. Jacques J. Weinstock, Yael Miron, and P. Murali Krishma (Radiation Applications, Inc., Long Island City, N. Y.). July 1963. 140 p.

An extensive research and development program produced vitreous enamel formulations suitable for the fabrication of a large planar beta radiation sources using Pm-147, Ce-144 or Sr-90 as the active component. The physical and chemical properties of these enamels were characterized, and actual source dosimetry patterns were established for Pm-147 and Ce-144 sources. These enamels allow the fabrication of beta sources containing up to 5 curies per square centimeter of Pm-147 in a 4-mil thick coating, up to 5 C/cm<sup>2</sup> of Ce-144 in an 8-mil coating, and up to 1 C/cm<sup>2</sup> of Sr-90 in an 8-mil coating. Such sources will deliver surface and near surface dose rates in the range of 1 to 10 megarads per hour. Actual dosimetry patterns are presented for enameled Ce-144 and Pm-147 sources containing approximately 0.1 millicurie per square centimeter. These dosimetry measurements are compared with theoretical calculations and show generally good agreement.

3. 5. 5. 20 HW-77375. SHIELDING REQUIREMENTS FOR PROMETHIUM SOURCES. H. H. Van Tuyl, F. P. Roberts, E. J. Wheelwright (Hanford Atomic Products Operation, Richland, Washington. April 1963.

A theoretical analysis of the shielding required for large amounts of promethium was made, including an analysis of the radiation contributions from Pm-146 and Pm-148 at different times after reactor discharge. The results are compared with experimental determinations on samples of promethium at different ages, and promethium is compared with other radionuclides which may be used in power sources. As a first approximation, shielding requirements for promethium aged less than 2 years from reactor discharge are comparable to those for Sr-90 or Cs-137. For promethium aged over 2 years, shielding requirements are comparable to those for Pu-238. Shielding an aged (over 2 years) promethium oxide source to 10 mr/hr at 1 meter would require about 1/4 inch of uranium for a 100 watt source, and 2/3 inch for a 1000 watt source. For optimum geometries, these shield weights correspond to 3 and 4 times the source weight, respectively. Shielding to a dose rate of 1 r/hr at 1 meter would require shielding of weight less than one tenth of the source weight. The former case would permit close approach of personnel, while the latter would afford ample protection for most auxiliary equipment.

3. 5. 5. 21 HW-77296. THE HALF LIFE OF PROMETHIUM-147. F. T. Roberts, E. J. Wheelwright, and W. Y. Matsumoto (Hanford Atomic Products Operation, Richland, Washington, General Electric Corporation) April 1963

((This report describes a Pm-147 half life determination based on absolute beta counting of a solution prepared from a weighted amount of very highly purified promethium oxide.)) The specific activity of Pm-147 has been determined to be  $2.026 \pm 0.048 \times 10^{15}$  disintegration/minutes/gram which corresponds to a half life of  $2.67 \pm 0.06$  years. This value was obtained by 4 pi beta counting known amounts of promethium. The chemical purity of the promethium was verified by emission in absorption spectroscopy and by chelometric titration. The promethium used to prepare a standard solution was purified from Hanford fission product waste. The recovery process was a chromatographic displacement technique from which more than 10 grams of highly purified promethium was obtained. The promethium used in making the standard solution was taken from the center of the elution

band to insure maximum chemical and radiochemical purity.

Promethium was precipitated as the oxalate after filtering and washing with dilute oxalate acid the precipitate was ignited at 750°C for 6 hours to convert to the oxide. A sample of the prepared oxide was accurately weighed, dissolved in 1.0 molar HClO<sub>4</sub> and diluted to a known volume with 1.0 molar HClO<sub>4</sub>. This solution was used in all experiments. The purity of the promethium was checked by emission spectroscopy and found to contain only trace amounts of metallic impurities. Radiochemical analysis show no detectable impurities other than Pm-146 and Pm-148. Both of these activities were less than 10<sup>-4</sup>% of that of Pm-147.

3. 5. 5. 22 MEASUREMENTS OF THE MEAN BETA DISINTEGRATION ENERGIES OF P-32 AND Pm-147 AT LOW TEMPERATURES. Vaeinoe Hovi and Lasse Niemelae (Univ. of Turku, Finland). Ann. Acad. Sci, Fennicae. Ser. A, VI, No. 103, 1-10 (1962) (In English)

By using an isothermal microcalorimetric method the mean beta disintegration energies of P-32 and Pm-147 were determined at liquid nitrogen temperatures. Taking into account the bremsstrahlung correction, the value of  $691 \pm 20$  kev was obtained for the mean beta disintegration energy of P-32, in agreement with experimental data and Fermi's theory. In the case of Pm-147 the measured mean beta disintegration energy was  $70.4 \pm 4$  kev, about 10% higher than that obtained from the beta spectrum and the Fermi theory.

3. 5. 5. 23 ACCURATE MEASUREMENT OF THE HALF LIFE OF THE 91 KEV STATE OF PROMETHIUM-147. Beekhuis H. Physica 28, 1199-1200, 1962

A half life of  $2.59 \pm 0.02$  nanoseconds (estimated probable error) was found for the 91 kev level in Pm-147 excited in the beta decay of Nd-147 from delayed coincidence measurements between beta rays and L conversion electrons.

3. 5. 5. 24 LOW ENERGY PHOTON EMISSION FROM RADIOISOTOPES. Farno L. Green, Transactions of the American Nuclear Society 5, 1, 207, June 1962

This paper is primarily concerned with the emission rates of low energy photons allowed by nature. The low energy range considered is approximately 20 to 300 kev. Table 1 ((not shown)) presents the

theoretical maximum number of photons in a metallic source of two millimeters diameter and two millimeters length in which every atom is considered to be radioactive. ((In addition to several other isotopes, the following information is given for promethium: half life, 2.6 years; disintegration rate (curies per pellet), 46; approximate photons per disintegration, 0.03; approximate photons per second,  $5.2 \times 10^{10}$ ).

- 3.5.5.25 HALF LIVES OF RADIONUCLIDES--II. E. I. Wyatt, S. A. Reynolds, T. H. Handley, W. A. Lyon and H. A. Parker (Analytical Chemistry Division Oak Ridge National Laboratory, Oak Ridge, Tennessee). Nuclear Science and Engineering 11, 74-75, 1961

Half lives of 21 radionuclides are reported. In general the activity of a purified sample was followed for two or more half lives. The data were analyzed by least squares using desk calculators or computers. Two half life values reported previously have been redetermined with improvement.

((Determination for Pm-147 is as follows: half life,  $2.50 \pm 0.03$  years; time observed in half lives, 2.0; instrument used for half life determination, a 2 pi internal source gas flow proportional counter.))

- 3.5.5.26 DIRECTIONAL CORRELATION OF GAMMA TRANSITIONS IN PROMETHIUM-147. Babulai Sarat, Jambuyairan, and M. R. Gonte (Atomic Energy Establishment Trombay, Bombay, India). Physical Review 124, 178-82, October 1961

The angular correlations of five different gamma ray cascades involving the states of Pm-147 excited in the decay of Nd-147 have been studied. The observed correlation functions for the various cascades are presented. The analysis of the above correlation functions with the ground state spin of Pm-147 as  $7/2$  and the consideration of the log ft value for the beta transitions from Nd-147 state of spin  $5/2$  give the spin values for the 91, 413, 491, 533, and 690 keV excited states as  $5/2$ ,  $7/2$ ,  $7/3$ ,  $5/2$  and  $5/2$ , respectively. The data of Bishop, et al, and Ambler, et al, on nuclear alignment experiments have been re-analyzed. Results are consistent with the above spin assignment for  $5/2$  for both the 91 and 533 keV levels.

3. 5. 5. 27 STUDY OF THE EMISSION OF NEGATIVE ELECTRONS ACCOMPANYING AND ACTIVITIES BY THE COINCIDENCE TECHNIQUE. Maurice Duquesne (College de France, Paris). Ann.Phys. (13), 6: 643-702 (May, June 1961) (In French)

A study is made to extend as far as possible the utilization limits of the coincidence method ( $\beta - e^-$ ) by working out a geometry and methods adapted to eliminate parasite coincidences. This coincidence technique is then applied to the study of some nuclear physics problems. An  $e^-$  spectrum in coincidence with the beta radiation was detected for the pure beta emitters S-35, P-32, Pm-147, and RaE. The maximum energy of the spectra is in sharp disagreement with the energies fixed by the theory of self-ionization; nevertheless, the total intensity of the phenomena is of the order of magnitude fixed by this theory of self-ionization; nevertheless, the total intensity of the phenomena is of the order of magnitude fixed by this theory and corresponds with the measurements of the intensity of the characteristic x radiation. A similar study was made for the alpha emitter Po-210 and it was shown that the energy and the intensity of the  $e^-$  emitted correspond to the order of magnitude fixed by the theory of self-ionization.

3. 5. 5. 28 GAMMA-GAMMA DIRECTIONAL CORRELATIONS IN Nd-147. Atam P. Arya (Department of Physics, The Pennsylvania State University Park, Pennsylvania). Physical Review 102, 1226, May 1961

Directional correlation measurements have been made on the 320- to 92-kev and 280- to 320-kev gamma-ray cascades in Pm-147 following the decay of 11.1 day Nd-147 with a coincidence scintillation spectrometer using NaI detectors. The observed correlation functions are:

$W(\theta) = 1 - (0.1030 \pm 0.0298) P_2(\cos \theta) + (0.0107 \pm 0.0099) P_4(\cos \theta)$   
and  $W(\theta) = 1 + (0.0710 \pm 0.0162) P_2(\cos \theta) - (0.0126 \pm 0.0103) P_4(\cos \theta)$ ,  
respectively, for the two cascades. The energy levels of Pm-147 at ground state, 92 kev, 410 kev, and 690 kev were found to be  $7/2^+$ ,  $7/2^+$ ,  $7/2^+$ , and  $5/2^+$ , respectively. It was found that the 92-kev gamma ray has a mixture of  $(95 \pm 2)\%$  M1 and  $(5 \pm 2)\%$  E2 with  $\int_{92} = (0.229 \pm 0.143)$ , the 320 kev gamma ray has a mixture of  $\%$  M1 with 99% E2 with  $\int_{320} = +9.95 \pm 0.11$ , and the 280-kev gamma ray has a mixture of 99% M1 and 1% E2 with  $\int_{280} = 0.11 \pm 0.11$ .



Three neodymium samples, in the form of metallic fused chips 99.9% pure, were irradiated at different times in the thermal neutron flux of the Pennsylvania State University Research Reactor for different lengths of time varying from 75 to 130 hrs. Because of the simultaneous production of Nd-149 ( $T_{1/2} = 26$  hrs), irradiated samples were allowed to decay from three to six weeks in order to get rid of these short-lived activities. The correlation experiments were started only when the activity left was mainly Nd-147 (11.1 days).

- 3.5.5.29 THE  $g$  FACTOR OF THE 92-KEV LEVEL AND OTHER ANGULAR CORRELATION MEASUREMENTS ON Pm-147. E. Bodenstein, H. J. Korner, F. Frisius, D. Hovestadt, and E. Gerdau (Universitat, Hamburg). Z. Physik 160, 33-46 (1960) (In German)

The gyromagnetic ratio of the 92 keV level of Pm-147 was determined by a measurement of the 321 keV-92 keV angular correlation in an external magnetic field of 15,000 gauss. The result ( $g = +1.42 \pm 0.20$ ) implies corrections for the paramagnetism of the 4f-electron shell and for a time dependent attenuation by internal fields. A new determination of the half life of the 92 keV level confirmed the known value. The half life of the 412 keV level was found to be  $T_{1/2} \leq 5 \cdot 10^{-10}$  s. The following angular correlations were measured: 321 keV-92 keV cascade  $W(\theta) = 1 - (0.087 \pm 0.008) \cdot P_2 - (0.001 \pm 0.003) \cdot P_4$ , 441 keV-92 keV cascade  $W(\theta) = 1 + (0.065 \pm 0.010) \cdot P_4$ , 400 keV-92 keV cascade  $W(\theta) = 1 - (0.022 \pm 0.008) \cdot P_2 - (0.002 \pm 0.009) \cdot P_4$ , 277 keV-92 keV triple cascade  $W(\theta) = 1 + (0.0016 \pm 0.0027) \cdot P_2 - (0.002 \pm 0.003) \cdot P_4$ , 277 keV-321 keV cascade  $W(\theta) = 1 + (0.0117 \pm 0.0025) \cdot P_2 - (0.0067 \pm 0.0033) \cdot P_4$ , 120 keV-321 keV cascade  $W(\theta) = 1 - (0.029 \pm 0.011) \cdot P_2 - (0.031 \pm 0.012) \cdot P_4$ , and 400 keV-198 keV cascade  $W(\theta) = 1 - (0.067 \pm 0.009) \cdot P_2 + (0.001 \pm 0.011) \cdot P_4$ .

The spins and multipolarities of different gamma transitions are derived from these results.

- 3.5.5.30 MEASUREMENT OF  $g$  FACTORS OF SEVERAL SHORT LIVED NUCLEAR STATES IN ODD-MASS NUCLEI. G. Manning and J. D. Rogers (California Institute of Technology, Pasadena, California). Nuclear Physics 15, 166-186, February 1960

((The first portion of the abstract has to do with As-75, Lu-174, and Hf-177 and has been omitted.)) The  $g$ -factor of the 91 keV level of

Pm-147 has also been studied. A definite rotation of the angular correlation being observed, computation of the g-factor from the observed data is uncertain because the time dependent attenuation of the gamma-gamma correlation is not known. The results are  $g = (0.9 \pm 0.2)/G_2$ . The effect of the thermomagnetic nature of the rare earth ions on such measurements on this group of nuclei is discussed as appendix. Other assumptions may be in deducing the g-factor from the observed rotations of the angular correlations are also discussed in this paper.

- 3.5.5.31 RESEARCH NOTES, THE DECAY OF Pm-147. R. J. Jakeways and W. G. Rosser (Physics Department, The University, Exeter). Proceedings of the Physical Society (London) 74, 478-9, October 1959

((The purpose of the work described in this article was to investigate the possibility that promethium decays via an excited level at 121 kev in Sm-147 using beta gamma coincidence techniques. The conclusion reached was that the 121 kev level does not arise from the decay of Pm-147.))

A sample of Pm-147 was obtained from the Radio Chemical Center, Amersham, in the form of a solution in 1 normal HCl. A source was prepared by evaporating a few drops of this solution on a thin mica disc. The source was covered with a collodion film.

- 3.5.5.32 DIRECT DETERMINATION OF THE HALF LIVES OF NINE NUCLIDES. J. Paul Cali and L. F. Lowe (Cambridge Research Center, U. S. Air Force, Bedford, Massachusetts). Nucleonics 17, 10, 86-8, October 1959

The half lives of nine isotopes have been newly determined by direct measurement after extensive radiochemical purification. These values are compared in the table with previous values frequently quoted in the literature. To insure radiochemical purity extensive chemical procedures were used for each of the elements after its irradiation ((half life determined for Pm-147 was  $2.7 \pm 0.1$  years.))

- 3.5.5.33 Refer to 3.2.5.18. HALF LIFE DETERMINATIONS OF SOME RADIONUCLIDES. Canad. Journ. Phys. 35, 16-20, 1957

3. 5. 5. 34 APPLICATIONS OF A MULTI-CHANNEL GONIOMETER.  
Torsten Lindquist and Erik Karlsson, Arkiv for Fysik 12,  
519-36, 1957

A multi-channel goniometer (MCG) for gamma-gamma angular correlation measurements is described. The apparatus has four channels. Various methods of measurement and a complete treatment of data are given. The MCG has been carefully tested with well-known correlations (Bi-207, Hs-181, and Co-60). As applications the angular correlations of gamma cascades from Ba-131 and Nd-147 have been investigated. Both cases involve forbidden magnetic dipole transitions from the mixing ratios determined in the correlation experiments. It is possible to investigate the strength of forbiddenness. An upper limit of the g-factor of the 92 kev level in Pm-147 is determined by studying the influence of a magnetic field on the angular correlation.

The anisotropy of the 320-92 kev cascades in Pm-147 was found to be  $A = 0.110 \pm 0.010$  which is large enough for observing an attenuation. The lifetime of the 92 kev level was  $3.5 \times 10^{-9}$  seconds, however, even with a fairly high field, 25,000 gauss, the attenuation was very small:  $A(25,000 \text{ gauss}) = -0.094 \pm 0.027$ . Taking the limits of everything into consideration it is possible to give only an upper limit to the g-factor in this case. It was not possible to measure the sign of g.

3. 5. 5. 35 CONTRIBUTION AU ETUDE DES PHENOMENES DE  
FREINAGE INTERNE ET D'AUTOIONISATION ASSOCIES  
A LA DESINTEGRATION BETA. Langevin (Joliot)  
Annules Physique 2, 16, 1957. Chap. 3 Rayonnement  
de freinage interne et externe de S-35 et Pm-147. ((This  
article is in French and no attempt at translation has been  
made.))

3. 5. 5. 36 INTERNAL BREMSSTRAHLUNG FROM Pr-143 AND Pm-  
147. N. Starfelt and J. Cederlund (Radiation Physics  
Department University of Lund, Lund, Sweden). Physical  
Review 105, 241-6, January 1957

A NaI(Tl) scintillation spectrometer was used for the study of the spectral distribution of the internal bremsstrahlung from the beta emitters Pr-143 ( $E_{\text{beta-max}} = 922 \text{ kev}$ ) and Pm-147 ( $E_{\text{beta-max}} = 226 \text{ kev}$ ). Both the shape of the corrected experimental spectrum and the

total yield of internal bremsstrahlung measured per beta decay disagree with the original Knipp-Uhlenbeck theory and with the improved version of this theory which takes into account the nuclear coulomb field. For Pr-143 the measure bremsstrahlung yield exceeds that of the Knipp-Uhlenbeck theory by factors of 1.58 (1.48) and 2.56 (1.50) at 50 and 500 kev, respectively. The numbers in parentheses refer to the coulomb corrected theory. The corresponding factors for Pm-147 are 1.58 (1.37) and 6.1 (2.54) at 25 and 150 kev, respectively. The sources were prepared from carrier free Pr-143 and Pm-147 solutions obtained from the Atomic Energy Research Establishment, Harwell. The absolute activities of the solutions were measured at Harwell by four pi counting and were given an accuracy of  $\pm 2\%$  for Pr-143 and  $5\%$  for Pm-147. The sources were made on 10 micrograms/cm<sup>2</sup> formvar by evaporation with an infrared lamp. Four sources with activities ranging from 34 to 156 microcuries activity and thicknesses between 160 and 600 micrograms/cm<sup>2</sup> and three Pm-147 sources with activities from 78 microcuries and average thicknesses between 20 and 200 micrograms/cm<sup>2</sup> were used in the IB measurements. The fact that all the sources used gave the same spectrum within the experimental errors of about  $5\% \pm$  at low photon energy strongly supports the assumption that the external bremsstrahlung emitted from the source was negligible in comparison with the internal bremsstrahlung ((Figure 2, shows experimental pulse height distributions and background spectra for 154 microcuries Pr-143 and 465 microcuries Pm-147; Figure 3, shows the total correction factor converting the scintillation pulse height distribution of the internal bremsstrahlung of Pr-143 and Pm-147 into the true photon spectrum; Figure 5, presents a graph of the internal bremsstrahlung from Pm-147; Figure 7, shows a comparison between experimental and theoretical internal bremsstrahlung yield for P-32, F-35, Pr-143 and Pm-147.)) ((Figures 2 through 7 are not presented.))

- 3.5.5.37 Refer to 3.1.5.14. DESIGN AND PERFORMANCE OF A THIN MAGNETIC LENS BETA RAY SPECTROMETER. Proceedings of the Indian Acad. Sei. 44A, 111-122, 1956
- 3.5.5.38 HALF LIVES OF Ce-144, Co-58, Cr-51, Fe-55, Mn-54, Pm-147, Ru-105, and Sc-46. R. P. Schuman, M. E. Jones, and Mrs. A. C. Mewherter (Knolls Atomic Power Laboratory, Schenectady, New York). Journal of Inorganic Nuclear Chemistry 3, 160-163, October 1956

Half lives have been obtained for a number of radionuclides by following the decay of purified samples over periods of time as long as six years. The data were treated by the method of least squares.

The half lives are:  $284.5 \pm 10$  days for Ce-144,  $71.3 \pm 0.2$  day for Co-58,  $27.8 \pm 0.1$  day for Cr-51,  $2.60 \pm 0.02$  year for Fe-55,  $2.78 \pm 5$  days for Mn-54,  $2.66 \pm 0.02$  year for Pm-147,  $366.6 \pm 0.9$  day for Ru-106, and  $84.1 \pm 0.3$  day for Sc-46.

The Pm-147 sample was isolated from fission products by a method which separates the rare earths other than cerium from the other fission products and cerium. Before the decay curves were started the sample had decayed several years so only the long lived rare earth fission products remained. The only fission products other than Pm-147 that would be present are 73 year Sm-151 which because of the very low energy beta rays would not count under the conditions used and 1.7 year Eu-155 which has a low fission yield, about 1% that of Pm-147. In calculating the half life no correction was made for the Eu-155 impurity.

- 3.5.5.39 PR-P-29 RESEARCH AND DEVELOPMENT, PROGRESS REPORT. January 1, 1956 to March 31, 1956 (Atomic Energy of Canada Limited, Chalk River Project, Physics Division, Chalk River, Ontario). Nuclear Physics 1 by J. N. Robson

Section 1.11 Decay of Nd-147 and the level scheme of Pm-147. Through the use of a ring focus beta ray spectrometer, operated at 0.8 resolution, the beta spectra in coincidence with the 532 kev and 690 kev gamma rays have been studied. Preliminary values of the end points of these spectra indicate beta transitions directly to levels at 532 kev and 690 kev in Pm-147. Rutledge, et al (Physics Review 86, 775, 1952) and Hans, et al (Physics Review 97, 1267, 1955) have proposed two differing level schemes for Pm-147. The present work tends to confirm that of Hans with the possible addition of another level to explain the observed results.

- 3.5.5.40 ON THE INTERNAL AND EXTERNAL BREMSSTRAHLUNG OF Pm-147 and Pr-143. Helene Langevin-Joliot. Compt. rend. 241, 1286-8 (1955, November 7) (In French)

The internal bremsstrahlung spectrum of Pm-147, obtained by a scintillation spectrometer in an appropriate arrangement, disagrees with theory in both its shape and intensity. The number of photons reaches 450% of the theoretical value above 60 kev. The first results on Pr-143 also indicate an excess of photons.

3. 5. 5. 41 THE RELATIVE ABUNDANCES OF NEODYMIUM AND SAMARIUM ISOTOPES IN THE THERMAL NEUTRON FISSION OF U-235 AND U-233. E. A. Melaika, M. J. Parker, J. A. Petruska and R. H. Tomlinson, Canadian Journal of Chemistry 33, 830, 1955

The relative fission yields of neodymium and samarium isotopes have been measured with a mass spectrometer for samples of natural uranium and U-233 that have been irradiated with moderated neutrons. The cross sections for neutron capture by Sm-149 and Sm-151 have been determined to be  $66,200 \pm 2500$  barns and 12,000 barns, respectively, relative to the cross section of a B-10 monitor. The half lives of Pm-147 and Sm-151 have been evaluated to be  $2.52 \pm 0.08$  year and approximately 93 years, respectively, from samarium fission yield data for samples differing in age by 7 years.

3. 5. 5. 42 THE BETA RAY ABSORPTION SPECTRUM OF Pm-147 AND ITS APPLICATION TO THICKNESS MEASUREMENT. L. Mandel, B.Sc., Ph.D., A. Inst. P. (Central Instrumentation Imperial Chemical Industries Limited, Welwyn). British Journal of Applied Physics 5, 287-9, August 1954

The absorption in aluminum of the beta rays from Pm-147 has been measured and found to be nearly exponential with an absorption length of  $6.78 \text{ mg/cm}^2 \pm 3\%$ . Curves are given which relate the measuring accuracy with absorber thickness in rapid response beta ray gages using Pm-147 and Tl-204 as sources. These show that under the same conditions the use of Pm-147 leads to greater accuracy at thickness measurement by a factor of 4.

The source for these measurements was supplied by the Radio Chemical Center, Amersham and consisted of an evaporated solution of Pm-147 in hydrogen chloride of approximately 1 mc strength covering an area of about  $1 \text{ cm}^2$ . ((Figure 2, not reproduced here, presents a comparison of the absorption spectra of Pm-147 and Tl-204.))

3. 5. 5. 43 DETERMINATION OF THE HALF LIVES OF SOME MAGNETIC DIPOLE GAMMA RAY TRANSITIONS. R. L. Graham and R. E. Bell, Canadian Journal of Physics 31, 377, 1953

A number of magnetic dipole gamma ray transitions have been studied using a coincidence circuit of short resolving time, a two lens single gamma ray spectrometer, a pair of lens spectrometers placed end to end with coincidence counting of the focused radiations from a single source, and a scintillation spectrometer. Life times have been measured using the delayed coincidence method and where feasible conversion coefficients and K/L ratios obtained. Comparison is made with theoretical estimates of the life time-energy relation for M-1 gamma ray transitions. The net results are as follows: ((Although several isotopes are mentioned only that information for Pm-147 is given)) E (kev), 91.5; half life (seconds,  $(2.44 \pm 0.08) \times 10^{-9}$ ; K-1 ratio, exp 7.3, M-1 7.7 E-2 0.5; alpha, exp 1.8, M-1, 1.9; total alpha 2.2.

3. 5. 5. 44 IDENTIFICATION AND CHARACTERISTICS OF APPROXIMATE FOUR YEAR Pm-147, PAPER 192.  
J. A. Marinsky and L. E. Glendenin, Radio Chemical Studies, The Fission Products, Book 2, part V, papers 53-199, McGraw-Hill Book Company, Inc. 1951

A detailed study has been made of the long lived isotope of promethium that occurs in fission. The separation and identification of this activity as an isotope of promethium was achieved by the Amberlite (ion exchange) resin absorption elution method. Its decay is characterized by a 0.20 Mev beta ray. No gamma ray is observed. An apparent daughter relationship has been shown to exist between the 11 day Nd and the long lived promethium. The fission yield of this chain is  $2.6 \pm 0.2\%$ . A mass of 147 has been assigned the chain by mass spectroscopy. The half life of the long lived promethium activity has been calculated as 4.8 years from its observed growth from 11 day Nd but as 4.4 years from the amount of activity formed in fission ((using the fission yield of the 11 day Nd parent)).

3. 5. 5. 45 ABSORPTION AND EMISSION SPECTRA OF PROMETHIUM.  
William F. Meggers, Bourdon F. Scribner and William R. Bozman, Journal of Research of the National Bureau of Standards, Vol. 46, No. 2, 85-98, February 1951, Research Paper 2179.

Five milligrams of Pm-147 separated from fission products at the Oak Ridge National Laboratory were loaned by the United States Atomic Energy Commission for this investigation. The absorption spectrum of this sample in solution was plotted between 2500 and

10,000 angstroms. The principal bands having wavelengths 494.5, 548.5, 568.0, 685.5, 735.5 millimicrons ( $\pm 0.5$  millimicrons). Small portions of the sample were dried on copper electrodes employed in photographing alternating current arc and spark spectra with a concave grating of 22 foot radius. Excepting Sm, into which Pm decays, no other rare earths could be detected in this sample, but common chemical contaminants were troublesome. Between 2200 and 6900 angstroms the wavelengths and relative intensities of more than 2200 new spectral lines were determined, but it is not possible to differentiate Pm I and Pm II lines with the light sources employed. The strongest Pm lines have wavelengths 3892.16, 3910.26, 3919.09, 3957.74, and  $3998.96 \pm 0.02$  angstroms. Hyperfine structure is suspected in some Pm lines indicating that the nuclei of Pm-147 atoms disposes mechanical and magnetic moments. Confirming the findings of the Oak Ridge National Laboratory, both the absorption and emission spectrum identify this fission product as a new element of rare earth type. They provide positive proof that the long sought element with atomic number 61 has been discovered.

((The balance of this article is concerned with the optical spectra of Pm-147 and is not described here.))

3.5.5.46 LOW ENERGY BETA RAY SPECTRA. Pm-147, S-35.  
L. M. Langer, J. W. Motz and H. C. Price, Jr.  
(Department of Physics, Indiana University, Bloomington,  
Indiana). Physical Review 77, 798-805, March 1950

The beta spectra of S-35 and Pm-147 have been measured in order to study further the nature of any low energy deviation from the Fermi theory of beta decay. Measurements were made within relatively uniform sources in both the 40 centimeter radius of curvature spectrometer and also in a small  $180^\circ$  focusing Helmholtz coil spectrometer designed specifically for low energy spectra. The thinnest sources were less than ten micrograms/cm<sup>2</sup>. Using zapon counter windows ranging from 1.5 to 3 micrograms/cm<sup>2</sup> and also a windowless counter technique, Fermi plots were obtained which showed how the measure distribution of particles at low energy depends on both source and counter window thickness. Favorable experimental conditions yielded a straight line Fermi plot for Pm-147 above 8 kev. Less favorable conditions resulted in a straight line plot for S-35 down to at least 50 kev. Thus, S-35, which is allowed, and Pm-147, which is probably once forbidden, are found to have spectra of the allowed shape. It is concluded that under very favorable experimental



conditions there is probably no real disagreement between the observed momentum distribution and that predicted for an allowed transition by the Fermi theory. On the basis of an improved calibration the following end points are obtained: Pm-147,  $223.2 \pm 0.5$  kev and S-35,  $167.0 \pm 0.5$  kev.

In view of the conflicting reports in the literature on the effect of source thickness, we made a study of auto-radiographs of sources prepared from chemical solutions. By this technique it was found that, in general, such sources, though appearing uniform, may in many cases have variations of intensity of as much as 100 fold. Under these circumstances, the average source thickness as reported by various investigators does not have much meaning. We are as yet unable to deposit completely uniform sources from chemical solution. Our best technique consists of wetting the portion of the backing foil on which the source is to be placed with one drop of 5% solution of Lily insulin, 40 units per cc in water. A drop of radioactive solution applied any place in this area will spread over the entire region defined by the insulin. Fairly uniform sources are easily made on larger areas by covering the region with narrow insulin lines and putting a small drop of liquid source on each. Extremely uniform sources can, if activity permits, be successfully prepared by thermal evaporation in vacuum.

- 3.5.5.47 LOW ENERGY BETA RAY SPECTRA PROMETHIUM-147 AND SULPHUR-35. H. C. Price, Jr., J. Motz, and L. M. Langer (Indiana University). Physical Review 77, 744, 1950

The beta spectra of S-35 and Pm-147 have been measured in an attempt to study further the nature of the low energy deviation from the Fermi previously reported. Fermi plots were obtained which for Pm-147 were straight down to 8 kev. The end point of Pm-147 was found to be  $223.2 \pm 0.5$  kev.

- 3.5.5.48 Refer to 3.3.5.54. THE BETA SPECTRA OF Cs-137, Y-91, Pm-147, Ru-106, Sm-151, P-32, and Pm-170. Harold M. Agnew (Institute of Nuclear Studies, University of Chicago, Chicago, Illinois). Physical Review 77, 655-60, March 1950

((Promethium-147 half life 3.7 years:  $t_{1/2} = 4.01 \times 10^7$ ; Sources of promethium weighing less than  $0.05 \text{ mg/cm}^2$  were prepared from material obtained from Oak Ridge.))

- 3.5.5.49 THE BETA SPECTRUM OF Pm-147. L. Lidowsky, P. Macklin and C. S. Wu (Pupin Physics Laboratories, Columbia University, New York, New York). Physical Review 76, 1888-9, December 1949

High specific activity Pm-147 has recently become available from the Isotopes Division of the AEC at Oak Ridge where it is made as a fission product. This high specific activity material, when used in the high transmission solenoidal focusing beta ray spectrometer, makes it possible to investigate its beta spectrum when using sources less than 30 micrograms/cm<sup>2</sup> thick. The Pm-147 received as Pm-147 Cl<sub>3</sub> in 0.1 normal HCl was evaporated to dryness and then re-dissolved in distilled water to which a small amount of wetting agent (Antorox) was added. A 0.01 milliliter drop placed on a collodion backing approximately 10 micrograms/cm<sup>2</sup> formed a uniform source spread over a circular area of about 0.5 cm<sup>2</sup> (an end point for Pm-147 of  $227 \pm 1$  kev was obtained). The plot was straight from the end point down to at least 35 kev. Below this energy the curve deviated slightly upward and then again downward in a manner that could be attributed to finite source thickness end window absorption. The above end point in conjunction with the reported value of the half life (3.7 years) yielded a ft value for the transition of  $1.2 \times 10^7$  which foreign element with a Z of 61 indicates a first or second forbidden spectrum. Although the curve has an allowed shape, this is not a contradiction since, for certain interactions, a forbidden transition may exhibit allowed shape.

- 3.5.5.50 THE K SPECTRA OF ELEMENT 61. L. E. Burkhar, W. F. Peed, E. J. Spitzer (Clinton Engineering Works, Carbide and Carbon Chemicals Corporation, Oak Ridge, Tennessee). Physical Review 75, 86-9, 1949

The K alpha 1, K alpha 2, K beta 1 and K beta 2 lines of the X-ray spectra of element 61 were obtained from 1.5 mg of sample of the chloride salt. The sample was fused on a copper target and bombarded with electrons from a 70 kev source. Wavelength measurements were made from a microphotometer tracing of the film. After one exposure of the sample neodymium and samarium were added to the spectrum of the three elements was recorded to show the position of element 61 in the periodic table.

- 3.5.5.51 MASS SPECTROGRAPHIC MASS ASSIGNMENT OF RADIO-ACTIVE ISOTOPES. Richard J. Hayden (Metallurgical Laboratory University of Chicago, Chicago, Illinois). Phys. Rev. 74, 650, Sept. 1948

((Characteristics of positive ion emission for various compounds on a tungsten filament are cited. Methods of analysis of mixtures of radioactive isotopes by mass spectrograph are described. The following mass assignments are made. Only those for elements of interest are given, 3.7 year element 61, 147; 21 year strontium, 90.))

3. 5. 5. 52 THE CHEMICAL IDENTIFICATION OF RADIOISOTOPES OF NEODYMIUM AND ELEMENT 61. J. A. Marinsky, L. E. Glendenen and C. D. Coryell, Journal of the American Society 69, 2781, 1947

((Article describes the chemical identification of Pm-147.))

3. 5. 5. 53 UCRL-10245. INVESTIGATION OF ELECTRONIC AND NUCLEAR PROPERTIES OF SOME RARE EARTH ISOTOPES. (University of California, Ernest O. Lawrence Radiation Laboratory). Page 22, Section A

Promethium, atomic number 61 has no stable isotopes, is a fission product, and has a half life of 2.6 years. Its beta spectrum has been measured accurately, and although first forbidden exhibits the allowed shape, there is evidence of a gamma spectrum.

### 3.6 POLONIUM-210

#### 3.6.1 Nuclear Characteristics

3.6.1.1 Atomic Number - 84

3.6.1.2 Atomic Weight - 210

3.6.1.3 Half Life

$138.3763 \pm 0.0017$  days

Reference 1

3.6.1.4 Energy Levels and Decay Schemes

Po-210  $\xrightarrow{\alpha}$  Pb-206

Alpha ( $5.3049 \pm 0.006$ ) Mev

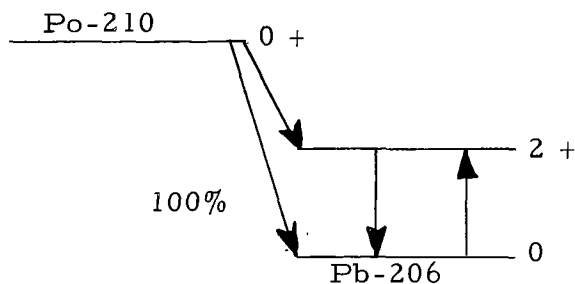
Reference 2

Gamma ( $0.800 \pm 0.006$ ) Mev

Reference 3

Probability of branching to the  
first excited level of Pb-206 =  
 $(1.2 \pm 0.2) \cdot 10^{-5}$

Reference 4



Reference 5

### 3.6.1.5 Other Sources of Radiation

Po-210 emits neutrons from ( $\alpha$ , n) reactions between the emitted alpha particles and impurities (oxygen) in the isotope compound. In the case of pure polonium metal a value of 70 neutrons/second curie has been measured at the Mound Laboratories (Ref. 6). This is equivalent to  $3.15 \times 10^5$  n/sec gram. Other compounds studied at the Mound Laboratories have emitted as many as 200 n/sec curie.

### 3.6.2 Chemical Characteristics

#### 3.6.2.1 Fuel Forms

Several possible polonium compounds (Ref. 7) are indicated in Table XVI. In addition to compounds in which polonium acts as a metal, a number of polonides are possible. The Mound Laboratories is presently developing high melting temperature polonides of lanthanum and similar metals. Early efforts have demonstrated that lanthanum polonide has a melting point higher than 1200°C. It is expected that compounds will be developed which are stable at temperatures in excess of 1600°C.

#### 3.6.2.2 Material Compatibility

At the present, the only form in which polonium has been produced for use in direct conversion systems, has been the metal. The SNAP-3 generator (demonstration device) showed that polonium can be contained in the elemental form at temperatures as high as 1300°F. However, it is expected that future systems will try to employ some compound of polonium, probably one in the lanthanum family. No chemical compatibility data was found in the open literature.

Table XVI (Ref. 7)

Polonium Compounds			
Compound	Melting Point (°C)	Specific Power Pure (w/gm)	Power Density Theoretical (w/cc)
Po metal	255 (pg. 21, Ref. 9)	141.3	1315
PoO <sub>2</sub>	900*	123.0	1130
PoCl <sub>4</sub>	294	84.2	421 (est)
PoBr <sub>4</sub>	324	55.9	335 (est)
PoNi <sub>2</sub>	625	91	1050

\*Decomposes at 885°C, appreciable O<sub>2</sub> pressure at lower temperatures.

### 3.6.2.3 Effects of Impurities on Nuclear Characteristics

Isotope purity obtainable with Po-210 is quite good, and is given as 95% by reference 8. In shielding calculations performed at the Martin Company, only the 0.804 Mev gamma emitted in the decay chain and neutrons produced by ( $\alpha$ , n) reactions with trace amounts of oxygen found in the metal after fabrication, are considered. This is compatible with the technique used by ORNL.

### 3.6.3 Thermal and Physical Properties

Since the power density and specific activity of pure Po-210 are very high Po-210 is mixed with a "filler" material to form a metal matrix which can be handled. The diluent metal must be chemically inert with a high melting point and high thermal conductivity. The diluents must also be nuclearly inert in the sense that as small a number as possible of secondary particles such as neutrons or gammas are generated due to nuclear reactions.

The allowable power densities are small ( $\sim 150 \frac{\text{watts}}{\text{cm}^3}$ ) compared with the power density attainable with Po-210 ( $1328 \frac{\text{watts}}{\text{cm}^3}$ ) hence a large percentage of diluent must be added and the metal matrix so formed will take on the heat transfer characteristics of the diluents.

The thermal and physical properties of possible filler materials may be found in Table XVII.

Assuming the power density of  $150 \left[ \frac{\text{watts}}{\text{cm}^3} \right]$  mentioned above and using a density of  $9.398 \frac{\text{gm (Po-210)}}{\text{cm}^3}$  (Ref. 9, p. 20) ( $\rho_m$ ) is then  $1.06 \frac{\text{gm (Po-210) in matrix}}{\text{cm}^3}$  and the activity of the matrix (SA)<sub>m</sub> is then  $4.77 \left[ \frac{\text{K curie}}{\text{cm}^3} \right]$  as calculated below:

$$\rho_m \left[ \frac{\text{gm (Po-210) in matrix}}{\text{cm}^3} \right] \times 141.3 \frac{\text{watts}}{\text{gm}} = 150 \frac{\text{watts}}{\text{cm}^3}$$

$$\rho_m = 1.06 \left[ \frac{\text{gm (Po-210) in matrix}}{\text{cm}^3} \right]$$

$$\begin{aligned}
 (SA)_m \frac{\text{curies}}{\text{cm}^3} &= 4500 \frac{\text{curies}}{\text{gm}} = 1.06 \left[ \frac{\text{gm (Po-210) in matrix}}{\text{cm}^3} \right] \\
 &= 4770 \left[ \frac{\text{curies}}{\text{cm}^3} \right] \\
 &= 4.77 \left[ \frac{\text{K curies}}{\text{cm}^3} \right]
 \end{aligned}$$

The detailed thermal and physical properties of Po-210 and a hypothetical matrix based on the preceding calculations are presented in Table XVII.

Table XVII

Physical and Thermal Properties

		Po-210 Metal	Hypothetical Matrix
Thermal Conductivity	$\frac{\text{cal cm}}{\text{sec } ^\circ\text{C cm}^2}$	0.24(a) Ref. 10	--
Specific Power	$\frac{\text{watt}}{\text{gm}}$	141.3(b) Ref. 8	--
Thermal Output	$\frac{\text{watt}}{\text{K curie}}$	32 (c) Ref. 8	32 Ref. 8
Heat Capacity	$\frac{\text{cal}}{\text{gm}^\circ\text{C}}$	0.177(a) Ref. 10	--
Heat of Fusion	$\frac{\text{cal}}{\text{mole}}$	26.31(a) Ref. 11	--
Weight Density	$\frac{\text{cal}}{\text{cm}^3}$	9.298 Ref. 9	8.5(d, e)
Melting Point	$^\circ\text{C}$	255(a) Ref. 11	--
Boiling Point	$^\circ\text{C}$	946.5(a) Ref. 11	--
Specific Activity	$\frac{\text{dis/min}}{\text{micro gram}}$	$1 \times 10^{10}$ (c) Ref. 9	--

### Key to Table XVII

- (a) The metal matrix takes on the properties of the diluent material.
- (b) Final Report, Radioisotope Evaluation Program MNSP-MK-5002. Dec. 23, 1964. Contract No. NAS 5-3879.
- (c) Calculated value.
- (d) The values 150 watts/cm<sup>3</sup> and 8.5 gm/cm<sup>3</sup> are realistic values for specific power and matrix density.
- (e) Weight density of the matrix.

#### 3.6.4 Isotope Production, Availability and Cost

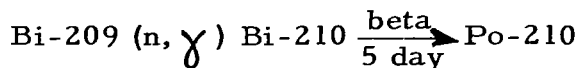
##### 3.6.4.1 Production Method

Po-210 can be produced by a large number of means, including nuclear decay, bombardment with high energy particles, and combinations of the two.

It is produced naturally as a daughter of Bi-210 (RaE), a member of the radium decay chain. A second method of production by decay is electron capture by At-210, a nuclear reaction with a half life of 8.3 hours.

Bombardment of Pb-208 or Bi-209 by charged particles such as alpha particles, deuterons, and protons produce Po-210.

Polonium is produced in quantity by the irradiation of Bi-209 in a heterogeneous thermal reactor to form Bi-210. The Bi-210 decays with a 5 day half life to Po-210.



The neutron absorption cross-section for bismuth is very small (20 millibarns) which results in a yield of roughly one gram of polonium per ton of bismuth irradiated. The bismuth is canned in aluminum container and irradiated for periods of two to three months.



Chemical processing is then employed to separate out the polonium as the pure metal.

#### 3.6.4.2 Availability and Costs

The following prospective production levels of Po-210 were published by the Division of Isotope Development, USAEC, in January 1963. These values are presented below:

	FISCAL YEAR							
	<u>1964</u>	<u>1965</u>	<u>1966</u>	<u>1967</u>	<u>1968</u>	<u>1969</u>	<u>1970</u>	<u>1980</u>
gm	50	100	100	1000	1000	1000	1000	1000
Kw(t)	7	14	14	140	140	140	140	140

The probable cost of Po-210 is very unsure. At present it is approximately 90,000 \$/gram. At a production rate of 100 grams/year, the cost is expected to decrease to 25,000 \$/gram and at 1000 grams/year, a cost of between 3000-9000 \$/gram could be anticipated.

## REFERENCES

1. MLM-1209. Mound Laboratories Monthly Progress Report for July 1964, pg. 12.
2. Absolute Precision Determination of Several Resonance Threshold Energies and the Alpha Particle Energy of Po-210, Part I. Helv Phys. Acta 34, 960-76, 1961.
3. Gamma Radiation from Pb-206. Physical Review 81, 523-6, Feb. 1961.
4. Determination of the Branching Ratio in the Disintegration Scheme of Polonium-210; J. Exptl & Theoret Fiz (USSR) 31, 174-7, Aug. 1956 (translated in Soviet Physics JETP Vol. 4, #2, 165 March 1957).
5. Table of Isotopes, Rev. Mod. Physics, Vol. 30 #2, pt II, April 1958.
6. Personal communication between Ivan Gray of Martin Company and Dr. Grove of Mound Laboratories.
7. Polonium, H. V. Moyer, TID-5221, U. S. Atomic Energy Commission, July 1956.
8. Isotope Property Data Sheet from General Electric Company, Hanford Laboratories, Richland, Washington, C. A. Rohrmann (6/4/63).
9. MIM-1165 (Tr), Translation of p. 2041 to 2122 of Nouveau Traite de Chrmie Minerale, Tome XIII, Part 1, 1961.
10. Nuclear Engineering Handbook, Etherington, McGraw-Hill Book Co. (1958).
11. Giorgi, Dessert, Alstr 17, 2166 (1957).

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gm	50	100	100	1000	1000	1000	1000	1000
Kw(t)	7	14	14	140	140	140	140	140

The probable cost of Po-210 is very unsure. At present it is approximately 90,000 \$/gram. At a production rate of 100 grams/year, the cost is expected to decrease to 25,000 \$/gram and at 1000 grams/year, a cost of between 3000-9000 \$/gram could be anticipated.

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1. MLM-1209. Mound Laboratories Monthly Progress Report for July 1964, pg. 12.
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3. Gamma Radiation from Pb-206. Physical Review 81, 523-6, Feb. 1961.
4. Determination of the Branching Ratio in the Disintegration Scheme of Polonium-210; J. Exptl & Theoret Fiz (USSR) 31, 174-7, Aug. 1956 (translated in Soviet Physics JETP Vol. 4, #2, 165 March 1957).
5. Table of Isotopes, Rev. Mod. Physics, Vol. 30 #2, pt II, April 1958.
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### 3.6.5 Annotated Bibliography

- 3.6.5.1 POLONIUM COMPOUND HEAT SOURCES. Martin R. Hertz (to U. S. Atomic Energy Commission). U. S. Patent 3, 154, 501. Oct. 27, 1964

Improved radioactive materials were developed for use in heat sources. Po-210 was reacted with rare earth elements such as scandium, yttrium, or the lanthanide series to form radioactive compounds or materials of lower vapor pressures and higher melting points than Po-210 itself.

- 3.6.5.2 ALPHA PARTICLE EXCITATIONS IN ATOMIC NUCLEI. L. Simons (Univ. of Helsinki). Phys. Letters, 12: 331-4 (Oct. 15, 1964)

The alpha-particle excitation model is investigated for even-even nuclei, assuming the time-independent Schroedinger equation to be valid and that no spin-orbit coupling exists. Excited states calculated in this model are compared with experimental values in Ca-40, Zn-66, Ce-140, Po-210, S-32, and Ce-142, as regards spin-parities and energy values.

- 3.6.5.3 (MLM-1209) MOUND LABORATORY PROGRESS REPORT FOR JULY 1964. J. F. Eichelberger, G. R. Grove, and L. V. Jones (Mound Lab., Miamisburg, Ohio). Sept. 25, 1964. Contract AT(33-1)-Gen-53. 18 p.

A calorimetric determination of half-life of Po-210 gives a value of  $138.3763 \pm 0.0017$  days. The half-life of Po-210 redetermined by the method of decaying pairs is  $138.7 \pm 0.4$  days.

- 3.6.5.4 (MLM-1216) MOUND LABORATORY PROGRESS REPORT FOR AUGUST 1964. J. F. Eichelberger, G. R. Grove, and L. V. Jones (Mound Lab., Miamisburg, Ohio). Aug. 20, 1964. Contract AT-33-1-GEN-53. 45p.

Six sources of Po-210 are being counted at irregular intervals to redetermine the half life of the isotope. The half life value was found to be  $139.3 \pm 0.1$  days, as compared with the accepted value of  $138.401 \pm 0.001$  days.

((Only the portion of report pertaining to Po-210 is reproduced.))

- 3.6.5.5 (MLM-1200) GAMMA SHIELDING REQUIREMENTS FOR PLUTONIUM-238 AND POLONIUM-210. K. W. Foster (Mound Lab., Miamisburg, Ohio). May 15, 1964. Contract AT(33-1)-Gen-53. 9p.

Gamma shielding requirements were computed for Pu-238 and Po-210. Literature values were used for the gamma energies, their relative abundances, and for the mass absorption coefficients of these decay components in various shielding materials. A point source and isotropic radiation were assumed; self-absorption was not considered. For Pu-238 the greatest shielding thicknesses were required to attenuate the 760-kev component; therefore, the shielding required to reduce the dose rate from this component to less than one mR per hour at one meter was used to compute the total dose rate. The following shielding thicknesses are required to reduce the dose rate from a bare 1-kg plutonium-238 source to less than 2.5 mR per hour at one meter: 1.95 cm lead, 3.77 cm iron, 1.03 cm uranium, and 11.6 cm concrete. The gamma radiation intensities were computed for 100-watt, 1-kilowatt, and 10-kilowatt Po-210 heat sources. The thicknesses of uranium required to attenuate the 0.803-Mev gamma radiation from each of the above sources to 1000, 100, 10.0, and 1.0 mR per hour at one meter were calculated (e.g., 0.95 cm uranium would reduce the dose rate of 1000-watt source (54.4 mR) to 10.0 mR per hour at one meter).

- 3.6.5.6 Refer to 3.10.12. (RSG-A-1) PRELIMINARY REPORT ON PUBLIC HEALTH HAZARD ANALYSES OF SPACE ISOTOPIC POWER SOURCES. J. J. Fitzgerald, Gordon L. Brownell, and N. Irving Sax - Jack J. Gabay, ed. (New York. State Dept. of Health, Albany). Mar. 1, 1964. Contract SAPH 76262. 87p.
- 3.6.5.7 RADIOISOTOPE POWER SYSTEMS FOR MANNED SPACE STATIONS. Alan J. Streb (Martin Co., Baltimore). New York, American Inst. of Aeronautics and Astronautics, 1964. Paper No. 64-711, 31p. (CONF-640901-3)

From 3rd Biennial Aerospace Power Systems Conference, Philadelphia, Sept. 1964.

Radioisotope power systems in the range of three to four kilowatts are considered for application to manned orbiting space stations. Isotope selection and availability, power conversion systems, and safety are considered for present station concepts. Systems using

Po-210 or Pu-238 and employing the Brayton cycle are concluded to offer a number of advantages over the conventional photovoltaic system in this power range.

- 3.6.5.8      POODLE - A DIRECT CYCLE RADIOISOTOPE-HEATED ROCKET ENGINE. J. S. Martinez and D. Jortner (TRW Space Technology Labs., Redondo Beach, Calif.). New York, American Inst. of Aeronautics and Astronautics, 1964, Paper No. 64-383, 6p. (CONF-600-35)

From American Institute of Aeronautics and Astronautics 1st Annual Meeting, Washington, D. C., June-July 1964.

The POODLE rocket engine uses isotopic energy to heat hydrogen which is expelled through a conventional nozzle. The results of a survey are summarized, showing that Po-210 is the most promising isotope for this use, and engine design considerations are discussed. A hydrogen-fueled, POODLE-powered stage for use with Titan II is described, and payload characteristics of chemical-POODLE and all-chemical systems are compared.

- 3.6.5.9      ALPHA SOURCES FOR LOW THRUST TASKS IN SPACE. W. Diethorn (Nuclear Engineering Department, Pennsylvania State University, Pennsylvania). International Journal of Applied Radiation and Isotopes 15, 127-31, March 1964

The recoil thrust exerted on a flat plate alpha source is calculated for alpha particle escape from one side of the source. The thrust is low even for the high specific activity alpha emitters Po-210, Pu-238 and Cm-244 now receiving increased production emphasis. It is suggested that these three alpha emitters may find some future applications in space exploration requiring low thrust devices.

((This article presents a theoretical discussion on the maximum thrust capabilities of the subject alpha emitters. Information is also presented in Table 1, not reproduced here, of the chemical and physical properties of the isotopes. The information presented in the table is taken from several other articles.))

- 3.6.5.10     THE MECHANISM OF ALPHA-DECAY, AND THE REDUCED LEVEL WIDTHS OF Po-210 AND Po-212. K. B. Baktybaev and B. M. Bukat. Izv. Akad. Nauk SSSR, Ser. Fiz., 27:1297-1304 (Oct 1963)

The simple model of the penetration of a potential barrier by an alpha particle can be modified by introducing the reduced width  $\gamma_L$ ; whose square characterizes the probability for formation of an alpha group within the nucleus:  $\gamma_L = (2\lambda_L/h)P_L$ . Here  $\lambda$  is the decay constant for the emission of an alpha particle with a definite orbital moment  $L$ , and  $P_L$  is the barrier penetrability. The value of  $\gamma_L$  is determined by the behavior of the internal wave function of the nucleus, and the probability of alpha decay can be related to the nuclear structure. The reduced widths for a decay are calculated for Po-210 and Po-212 by taking into account the role of correlations associated with residual pair interactions in the nucleus. A number of alpha transitions to excited states are strongly forbidden due to the low probability of a particle formation in the nucleus. Values of  $\gamma_{\text{pure}}^2$ , calculated on the assumption that the nuclear states are pure shell states, are compared with values of  $\gamma_{\text{mixed}}^2$ , where the residual pair interactions lead to mixed configurations. It is shown that the probability of alpha decay in Po-210 and Po-212 is strongly dependent on tunneling effect.

3. 6. 5. 11 MEASUREMENT OF THE LEAD M PHOTON INTENSITY IN Po-210 DECAY; A FURTHER GROSS INADEQUACY OF THE THEORY. William Rubinson (Department of Chemistry, Brookhaven National Laboratory, Upton, New York). Physical Review 130, 2011-202, June 1963. ((See also BNL-6166 in this section. The present article is by and large an updated version of that report.))

The probability that a Pb M photon is emitted in Po-210 decay is  $0.91 (\pm 15\%) \times 10^{-3}$  photons per alpha as measured by means of a proportional counter and pulse height analyzer. This value is in gross disagreement with theoretical estimates. A Pb M spectrum from Po-210 decay shows definite differences from a conventional Pb M spectrum, indicating considerable injection of higher shell electrons in the Po-210 decay act. In particular the spectrum from Po-210 contains a 3.7 keV component whose intensity is about 4% of the total spectral intensity and whose energy is consistent with the allowed but hitherto unreported transition  $M_{2,3}$ . The theory of electron ejection in alpha decay is reviewed and previous calculations of ejection probabilities are extended to include ejection to bound states. The estimates of K, L, and M photon yields in this theory are orders of magnitude lower than the measured yields. Possible sources of the discrepancy are discussed.



About 30 millicuries of Po-210 were obtained from the Monsanto Chemical Company as an invisible quantity of the dry nitrate. The supplier stated it to assay at 99.9% Po-210. Possible impurities being Bi and Pt. Without further purification the Po-210 was taken up in three milliliters of a 2.4 normal nitric acid solution that had been prepared with triply distilled water. This constituted our Po-210 stock solution which assayed by the method of dilution and alpha counting proved to have a specific activity of  $1.956 \times 10^{10} \pm 2\%$  disintegrations per minute per milliliter 8.81 millicuries per milliliter, a value of about 1% higher than that computed from the supplier's states assay. In order to test for possible effects of source thickness, sources of strength ranging over a factor of 10 were prepared namely with aliquot volumes 5, 10, 25, and 50 lambda (0.044 to 0.44 millicuries) for each of these strengths at least two sources were made. The measured X-ray intensities proved to be independent of source strengths over this range.

- 3.6.5.12 Refer to 3.4.5.10. EVALUATION OF ISOTOPE THERMIONIC GENERATORS. Robert Harvey, et. al. Transactions of the American Nuclear Society 6, 88-91, June 1963
- 3.6.5.13 RADIOAKTIVNYE EMANATSCH. (Radioactive Emanation). V. M. Permyakov, Moscow and Leningrad, Publishing House of the Academy of Sciences, 1963. 176p.

The physical and chemical properties of Rn-222, Rn-220, and Rn-219 are discussed. These include decay spectra, half life, absorption, diffusion, chemical effects, units of activity and concentration, and other properties. The properties and activity of Po-218, Pb-214, Bi-214, Po-210, Bi-210, Pb-210, Po-215, Pb-211, Bi-211, Po-216, Pb-212, Bi-212, Po-212, and Tl-208 are also briefly discussed. A bibliography of 442 references is included.

- 3.6.5.14 LIFETIMES OF THE 246 and 46.7 KEV TRANSITIONS IN POLONIUM-210. E. G. Funk, Jr., H. J. Prask, F. Schima, J. McNulty, and J. W. Mihelich (University of Notre Dame, Notre Dame, Indiana). Physical Review 129, 757-9, January 1963

The half lives of the 1431 and 1478 kev levels in Po-210 have been measured. The experimental values being  $1.8 \pm 0.2$  nsec and  $29 \pm 6$  nsec, respectively. These levels are depopulated by E2 transitions

of 246 and 46.7 keV whose transition probabilities are approximately three times greater than the single part nickel estimates. One of the most sensitive tests of nuclear models is the determination of gamma ray transition life times. We have undertaken an extensive study of the levels of Po-210 as populated by the electron capture decay of 8.3 hour At-210. This nucleus is of particular interest since it has a closed neutron shell ( $n = 126$ ) and 2 protons outside of the closed proton shell at  $Z = 82$ .

- 3.6.5.15 ABSOLUTE PRECISION DETERMINATION OF SEVERAL RESONANCE THRESHOLD ENERGIES AND THE ALPHA PARTICLE ENERGY OF Po-210, PART II. A. Rytz, H. H. Staub, H. Winkler and W. Zych (Physik Institut der Universtitat, Zurich). *Helv. Phys. Acta* 35, 341-50, 1962

In Part I of this paper\*, we described absolute measurements of the thresholds of the reactions Li-7 (p, n) Be-7 and T (p, n) He-3 and the alpha particle energy of Po-210 with a semicircular magnetic spectrometer. The same apparatus and method has been used for the remeasuring absolute energy values of the reactions A-127 (p, gamma) Si-28 at 992 keV and F-19 (p, gamma) O-16 at 872 keV both of which were used as calibration standards. ((The balance of this article does not contain any new information on the alpha particle energy of Po-210. The article is referenced here primarily to provide continuity with Part I of this work.))

\**Helv. Phys. Acta* 34, 960-76, 1961

- 3.6.5.16 COMPARISON OF ALPHA PARTICLE ENERGIES FROM Po-210 AND Po-214 AND THE ENERGY OF Po-210 ALPHA PARTICLES. Cornelius B. Brown (University of Notre Dame, Notre Dame, Indiana). *Physical Review* 124, 1139-42, May 1962

The energies of alpha particles emitted by Po-210 and Po-214 were compared with a broad range spectrograph. Techniques were the same as those used in recent comparison of the Po-210 alpha particle energy with the Li-7 (p, n) Be-7 reaction threshold energy. The result agrees with other measurements and thus the discrepancy between Po-210 alpha particle energies measured against the Briggs value for Po-214 and the Po-210 alpha particle energy measured absolutely or against the Li-7 (p, n) Be-7 threshold remains. If Rytz's recent absolute value for the Po-210 energy is used\*, the

discrepancy disappears. A summary of all measurements show that the energy of alpha particles from Po-210 is very close to 5.3045 Mev.

\*Ref. 3.5.8. Helv. Phys. Acta 34, 960-76, 1961

- 3.6.5.17 MODERN PROBLEMS OF NUCLEAR SPECTROSCOPY AND THE HUNGARIAN RESULTS. Denes Berenyi (Nuclear Research Inst., Hungarian Academy of Sciences, Debrecen). Magy. Fiz. Folyoirat, 10: 515-26 (1962) (In Hungarian).

The alpha-spectrum of Po-210 was studied by an alpha-gamma coincidence technique, it was found that the energy of the low-intensity group of that spectrum is  $4.525 \pm 0.005$  Mev.

- 3.6.5.18 MASS NUMBER ASSIGNMENTS AND ALPHA ACTIVITIES OF LIGHT POLONIUM ISOTOPES. Wilhelm Forsling and Torsten Alvater, Arkiv fur Physik 19, 353-68, 1961

Light polonium isotopes have been produced mainly by utilizing heavy ion bombardments. Mass number assignments of these nuclides have been made by use of an electromagnetic isotope separator. The alpha activities of Po-198, 199, 200, 201, 202, 203, 204, 205, 206, 208, and 210 were studied in this work.

The samples of Po-210 used in the present experiments were obtained from small glass tubes which had been filled with radon and used in therapy at the Institute of Radio Physics in Stockholm. As is well known, Rn-222 decays to Po-210 which is found in the deposit on the walls of such tubes. After about three years the Po-210 activity had reached its equilibrium. The active deposit was extracted from the radon bulbs ((and)) the Po-210 samples were then made by electrochemical replacement onto silver foils in 0.4 M nitric acid. ((As a result of the work described in this article, the alpha particle energy of Po-210 is determined to be  $5.30 \pm 0.02$  Mev.))

- 3.6.5.19 ABSOLUTE PRECISION DETERMINATION OF SEVERAL RESONANCE THRESHOLD ENERGIES AND THE ALPHA PARTICLE ENERGY OF Po-210, PART I. A. Rytz, H. H. Staub, and H. Winkler (Physik Institut der Universitat, Zurich). Helv. Phys. Acta 34, 960-76, 1961

Po-210 is still used extensively as an energy standard despite its well known detrimental properties. It therefore seems desirable to make a new absolute measurement of this energy value with our spectrometer. The quality of every Po-210 alpha measurement depends to a high degree on the source properties. The most successful procedure seems to consist in deposition by volatilization in vacuum on a polished tantalum surface. Our source has been prepared by the Laboratoire de l'Amimant Permanant at Orsay, France, where this technique has been developed by R. J. Walen. The polonium had been deposited on only one narrow side ( $8 \times 0.3 \text{ mm}^2$ ) of a piece of tantalum  $10 \times 8 \times 0.3 \text{ mm}$  which had been polished previously. ((As a result of the work described in this article, the Po-210 alpha particle energy is determined to be  $5304.93 \pm 0.60 \text{ kev}$ .) )

- 3.6.5.20 COMPARISON OF ALPHA PARTICLE ENERGIES FROM VARIOUS POLONIUM-210 SOURCES. C. P. Brown (University of Notre Dame, Notre Dame, Indiana) and T. A. Eastwood (Atomic Energy of Canada Limited, Chalk River, Canada). Physical Review 124, 1494-6, December 1961

A comparison of the energies of alpha particles from six different Po-210 sources was made with the same broad range spectrograph used for a recent comparison of the Po-210 alpha particle energy with the Li-7 (p, n) Be-7 threshold energy. The sources were prepared by different techniques and some were on flat backings whereas others were on cylindrical backings. It is concluded that the discrepancies in most recent measurements of the alpha energy does not arise from source preparation techniques but the lower values of the older measurements may have been caused by source aging. ((This report was part of a project whose goal was to eliminate the variable of source condition. The energy of alpha particles from source made by one person by one technique were measured in dfiferent laboratories and compared with results obtained using sources made by the various techniques used in those laboratories.))

- 3.6.5.21 ALPHA PARTICLE REDUCED WIDTHS IN HEAVY NUCLEI. Kichinosuke Harada (Nuclear Physics Department, Japan Atomic Energy Research Institute, Tokai-mura, Ibaraki-ken). Progress in Theoretical Physics 26, 5, 667-79, November 1961

The alpha particle reduced widths ( $R\alpha^2$ ) for the ground state in Po-210 and Po-212 are calculated on the basis of the nuclear shell model. The calculations are made taking the boundary condition in an approximate way into consideration. The effects of the consideration mixing of the parent and daughter nucleus wave functions on  $\gamma_{\alpha}^2$  are examined and it is found that they give quite large contributions to  $\gamma_{\alpha}^2$ . Some features of the distortion of the two nucleon wave functions arising from the configuration mixing are discussed graphically. It is shown that the wave function of relative motion with the mixing of the level which is lowered from the upper band by spin orbit force would correspond to the bound electron pair in the superconducting metals. Although there are some unavoidable uncertainties in the course of the calculation it is concluded that wave functions derived by conventional shell model calculations can explain the major part of the experimental values of  $\gamma_{\alpha}^2$ . ((This article is entirely theoretical in nature.))

3. 6. 5. 22 STUDY OF THE AUGER L ELECTRONS FOLLOWING THE AUTOIONIZATION PHENOMENON IN Po. A. Juillard and A. Moussa (centre d'Etudes Nucleaires, Grenoble, France). J. Phys. Radium 22: 677-9 (Oct. 1961) (In French)

A study is carried out of the rearrangement Auger L electrons following the autoionization process after disintegration of Po-210. Experimental results are between  $1.3 \times 10^{-3}$  to  $2.5 \times 10^{-3}$  Auger L electrons per disintegration. These results are compared with measurements of rearrangement X-K and X-L photons and with the theoretical value.

3. 6. 5. 23 ABSOLUTE MEASUREMENT OF A SET OF ENERGY CALIBRATION STANDARDS. E. H. Beckner, R. L. Bramblett and G. C. Phillips (Price University, Houston, Texas), and T. A. Eastwood (Atomic Energy of Canada Limited, Chalk River, Ontario, Canada). Physical Review 123, 2100, September 1961

A 180° magnetic spectrometer has been employed to measure the energy of several neutron thresholds and gamma ray resonances as well as the energy of the alpha particles emitted by Po-210. The primary reason for performing these experiments was to obtain a set of energy standards with consistent experimental techniques for all the measurements. The neutron thresholds studied were Li-7, (p, n) Be-7; B-11 (p, n) C-11; C-13 (p, n) N-13; and F-19 (p, n) Ne-19.

The gamma ray residences at 872 kev in F-19 (p, alpha-gamma) 0-16 and 992 kev in Al-27 (p, gamma) Si-28 were observed. The same instrument used to make energy measurements in these experiments was also employed to determine the energy of the alpha particles emitted by Po-210.

The sources used for these experiments were prepared by currentless electrodeposition on pure silver foils using two slightly different techniques. Nine sources were prepared by one of the authors by dipping the source backings into the polonium solution for periods of from 2 to 10 minutes. These sources were used immediately after preparation in order to avoid the familiar problem of polonium diffusion into the backing. Several other sources were prepared by the second author by a somewhat different technique. These latter sources were prepared by placing a small amount of the polonium solution directly on the surface of the source backings. These sources were also used immediately after their preparation. In no case were sources more than 6 hours old employed in these measurements. Sources of various activities were employed by using the source preparation time to range from 2 to 20 minutes.

3. 6. 5. 24 GAMMA RADIATION FROM Pb-206. D. E. Alburger, and G. Friedlander (Brookhaven National Laboratory, Upton, New York). Physical Review 81, 523-6, February 1961

Gamma rays accompanying the electron capture decay of 6. 4 day Bi-406 and the alpha decay of 138 day Po-210 have been observed by measurement of internal conversion electrons and photoelectrons with a lens spectrometer. The bismuth activity showed the resolved lines corresponding to the gamma rays of 182, 234, 260, 341, 470, 505, 536, 590, 803, 880, 889, 1020, 1097, and 1720 kev. A thin Po-210 source of 480 mc alpha particle strength was found to emit gamma rays of  $800 \pm 6$  kev energy which yielded K and L internal conversion lines in the ratio of  $3.7 \pm 0.5$  to 1. The 803 kev gamma ray observed in both the Bi-206 and Po-210 activities is identified with a transition from a first excited state of Pb-206 to the ground state.

The Po-210 source was specifically prepared for this work by the AEC. It consisted of carrier free polonium deposited on a nickel disc over an area 0. 5 centimeter in diameter and covered with 0. 2 mil gold foil (about 10 mg/cm<sup>2</sup>) for the purpose of reducing the spread of contamination. The foil had been clamped with a metal ring and sealed at

the outer edges with a vacuum evaporated coating. At the time of fabrication, the alpha particle strength was 478 millicuries and from this the thickness of the source is estimated to be about 0.5 mg/cm<sup>2</sup>.

- 3.6.5.25 BNL-6166. THE LEAD X-RAYS THAT ACCOMPANY POLONIUM-120 DECAY. MEASUREMENT OF THE M PHOTON YIELD AND THEORY. William Robinson, Department of Chemistry, Brookhaven National Laboratory, Upton, Long Island, New York. 1960. 46 pgs.

The probability that the lead M photon is emitted in the alpha decay of Po-210 atom is  $0.91 \pm 15\% \times 10^{-3}$  photons per alpha, as measured by means of a proportional counter and pulse height analyzer. The value is in gross disagreement with theoretical estimates. A lead M spectrum from Po-210 decay shows definite differences from a conventional spectrum, indicating extensive ejection of the outermost electrons in the decay act. In particular, the spectrum from Po-210 contains a 3.7 kev line whose intensity is about 4% of the total spectral intensity, and whose energy is consistent with the allowed but hitherto unreported transition  $M_1O_{2,3}$ .

The theory of electron ejection in alpha decay is reworked and the results are compared with the measured values of K, L, and M ejections in Po-210 decay. It is concluded that the apparent agreement between measured probability of K ejection and the theoretical calculations of Midgal and of Schaefer is fortuitous and that for K as well as for L ejections there is an order of magnitude difference between theory and experiment. And too, the failure of the theory can be ascribed in a large part to the unjustifiable set of hydrogenic wave functions.

- 3.6.5.26 MEASUREMENT OF THE ENERGY OF THE WEAK GROUP AND THE ALPHA SPECTRA OF Po-210. T. Fenyés (Institute for Experimental Physics, Debrecen, Hungary). Nuclear Physics 16, 529-533, 1960

The nuclear spectrum of Po-210 was examined with a scintillation alpha-gamma coincidence equipment combined with an electrostatic alpha spectrometer accepting  $5.3054 \pm 0.0010$  Mev (absolute  $\nu$ ) as the value for the kinetic energy of the main group, that of the weak group was found to be  $4.525 \pm 0.005$  Mev. As is well known Po-210 emits mostly alpha particles of 5.3 Mev kinetic energy but it has also some gamma radiation of very low intensity. The gamma spectrum has

been investigated by a number of authors. The measurements have revealed that the gamma radiation of Po-210 consists of a single line of about 800 kev. No other lines have been observed, at any rate, not in the interval between 25 kev and 2.5 Mev. The aim of this investigation was the precise determination of the energy ratio between the weak and main alpha groups through direct alpha spectroscopy. The Po-210 source was prepared ((by the method of)) volatilization and concentration of polonium in a hydrogen stream.

- 3.6.5.27 COMPARISON OF Po-210 ALPHA PARTICLE ENERGY WITH THE Li-7 (p, n) Be-7 REACTION THRESHOLD ENERGY. C. P. Browne, J. A. Galey, J. R. Erskine and K. L. Walsh (Department of Physics, University of Notre Dame, Notre Dame, Indiana). Physical Review 120, 905-13, November 1960.

Recent absolute measurements of the Po-210 alpha particle energy disagree with the older value used as the standard for many nuclear reaction energy measurements. A new comparison with the Li-7 (p, n) Be-7 reaction threshold energy was made using the Notre Dame electrostatic accelerator and broad range spectrograph. Four separate methods of comparison were used. In the first three, the threshold was run and then protons or deuterons were scattered from appropriate targets so that the scattered group was recorded on the spectrograph plate near the alpha group from a source placed at the target position. First the spectrograph, and second the beam analyzer, were used to compare particle momenta. Third, with both fields held constant after the threshold was run with the molecular beam, deuterons were scattered giving particles of the same  $B_p$  as the alphas. In the fourth method several reaction energies that were precisely known in terms of the Li-7 (p, n) Be-7 reaction threshold energy were measured in terms of the Po-210 alpha particle energy. These were the Mg-24 (d, d') Mg-24\* reaction to the first excited state of Mg-24 and the N-14 (d, p) N-15 reaction leading to three excited states of N-15. The four measurements agree and give  $5.3086 \pm 0.003$  Mev for Po-210 alpha particle energy based on 1.8811 Mev for the Li-7 (p, n) Be-7 reaction threshold energy.

- 3.6.5.28 ABSOLUTE MEASUREMENT OF SOME ALPHA ENERGIES. Albrecht Rytz. Compt.rend. 250, 3156-8 (1960) May 9 (In French)



The absolute energies of the radiation of Po-210, Po-212, Po-214, Bi-211, and Bi-212 were determined by magnetic spectrography with field controlled by nuclear resonance and absolute measurement of the trajectory lengths.

3. 6. 5. 29    GAMMA RAYS FROM A POLONIUM OXIDE 18 NEUTRON SOURCE.    E. M. Tsenter, A. G. Khabakhpashev and I. A. Pirkin, J. Exptl. Theoret. Phys. 37, 1133-1134, October 1959 ((translated in Soviet Physics J. E. Tepee 37, 4 April 1960

In a previous paper it has been shown that a Po O neutron source the (alpha, n) reaction operates on the isotope O-18. The neutrons yielded by the reaction O-18 (alpha, n) Ne-21 are accompanied by 0.35 Mev gamma rays with a relative intensity of  $30 \pm 10\%$ . The Po O-18 spectrum contains a certain number of pulses with energies up to 2.8 Mev. Some of these are evidently due to neutrons registered by the sodium iodide crystals. Others are possibly due to hard gamma rays which could not be detected in the course of the measurements because of low intensity. The intensity of the 0.35 Mev and 1.38 Mev gamma lines were determined from the areas under the full energy peaks. The crystal counting efficiency in a ratio of the area under a photopeak to that under the entire spectral curve was taken from other references. The intensity of the 0.35 Mev line relative to the neutron yield was found to be  $45 \pm 5\%$ . The intensity of the 1.38 Mev line was  $10 \pm 2$ . Upper limits for the relative intensity for the 1.73 Mev gamma line and the 2.84 Mev line were determined from the complete gamma spectrum of the Po-O-18 source. The upper limit was 1% for the 1.73 Mev line and 2% for the 2.84 Mev line.

3. 6. 5. 30    ON THE NUCLEAR SHELL STRUCTURE THEORY.  
Ming Yu (Inst. of Nuclear Energy, Academy of Sciences).  
Wu Li Hsueh Pao 15, 420-39 (1959) August (In Chinese

The problems of nucleon-nucleon interactions outside saturated shells are analyzed by perturbation methods. The method of wave expansion was developed for calculating Pb-206, Po-210, and Bi-210 energy spectra. The results are in good agreement with experimental data. It was shown that nuclear energy spectra are determined by S-wave interactions. The interaction intensity is in agreement with the effective radius of nuclear force action and scattering path and does not depend on the shape of the nuclear force. In special cases P-wave interactions play an important part. The analysis showed that excluding the S-wave interactions the repulsive force of  $^3P_0$  wave interactions is the strongest force between two nucleons.

3. 6. 5. 31 POLONIUM-210 AS A STANDARD FOR GAMMA RADIATION. Genevieve Bastin-Schoffier and Robert J. Walen, Compt. rend, 247, 2333-5 (1958) Dec. 22 (In French)

The alpha magnetic structure of Po-210 was studied to determine the branching ratio in the fine structure. The ratio of the weak alpha to the strong alpha emission was determined, and the gamma/alpha ratio was found to be  $1.006 \times 10^{-5}$ .

3. 6. 5. 32 THE EMISSION OF LONG RANGE ALPHA PARTICLES WITH  $E_{\alpha}$  OF 10 MEV BY Pu-239 AND Po-210. V. N. Andreev and S. M Sirotkin. Isv. Akad. Nauk SSSR, Ser. Fiz. 27: 1250-2 (Oct. 1958)

Ader in the J. Phys. et Radium 15:60, 191 (1954) and 17:45 (1956) observed the emission of  $10^{-6}$  to  $10^{-8}$  long range alpha particles for each alpha decay. These alpha particles have a range of 100 to 300 u in photoemulsions (12 to 25 Mev), and are thought to be due to spontaneous or forced triple fission. A specially constructed ionization chamber was set up to count the long range alpha particles in the presence of ordinary alpha radiation. A 10 mg sample of Pu-239 was deposited on an aluminum disc having a density of 0.5 mg/cm<sup>2</sup>, and was covered with an aluminum foil 28 u thick. The 70 ug sample of Po-210 was electrodeposited on a nickel plate, and was covered with a tinfoil 15.3 u thick and with two aluminum foils having a total thickness of 14.4 u in order to avoid the interference from an (a, p) reaction on the aluminum. The total count rates were found to be  $0.55 \pm 0.09$  counts/hr for the Pu-239 sample, and  $1.0 \pm 0.1$  counts/hr for the Po-210 sample for 10 to 15 Mev alpha particles. It was concluded that Pu-239 emits  $10^{-10}$  long range alpha particles per alpha decay, and Po-210 emits  $7 \times 10^{-13}$  long range alpha particles per alpha decay at the 95% confidence level.

3. 6. 5. 33 ENERGY LEVELS OF POLONIUM-210. R. W. Hoff and J. M. Hollander (University of California Radiation Laboratory, Livermore and Berkeley, California). Physical Review 109, 447-56, January 1958

The energy levels of Po-210 have been studied as populated by the electron capturing decay of At-210 and experimental level scheme has been constructed by using data obtained by conversion electron and gamma ray measurements made with beta ray and scintillation spec-

trometers and coincidence counting techniques. A theory of the energy levels of Po-210 has been developed using the method of Pryce to predict the levels of a nucleus containing two odd protons beyond the double closed shell from the experimentally known levels of Bi-209, the nucleus containing a single odd proton beyond the closed shell. Certain features of the theoretically predicted level scheme and the experimental level scheme show reasonable agreement. The spin assignment for At-210 has been discussed with respect to the log ft values for its electron capture decay. ((Figure 4 of this article presents an experimental Po-210 level scheme (energies in kev.)))

3. 6. 5. 34 ABSOLUTE ENERGY MEASUREMENT OF ALPHA PARTICLES FROM Po-210. F. A. White, F. M. Rourke, J. C. Sheffield and R. P. Schuman (Knolls Atomic Power Laboratory, Schenectady, New York) and J. R. Huizenga (Argonne National Laboratory, Lemont, Illinois). Physical Review 109, 437-422, January 1958

The absolute alpha energy of Po-210 alpha particles was measured by a 180° magnetic spectrometer. The value obtained was  $5.3054 \pm 0.0010$  Mev (absolute volts). Measurement of the energy ( $\epsilon_0$ ) of the alpha particles from Cm-214 leading to the ground state of Pu-240 was made relative to the energy of the Po-210 alpha particles. A value of  $5.8025 \pm 0.002$  Mev was obtained for this Cm-244 energy. The energy difference between the ground state transition  $\epsilon_0$  and the transition  $\epsilon_1$ , to the first excited level of Pu-240 from Cm-24 was found to be  $43.5 \pm 1$  kev.

The polonium sample was prepared from chemically purified polonium purchased from the Mound Laboratories. No further purification of the polonium was made. The sample used for the energy determination was plated on polished platinum. First a very thin film of copper was electroplated on the platinum; then the polonium was deposited on the platinum from a chloride solution. The final sample showed no discoloration due to either impurities or residual copper.

The curium sample was produced by the very long term irradiation of plutonium in the Materials Testing Reactor. The curium was separated from other actinides and purified from fission products by a series of cation and anion ion exchange column processes. The final curium contained about 98% Cm-244 alpha activity and 2% Cm-242 alpha activity. Just before the source was prepared, plutonium daughter activity was separated from the curium by an anion exchange column process.

The curium sources were made by vacuum-subliming the curium from a tantalum filament through a defining slit onto the polished platinum source holder. The sources showed a slight discoloration of the platinum due either to sublimed tantalum or to solid impurities deposited on the filament with the curium.

3. 6. 5. 35 ON THE RELATIVE INTENSITY OF Po-210 GAMMA RAYS.  
E. E. Ovechkin, Izvestia Akad Nauk SSSR 21, 12, 1641-2  
December 1957

Experimental evaluation of the relative intensity of the 803 kev gamma rays emitted from the first excited level of Pb-206 in alpha decay of Po-210 is of undoubted interest. In view of the divergence in the data published prior to 1952, we undertook a repeat measurement of the ratio  $K_{\text{gamma}} = n_{\text{gamma}}/n_{\text{alpha}}$ . The polonium source was prepared by precipitation from a Po-210 nitrate solution after repeated purification to remove all extraneous impurities. The source was sealed in a brass tube 8 millimeters long and 5 millimeters in diameter with a wall thickness of 0.5 millimeters, and had an alpha activity  $n_{\text{Po}} = 4.10 \pm 0.5$  microcuries according to a calorimetric determination. ((As a result of the work described in this article, the  $K_{\gamma}$  ratio is set equal to  $(1.22 \pm 0.09) \times 10^{-5}$  quanta per decay.))

3. 6. 5. 36 ENERGY OF ALPHA PARTICLES FROM Po-210. I. I. Agatkin and L. L. Gol'din, Izvestia Akad Nauk SSSR 21, 909-12, July 1957

The sources ((used in the experiment described in this article)) were prepared by vacuum evaporation of the radioactive materials Po-210 and Ra-224 from a heated filament onto a glass support. We suggest ((from the results of the experimentation described in the article)) the value of  $5297.8 \pm 1.5$  kev for the Po-210 alpha particle energy.

3. 6. 5. 37 DETERMINATION OF THE BRANCHING RATIO IN THE DISINTEGRATION SCHEME OF POLONIUM-210. N. S. Shimanskaia (Radium Institute, Academy of Sciences, USSR). J. Exptl. Theoret. Phys. (USSR) 31, 174-177, August 1956 (translated in Soviet Physics JETP, Vol. 4, #2, 165, March 1957)

Measurements were made of the branching ratio in the disintegration scheme of Po-210. The probability of transitions to the excited level Pb-206 (with an energy of  $E = 800$  kev) was found to be  $1.2 \pm 0.2 \times 10^{-5}$ .

The source used by us was a preparation of pure Po-210. Absence of any noticeable radioactive impurities was confirmed by long time (8 months duration) calorimetric measurements of its disintegration curve as well as of the absorption curve of its gamma radiation in lead. The decrease of the activity of the source also corresponded to the half life of Po-210 (138.5 days). The values of the absolute number of gamma transitions in Po-210 and therefore the values of the branching ratio in the disintegration scheme of this isotope obtained by us were considerably less than those reported in ((3 previous investigations)). Since the usual ionization or impulse chamber methods were used by these investigators to determine the activity of the polonium source, it was possible that some of the alpha particles were not registered as mentioned above due to the diffusion of the active atoms into the backing material and absorption into the surface films. This could result in raising the value obtained for the absolute intensity of nuclear radiation from Po-210.

- 3.6.5.38 ON THE RATIO BETWEEN GAMMA AND ALPHA ACTIVITIES IN Po-210. A. Ascoli, M. Asdente and E. Germagnoli (Laboratory C.I.S.E. Milano). Nuovo Cimento 10, 4, 946-7, October 1956

Po-210 is known to be a practically pure alpha emitter. Its decay scheme is given in Figure 1 and its half life is 138.13 days. A direct measurement of the intensity of emitted alpha particles may be carried out with satisfactory accuracy if the source is very thin and weak enough to be introduced into an ionization chamber of well defined geometry. If the intensity of the source is remarkable its absolute calibration by means of alpha counting becomes more troublesome and a current chamber intrinsically capable of less accuracy is generally used. For the present measurement a thin source whose activity is approximately four millicuries has been used. It has been supplied by the Radio Chemical Center of Amersham, England. Po-210 is electroplated onto platinum and covered with a thin mica sheet.

- 3.6.5.39 FINE STRUCTURE AND ANGULAR CORRELATION IN POLONIUM-210. S. DeBenedetti and G. H. Minton (Carnegie Institute of Technology, Pittsburgh, Pennsylvania) Physical Review 85, 944-5, 1955

Polonium is usually considered as a pure alpha emitter although the presence of weak gamma radiation (energy 803 kev, intensity about  $10^{-5}/\alpha$ ) is definitely established. The low energy alpha particles preceding these gamma rays have escaped observation despite repeated

attempts to study the fine structure of polonium. By using a coincidence method we were able to detect the low energy alpha group and to study its angular correlation relative to the gamma rays following it. The number of coincidences were also studied as a function of artificial delays introduced in the coincidence selector. The result indicated that the half life of the gamma emitting state of Pb-206 is less than  $10^{-9}$  seconds.

3. 6. 5. 40 THE RELATIVE NUMBER OF GAMMA RAYS FROM POLONIUM-210. Onofre Rojo, M. A. Hakeem and Max Goodrich (Louisiana State University). Physical Review 99, 1629, 1955. ((The following is the entire article. It is not an abstract.))

It has long been known that the alpha decay of Po-210 is accompanied by a weak gamma ray of about 800 kev. The number of these gamma rays has been reported to be from  $1.5 \times 10^{-5}$  to  $1.8 \times 10^{-5}$  times the number of alpha rays. In the course of some work on Po-210, the authors have redetermined this ratio using scintillation spectrometer techniques. The alpha ray measurements were made with a plastic scintillator using a diaphragm while the gamma ray measurements were made with a large ((3 inch diameter by 3 inch long)) NaI crystal. The resulting ratio of  $1.2 \times 10^{-5}$  for  $N_{\text{gamma}}/N_{\text{alpha}}$  (estimated error 10%) is believed to be significantly below the published values.

3. 6. 5. 41 EXPERIMENTAL STUDY OF INTERNAL IONIZATION IN EMISSION. Michel Riou (Institute du Radium, Paris). J. Phys. Radium 16, 583-8 (1955) July (In French)

A review is given of the results on disintegration of Po-210, the most favorable example for the observation of internal ionization. Intensities of K and L rays of lead and of the gamma ray of 800-kev emitted by Po-210 were determined. A deduction of the experimental probabilities of internal ionization of K and L shells, comparison with theory, and results on the electrons emitted by Po-210 are given.

3. 6. 5. 42 BRANCHING RATIO IN THE DECAY OF POLONIUM-210. R. W. Hayward, D. D. Hoppes and W. B. Mann, National Bureau of Standards Bulletin 54, 1, 47-50, January 1955

Using a microcalorimeter to determine the alpha particle activity and a sodium iodide scintillation counter of high efficiency to measure the gamma ray intensity, the branching ratio of Po-210 has been found to

be equal to  $(1.22 \pm 0.060) \times 10^{-5}$ . This value is based on the assumption that the energy of the main alpha particle group from Po-210 is 5.301 Mev. In the calibration of the scintillation counter, the angular anisotropy of the gamma ray from Co-60 were found to be  $1.164 \pm 0.002$ .

((Of the two Po-210 sources prepared, the second source was used. It was prepared from some 60 millicuries of solution that had been carefully purified prior to delivery. This purification consisted of depositing Po-210 onto a silver foil from a stock solution containing approximately 130 millicuries of Po-210.)) The silver foil was then washed free from solution and dissolved in concentrated nitric acid. After dispelling most of the nitric acid by evaporation, the silver was precipitated by hydrochloric acid. The solution was filtered from the silver chloride and its acidity adjusted for deposition on a second silver foil which was again processed in the same way as the first. The filtrate from the silver chloride being the final solution containing some 68 millicuries of purified Po-210.

- 3.6.5.43    PARTICLES WITH LONG PATH EMITTED FROM  
SOURCES OF POLONIUM. Marie Ader (College de  
France, Paris). J. Phys. Radium 15, 60 (1954)  
January (In French)

The origin of particles with long path emitted from Po is uncertain. It was thought that the particles could be caused by the action of the alpha rays of Po on the H<sub>2</sub> of the air, the H<sub>2</sub> of the source support, or the humidity retained in the source support, or the humidity retained in the source. However, the 5.2-Mev alpha ray can project in a hydrogenated substance protons of only 80 u, and paths longer than 300 u, were observed. The type of source support did not change the number of long paths. It was suggested that perhaps a spontaneous emission of the nuclei of a polonium atom occurred.

- 3.6.5.44    CALORIMETRIC DETERMINATION OF THE HALF-LIFE  
OF POLONIUM-210. J. F. Eichelberger, K. C. Jordan,  
S. R. Orr, and J. R. Parks, (Mound Laboratory,  
Monsanto Chemical Company, Miamisburg, Ohio)  
Physical Review 96, 719-721, November 1954

Six determinations have been made of the half-life of polonium with four different steady-state resistance-bridge calorimeters and five different samples of polonium. These six values of the half-life

have been weighed and combined to give a grand-mean value of the half-life of  $138.4005 \pm 0.0051$  days (internal consistency) or  $\pm 0.0058$  day (external consistency).

Five different Po-210 samples were used during a period of three years. The polonium was prepared by standard methods but was sealed in containers fashioned to fit the calorimetric-research needs. The chemical purity of these samples was not accurately known. However, the precision of the half-life measurements indicated that over the period of the measurements only negligible quantities of radioactive impurities could have been present. Gamma counts and neutron counts of the samples did not indicate the presence of any radioactive impurities in measurable amounts. ((Cited as best value in Rev. Mod. Phys. Vol. 30, No. 2, Pt. II, April 1958.))

- 3.6.5.45 EXPERIMENTAL VERIFICATION OF THE THEORY OF MIGDAL: CASE OF POLONIUM (Po-210). Alphonse Lagasse and Jacqueline Doyen. Compt. rend. 239, 670-2, (1954) Sept. 13. (In French)

Electron and gamma radiations from Po-210 are found to agree with a theory due to Migdal (J. Exp. Theoret. Phys. (USSR) 11, 207 (1941)).

- 3.6.5.46 ON THE THEORY OF MIGDAL. R. R. Roy and M. L. Goes (Univ. Libre de Bruxelles, Belgium). Bull. Classe Sci. Acad. 40, 143-9 (1954) Feb. (In French)

The probability of ionization during the alpha emission of Po-210 was calculated according to the theory of Migdal and compared with experimental results. The general expressions of the energy distribution of electrons are given. A method which allows the separation of the Migdal electrons from the internal conversion electrons is indicated.

- 3.6.5.47 ON THE DECAY OF Po-210. Radha Raman Roy and Marie-Louise Goes. Compt. rend. 237, 1515-17 (1953) Dec. 9. (In French)

Experiments are described for testing a theory due to Migdal concerning the origin of soft 84-kev radiation emitted by the alpha decay of Po-210 to Pb-206. Although accurate agreement was not obtained (within an order of magnitude) between the Migdal prediction and experiment, it is concluded that the observed level is due to the ionization of the atom following alpha decay according to a Migdal process.



- 3.6.5.48 (AECD-3536; MLM-575). HALF LIFE DETERMINATION OF POLONIUM-210 BY ALPHA COUNTING. Mary Lou Curtis (Mound Laboratory, Miamisburg, Ohio). ((Also published in Physical Review 92, 1458, December 1953.))

A half life of  $138.374 \pm 0.032$  days for Po-210 was determined by alpha counting a sample of approximately 0.5 millicuries over a period of 328 days.

The sample was prepared from a solution of purified Po-210 in nitric acid onto a glass slide. Mica weighing  $0.92 \text{ mg/cm}^2$  was cemented over the sample to prevent migration of activity from the slide. Tests made by adding air to the evacuated counting chamber and counting the various air pressures showed that the sample was sufficiently thin that no counts were lost by absorption or would be lost by diffusion into the glass. Over a period of 328 days 81 measurements were made. Each measurement was of sufficient duration to total at least 500,000 counts to reduce the statistical probable error to 0.1% per measurement. No geometry factor was used since the decay could be followed from the counting rate.

- 3.6.5.49 X AND GAMMA RADIATION EMITTED BY POLONIUM-210. Michel Riou. J. Phys. Radium 13, 244 (1952) April (In French)

Between 15 and 4000 kev, Po-210 was found to emit only a single gamma ray of 800-kev energy, which is internally converted with emission of Pb K rays with an intensity of  $1.6 \pm 0.5 \times 10^{-6}$  photons/alpha particle. Pb L rays with an intensity of  $2.2 \pm 0.5 \times 10^{-4}$  photons/alpha were found to arise through alpha particle excitation of Pb present in the source, not through conversion. The internal-conversion coefficient in the K shell is  $10 \pm 3\%$ , indicating electric multiple radiation with  $2^4$  or  $2^5$  polarity.

- 3.6.5.50 FINE STRUCTURE AND ANGULAR CORRELATION IN Po-210. S. DeBenedetti and G. H. Minton (Carnegie Institute of Technology, Pittsburgh, Pennsylvania) (Received 18 January 1952). Phys. Rev. 85, 944-5, Nov. 1952

Po-210 is usually considered as a pure alpha-emitter, although the presence of a weak gamma radiation (energy 803 kev, intensity about  $10^{-5}$  per alpha) is definitely established. The low energy particles

preceding these gamma rays have escaped observation despite repeated attempts to study the fine structure of polonium.

By using a coincidence method we were able to detect the low energy alpha group and to study its angular correlation relative to the gamma rays following it.

3. 6. 5. 51    SOME OBSERVATIONS ON THE GAMMA RADIATION FROM POLONIUM.    R. W. Pringle, H. W. Taylor and S. Standil (Physics Department, University of Manitoba, Winnipeg, Canada).    Phys. Rev. 87, 384-5, July 1952

((In this work)) certain observations have been made on the radiations involved ((in the decay of Po-210)) using a coincidence scintillation spectrometer and a weak source of polonium in solution from which all traces of radium and RaD have been removed.    Single channel pulse height distribution analysis gave a value of  $804 \pm 5$  kev for the energy of the hard component.    Several standard gamma ray energies were used for purposes of calibration and the value of 804 kev is a mean for a number of experiments.    No evidence could be found to suggest the presence of any other component in the gamma radiation from polonium in the range 100 kev to 2 Mev.    An estimate has been made of the intensity of the soft component relative to the hard component from a knowledge of the relative counting rates in the apparatus and the detection efficiency of the two radiations.    On the assumption that a soft component is entirely X-radiation following internal conversion of the 804 kev gamma ray, a conversion coefficient is obtained for this radiation of 20 to 30%.

3. 6. 5. 52    EVIDENCE FOR K SHELL IONIZATION ACCOMPANYING THE ALPHA DECAY OF Po-210.    W. C. Barber and R. H. Helm (Department of Physics, Stanford University, Stanford, California).    Physical Review 86, 275-80, May 1952

The radiations of Po-210 have been studied using a NaI scintillation counter in agreement with Grace, Allen, West and Halben.    It is concluded that the soft electromagnetic component probably entirely consists of X-rays of lead.    The region from 25 kev to 2.5 Mev have been examined and with the exception of the known gamma ray of 800 kev, no nuclear gamma rays were observed.    The ratio of the number of K X-rays to the number of 800 kev gamma rays was measured as  $0.134 \pm 0.025$  to 1.    The K shell internal conversion

coefficient of the 800 kev transition has been reported as about 0.05 and hence the K X-ray intensity is too great to be explained by internal conversion alone. The residual gamma rays are attributed to the process whereby the emission of the alpha particle causes ionization of the atom. Comparison of the relative intensity of K X-rays and alpha particles shows qualitative agreement with the probability of this ionization process as calculated by Migdal.

The polonium was supplied by the Eldorado Mining and Refining Company who report radioactive impurities of unspecified form of about  $6 \times 10^{-6}$  milligrams of radium equivalent per millicurie of polonium. The sources were prepared for use and further purified by precipitation on nickel foils in 0.1 normal HCl. Later some of the polonium was purified a second time in the manner described by Lee and Libby. No spurious lines were observed in the spectra of the sources either before or after purification. The sources used for alpha counting were prepared on 0.005 inch foils thick enough to stop alphas completely so that only one side was effective for alpha counting.

3.6.5.53 INVESTIGATION OF THE GAMMA RAYS FROM  
POLONIUM. M. A. Grace, R. A. Allen, D. West, and  
H. Halban (Clarendon Laboratory, Oxford, England).  
Proceedings of the Physical Society (London) 64A, 493-  
507, May 1951

A method of standardizing polonium sources by measurement of the number of gamma rays is described. The hard gamma radiation of 0.773 Mev energy emitted in the decay of Po-210 has been found to have an intensity of  $1.8 \pm 0.14 \times 10^{-5}$  quanta per alpha particle. The internal conversion of this line to the extent of  $6.7 \pm 1.7\%$  gives rise to K X-radiation of lead. No other soft radiation is detected. The decay scheme of polonium is discussed.

We were interested in a quantitative determination of the intensity of hard gamma radiation as a means of providing a simple method of standardizing polonium. At the same time we investigated the soft radiation found by Zajac, et al. Whilst our result for the intensity of the hard radiation is in agreement with the estimate of Zajac, et al, we have found evidence that the soft radiation is the K radiation of lead. The following experiments were carried out: (1) measurement of the source strength by counting alpha particles, (2) measurement of the intensity of the hard gamma rays, (3) search for gamma rays excited by alpha particle bombardment of certain elements, (4) critical

absorption measurements on the soft radiation using a scintillation counter, (5) search for coincidences between soft and hard gamma rays, (6) critical absorption measurements on the soft gamma rays using a proportional counter, (7) intensity of electron radiation. The source was prepared by the electrolytic deposition of about 200 millicuries of Po-210 from an RaD solution on one side of a platinum foil. This foil was one centimeter square and about 30 milligrams per square centimeter thick. The Po-210 had previously been extracted twice by electrochemical deposition on silver so as to remove as much of the RaD and RaE as possible. The strength of the RaE content was found in later measurements to be less than  $7 \pm 10^{-3}$  microcuries.

((Cited as best value for decay scheme in Rev. Mod. Phys. Vol. 30, No. 2, pt. II, April 1958.))

- 3.6.5.54 NP-1642. AN INVESTIGATION OF THE ALPHA RAY SPECTRUM OF POLONIUM. John J. Wagner, 1949, 141 pages. A dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the University of Michigan.

((This document could not be located during the course of the subject program. Therefore no detailed information can be presented.))

- 3.6.5.55 THE PHYSICAL PROPERTIES OF POLONIUM. III THE HALF LIFE OF POLONIUM. William H. Beamer and William E. Easton (University of California, Los Alamos Scientific Laboratory, Los Alamos, Nevada). Journal of Chemical Physics 17, 1298-1300, 1949

The half life of polonium has been measured by a calorimetric method. The reproducibility and precision of the data is much greater than has been previously reported. The value is found to be  $138.3 \pm 0.1\%$  days.

In this investigation two samples were studied. The pure polonium was electroplated from a dilute nitric acid solution onto a platinum foil. This foil was placed inside a platinum capsule in a nitrogen atmosphere and then this was sealed off in a glass ampule after evacuation and filling with pure helium.

- 3.6.5.56 THE ALPHA RAY SPECTRUM OF POLONIUM. Walter G. Wadey (Randall Laboratory, University of Michigan, Ann Arbor, Michigan). Physical Review 74, 1846-53, 1948

Measurements of the alpha ray spectra of polonium mounted on nickel, silver, and cadmium by means of a  $180^\circ$  focusing magnetic spectrograph are reported and discussed. The weak line series reported by Chang is not apparent. The hypothesis is proposed that the alpha ray spectra seen to date have not been caused by the element polonium but by the diffusion of the polonium into the mounting metal.

The sources consisted of polonium metal deposited by the Canadian Radium and Uranium Corporation on one  $1\frac{7}{16}$ " by 0.020" edge of a strip of metal  $1\frac{7}{16}$ " by 0.020" by  $\frac{1}{8}$ ". We were informed that deposition was made spontaneously by immersion in a polonium solution relatively free from metallic ions. The plating time was approximately 6 hours for a 0.1 millicurie source. The source strengths were 0.25 millicurie, 0.1 millicurie, and 0.1 millicurie.

3. 6. 5. 57    A FURTHER STUDY OF THE GAMMA RADIATION FROM POLONIUM. B. Zajac, E. Broda and N. Feather (Department of Natural Philosophy, University of Edinburgh) Proceedings of the Physical Society (London) A60, 501-8, 1948

Absorption experiments using lead, gold and tungsten have shown that in addition to the known gamma radiation of 0.77 Mev energy, polonium emits other (soft) radiations of which the most intense has a quantum energy of  $84 \pm 4$  kev. The intensity of this radiation is of the same order of magnitude as that of the hard radiation (roughly one quantum per  $10^5$  disintegrations). Experiments by the recoil method indicate that the emission of the polonium gamma radiation is not delayed by more than  $10^{-1}$  seconds. The results of Chang, 1946, concerning the fine structure of the alpha particles of polonium remained uncorrelated with all the other experimental evidence. Whilst present information regarding gamma ray energies and intensities may be reasonably explained, satisfactory explanation of the alpha particle fine structure "data" appears as remote as ever.

The source was deposited by evaporation of an HCl solution drop by drop on a polythene disc,  $50 \text{ mg/cm}^2$  thick over an area 0.5 centimeters in diameter.

3. 6. 5. 58    THE GAMMA RAYS OF POLONIUM-210. S. DeBenedetti and E. H. Kerner (Clinton Laboratories, Oak Ridge, Tennessee). Physical Review 71, 122, 1947

In connection with Chang's studies on the fine structure of the alpha particles of polonium and with Tether's discussion on the subject, it might be useful to report some recent absorption experiments on the gamma rays of this element. Within the accuracy of the absorption method ((used in the subject experiment)) it appears that the gamma radiation of polonium consists of a single component of half thickness equal to 8.5 grams per square centimeter of polonium. The energy of the radiation can be evaluated to be 0.8 Mev. The intensity per curie of polonium is equivalent to that of one of the gamma rays from  $7.0 \times 10^{-6}$  curies of radium when both curves are extrapolated to zero absorption. A search for softer radiation showed no other components until with absorbers thinner than 0.5 grams per square centimeter, aluminum, one finds a radiation whose mass absorption coefficient in aluminum is 18 square centimeters per gram, and which is probably the same component observed by Curie and Joliot and attributed by them to the L line of polonium.

- 3.6.5.59 GAMMA RADIATION FROM POLONIUM AND FROM LITHIUM BOMBARDED WITH ALPHA PARTICLES. Kai Siegbahn (Nobelinstitutet for Fysik) Nature 159, 471-3, 1947

((The work described in this article shows that)) the gamma radiation from the polonium is monoenergetic in contradiction to the result obtained by Bothe. Bothe's value for the energy in the strongest photo line 0.798 agrees well with ours. If a gamma radiation from any other energy than 0.773 Mev is emitted from polonium, it is at any rate extremely weak in comparison with the 0.77s radiation.

- 3.6.5.60 A STUDY OF THE ALPHA PARTICLES FROM POLONIUM WITH A CYCLOTRON-MAGNET ALPHA-RAY SPECTROGRAPH. W. Y. Chang, Palmer (Physical Laboratory, Princeton University, Princeton, New Jersey) Physical Review 69, 60, 1946

A description is given of an alpha ray spectrograph consisting of the Princeton cyclotron-magnet and a plexiglass deflection chamber in which the alpha particles can be bent into a semicircle of about 80 centimeters maximum diameter. Three different methods of detection have been employed according to the different strengths of the radioactive sources. They are the ordinary photographic method, counting method and the method of photographic tracks. The behavior of the spectrograph has been investigated with polonium

alpha particles by the three methods of detection. The forms of the energy distribution have been determined respectively by these three methods and agree fairly well with one another. The half width of the main line under favorable conditions is less than one half millimeter which is equivalent to about 0.01 Mev. Microscopic examination ((of the experimental data)) reveals distinctly a series of weak groups in the low energy region while in the high energy region no indication of any discreet group has been found.

The behavior of the above spectrograph has been investigated with polonium alpha particles by using the three methods of detection. Sources of different forms have been prepared by spinning in RaD solution thin platinum wire, narrow strips of nickel foils and the edges (about 0.2 millimeter wide) of nickel foils. All surfaces to be used for coating with the source have been well polished previously.

- 3.6.5.61 BNL-7333. THE PRESENT EXPERIMENTAL AND THEORETICAL STATUS OF THE PROBLEM OF ELECTRON EJECTION IN THE ALPHA DECAY OF Po-210. William Robinson (Chemistry Department, Brookhaven National Laboratory, Upton, New York, USA), 11 pages

((This document could not be located either in total or in reference. Therefore no comments can be made on it.))

- 3.6.5.52 MLM-1165 (TR). POLONIUM. M. Haissinsky. Translated by Robert C. Lange. AEC Research and Development Report (Monsanto Research Corporation)

Table I Polonium isotopes and their nuclear properties (only Po-210 is described here). Po-210 (RaF: half life, 138.4 days; emissions, alpha; alpha energy, 5.3054 Mev.

THE RADIATIONS. The alpha rays of Po-210 are essentially mono-energetic. Their energy is 5.3054 Mev which corresponds to a mean range 3.84 centimeters in air at 15° centigrade and 760 millimeter mercury. The range in water is 39 microns and in nuclear emulsions 21 microns. Polonium emits approximately  $10^5$  alpha rays with 5.305 Mev for every particle with 4.5 Mev. The emissions correspond to the formation of an excited state of radium G. This energy difference is found as gamma rays of 0.8 Mev and their measured intensity corresponds to 1.25 quanta for every  $10^5$  alpha particles. Two other electromagnetic rays of 77 and 10 kev and have weak

intensities are also associated with the disintegration of Po-210. They rise from the perturbation of the electron periphery by the passage of nuclear radiation. One milligram of Po-210 corresponds to 4.5 curies or approximately  $10^{13}$  disintegrations per minute. Therefore, the equivalence between 1 curie and 222.2 micrograms of polonium can be calculated. One curie of Po-210 produces  $27.24 \pm 0.11$  calories per hour.



### 3.7 PLUTONIUM-238

#### 3.7.1 Nuclear Characteristics

3.7.1.1 Atomic Number - 94

3.7.1.2 Atomic Weight - 238

3.7.1.3 Half Life

(alpha) -  $87.48 \pm 0.08$  years Reference 1

(spontaneous fission) -  $5.2 \times 10^{10}$   
years Reference 2

3.7.1.4 Neutrons/Spontaneous Fission

$2.33 \pm 0.08$  Reference 3

3.7.1.5 Neutrons from Spontaneous Fission

2640 neutrons/sec gram (Pu) Reference 4

3.7.1.6 Neutrons by ( $\alpha$ , n) with  $\text{PuO}_2$

$2 \times 10^4$  neutrons/sec gram (Pu) Reference 5

3.7.1.7 Energy Levels and Decay Scheme

Pu-238  $\xrightarrow{\alpha}$  U-234

Alpha Energies  
(Mev)

Alpha Particles per  
Disintegration (%)

5.4912

72

5.4482

28

References

5.352

0.13

6, 7

5.208

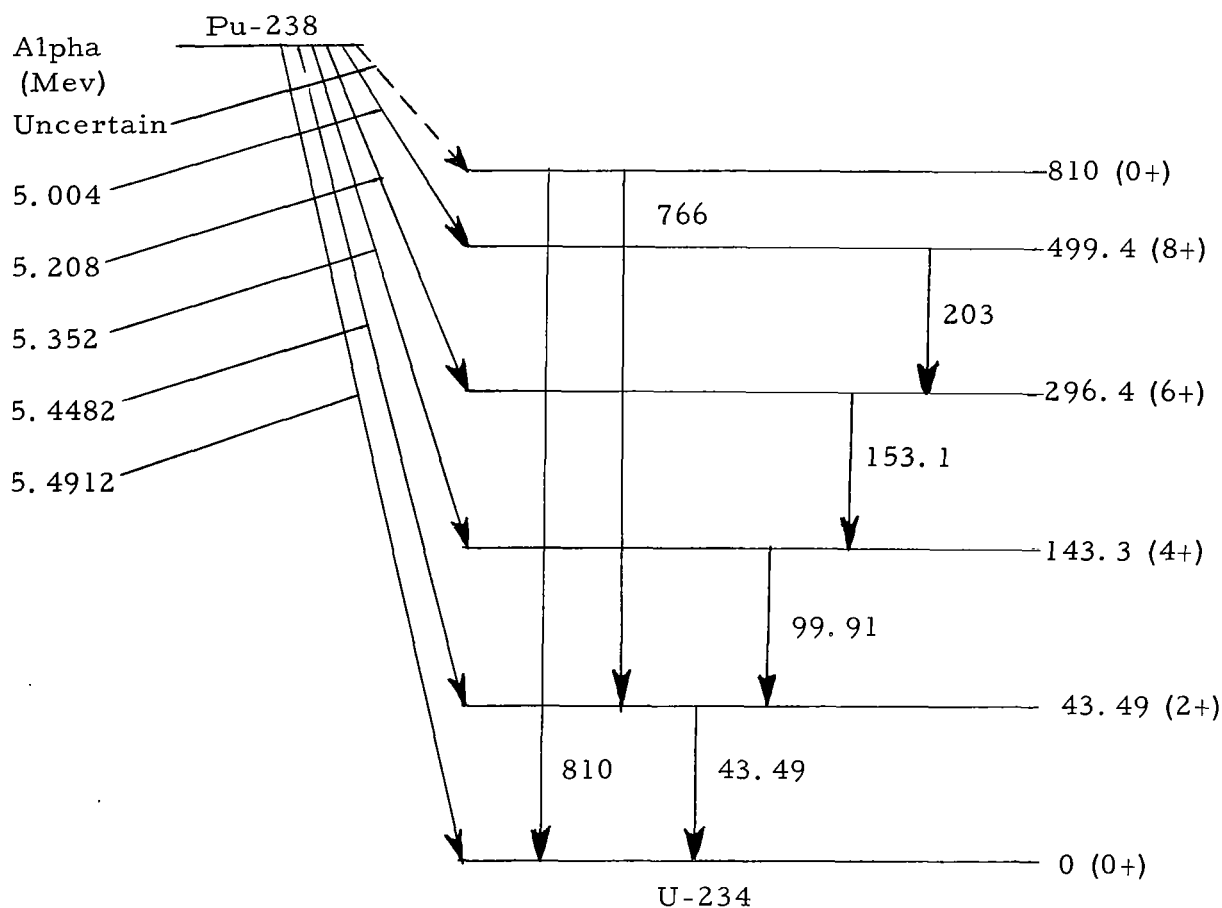
$5 \times 10^{-3}$

5.004

$7 \times 10^{-6}$

Gamma Energies (kev)	Percentage %	References 8, 9, 10
810	$10^{-5}$	
766	$5 \times 10^{-5}$	
203.0	$4 \times 10^{-6}$	
$153.1 \pm 0.6$	$1 \times 10^{-3}$	
$99.91 \pm 0.4$	$8 \times 10^{-3}$	
$43.49 \pm 0.08$	$3.8 \times 10^{-2}$	

There is a strong possibility that some of the gamma radiation attributed to the decay of Pu-238 may actually be the result of spontaneous fission. Reference 10.



Refs. 6, 7, 8, 9

### 3.7.1.8 Other Sources of Radiation

Pu-238 has inherent gamma radiation from spontaneous fissioning and from fission products. A survey of the literature revealed no specific information on the prompt fission gammas from the spontaneous fissioning of Pu-238. Information on the prompt gammas from U-235 fissioning is available (reference 11) and was used by shield designers from the Martin Company in estimating the effect of this source of radiation on shielding. The energies and abundance for U-235 adapted to Pu-238 are given in Table XVIII of this report.

Reference 12 also presents values for the gamma radiation due to spontaneous fissioning and is also presented in Table XVIII.

Table XVIII.

Gamma Radiation Associated with Pu-238 based  
on U-235

<u>Prompt Fission</u>		<u>Fission Products</u>	
<u>Energy (Mev)</u>	<u>Gammas Per Sec-Gram of Pu</u>	<u>Energy (Mev)</u>	<u>Gammas Per (Ref. 11) Sec-Gram of Pu</u>
0.5	4500	0.15	2400
1.0	2800	0.75	7100
1.5	1200	1.2	730
2.0	800	1.6	920
2.5	420	2.2	450
3.0 and up	500	2.4	170
		2.7	17.0

<u>Energy (Mev)</u>	<u>Gammas Per Sec-Watt</u>	<u>Energy (Mev)</u>	<u>Gammas Per (Ref. 12) Sec-Watt</u>
1.0	$6.60 \times 10^3$	0.63	$5.84 \times 10^3$
1.5	$1.654 \times 10^3$	1.10	$2.16 \times 10^3$
2.3	$1.75 \times 10^3$	1.55	$2.53 \times 10^3$
3.0	$3.09 \times 10^2$	2.30	$4.89 \times 10^2$
5.0	$4.72 \times 10^2$	2.75	$7.24 \times 10^2$

### 3. 7. 2      Chemical Properties

#### 3. 7. 2. 1    Fuel Forms

A number of plutonium compounds are reported in the unclassified literature. A summary is presented in reference 13 and some of their physical characteristics tabulated in Table XIX of this report. To date, only the metal and oxide forms have been seriously considered for use as a heat source in power conversion systems.

Table XIX    (Refs. 10, 13)

Properties of Plutonium Fuel Forms

<u>Compound</u>	<u>Molecular Weight</u>	<u>Wt. % Metal</u>	<u>Density g/cm<sup>3</sup></u>	<u>Sp. Power w/gm</u>	<u>Power Density w/cm<sup>3</sup></u>	<u>Melting Point °C</u>
$\alpha$ -Pu	238.0	100.0	15.92	0.328	5.22	639
$\beta$ -Pu	238.0	100.0	19.82	0.328	6.49	-
PuO <sub>2</sub>	270.0	88.15	11.46	0.289	3.31	2250
Pu <sub>2</sub> O <sub>3</sub>	524.0	90.84	11.47	0.298	3.42	2250
PuB <sub>6</sub>	302.87	78.58	7.31	0.257	1.88	-
PuB <sub>4</sub>	281.24	84.63	9.36	0.277	2.59	-
PuC	250.01	95.20	13.99	0.312	4.36	1650
Pu <sub>2</sub> C <sub>3</sub>	512.03	92.96	12.7	0.305	3.87	1900
PuN	252.01	94.44	14.23	0.309	4.40	2200
$\beta$ -PuSi <sub>2</sub>	294.17	80.91	9.18	0.265	2.43	-
PuP	268.97	88.49	10.18	0.290	2.95	-
PuS	270.06	88.13	10.60	0.289	3.06	-
Pu <sub>2</sub> O <sub>2</sub> S	540.06	88.14	9.95	0.289	2.88	-
Pu <sub>2</sub> S <sub>3</sub>	572.19	83.19	8.41	0.273	2.30	-

#### 3. 7. 2. 2    Material Compatibility

Compatibility studies have been made at the Martin Company to evaluate various metals as containment barriers for the selected fuel forms of plutonium (reference 14). Simulated fuel materials were used in these studies. Cerium was chosen to simulate plutonium in the experimental determinations. Cerium dioxide, cerium dicarbide, and cerium sesquicarbides were used to simulate the respective plutonium compounds. Except for the tests which included iron and nickel, studies were conducted with pellets of

the compound placed in direct contact with the various metals under investigation. Iron and nickel were tested in cermet form. Compatibility was determined metallographically. Table XX lists the experimental results at 2000°F. From the results, it would appear that only the refractory metals are attacked by the cerium compounds.

Table XX (Ref. 9)

Compatibility of  $\text{CeO}_2$  and  $\text{CeC}_2$

<u>Materials</u>	<u>Time (hr)</u>	<u><math>\text{CeO}_2</math> Compatibility</u>	<u><math>\text{CeC}_2</math> Compatibility</u>
Haynes 25	150	No attack	No attack
Hastelloy-C	150	No attack	No attack
Iconel-X	150	No attack	No attack
Type 316 SS	150	No attack	No attack
Molybdenum	150	Intergranular attack	Attack
Tantalum	150	Intergranular attack	Attack

Substitute for plutonium compounds with metals at 2000°F in an argon environment.

Literature survey compatibility information is presented in Table XXI. Only those results pertaining to the plutonium oxide are reported here since this is the most probable compound. Results of experimental work done at the Martin Company is presented in Table XXII.

Table XXI

Compatibility Information Obtained from a Literature Survey

<u>Isotope Form</u>	<u>Cladding Material</u>	<u>ATM</u>	<u>Temp.</u>	<u>Time</u>	<u>Reaction</u>
$\text{PuO}_2$	Fe, Aust. St. Steel, Nd	Air	1500°C	100 hrs	Very little

Table XXI (continued)

<u>Isotope Form</u>	<u>Cladding Material</u>	<u>ATM</u>	<u>Temp.</u>	<u>Time</u>	<u>Reaction</u>
PuO <sub>2</sub>	Tantalum	H <sub>2</sub>	1500°C	--	High diffusion rate
	Molybdenum	H <sub>2</sub>	1500°	--	None
	Tungsten	H <sub>2</sub>	1500°	--	None
PuO <sub>2</sub>	Mo, Ta, W, Al <sub>2</sub> O <sub>3</sub> , BaO, MgO, ZrO <sub>2</sub>	H <sub>2</sub> or A	1700°	1-1/2 hrs	All pellets melted because of Pu <sub>2</sub> O <sub>3</sub> content. MgO approval reasonably compatible
PuO <sub>2</sub>	304, 316, 318 347 std steel and inconel	--	2150°C	2 hrs	PuO <sub>2</sub> pellets remained intact
PuO <sub>2</sub>	Tungsten	H <sub>2</sub>	1593°C	20 hrs	Showed core pits
Molten PuO <sub>2</sub>	Tungsten	--	--	30 sec	Did not decompose
PuO <sub>2</sub>	Tantalum				

Table XXII

Summary of Experimental Work Performed at  
the Martin Company

<u>Isotope Form</u>	<u>Cladding Material</u>	<u>ATM</u>	<u>Temp.</u>	<u>Time</u>	<u>Reaction</u>
PuO <sub>2</sub>	Tantalum	argon	3100°F	7 hrs	No reaction
PuO <sub>2</sub>	Sintered with MgO	H <sub>2</sub>	2900°F	4 hrs	No reaction
PuO <sub>2</sub>	Sintered with ZrO <sub>2</sub>	H <sub>2</sub>	2900°F	4 hrs	Alloyed. Formed continuous solid solution.

Table XXII (continued)

<u>Isotope Form</u>	<u>Cladding Material</u>	<u>ATM</u>	<u>Temp.</u>	<u>Time</u>	<u>Reaction</u>
PuO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub> of un-known purity	H <sub>2</sub>	2900°F	4 hrs	Formed perovskite Pu AlO <sub>3</sub>
PuO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub> 99.99% pure	H <sub>2</sub>	2900°F	4 hrs	No reaction

### 3.7.2.3 Effects of Impurities on Nuclear Characteristics

Although the isotopic purity obtained for Pu-238 is quite high (approximately 80%, reference 15), it still can be expected to be contaminated with other plutonium isotopes. These isotopes act only as diluents and do not alter the spectrum or quantities of emitted radiation. In the shielding calculations performed at the Martin Company, only the pure Pu-238 energy spectrum (alpha and gamma and neutrons) is considered.

### 3.7.3 Thermal and Physical Properties of the Most Probable Fuel Form(s)

To date only two plutonium fuel forms have been given consideration as a heat source. These are the metal and the dioxide. Table XXIII presents the thermal and physical properties of these two fuel forms. The values of power density shown are given in units of w/gm. In each case this refers to watts/gm of Pu-238.

Table XXIII

#### Physical Properties

		<u>Pu Metal</u>	<u>PuO<sub>2</sub></u>
Thermal conductivity	$\frac{\text{cal cm}}{\text{sec } ^\circ\text{C cm}^2}$	0.012 $\pm$ 10( Pu) (a) Ref. 6	0.014 (400°C) (b)(c)(Table XXIV)Ref. 17
Specific power	$\frac{\text{watts}}{\text{gm}}$	0.55 Refs. 12, 18	0.305 Ref. 4

Table XXIII(continued)

		<u>Pu Metal</u>	<u>PuO<sub>2</sub></u>
Thermal output	$\frac{\text{watts}}{\text{K curie}}$	32.1(d) Ref. 13	32.1(d) Ref. 13
Heat capacity	$\frac{\text{cal}}{\text{gm } ^\circ\text{C}}$	0.0456(e) Table XXV Ref. 17	0.059(c)(f) Ref. 16
Heat of fusion	$\frac{\text{cal}}{\text{mole}}$	940 $\pm$ 150 (g) Table XXV Ref. 17	16,000(c) Ref. 16
Weight density	$\frac{\text{gm}}{\text{cc}}$	19.86(h) Table XXVI Ref. 23	11.46 Refs. 19, 20
Melting point	$^\circ\text{C}$	640 Ref. 19	2240(i) Ref. 19
Boiling point	$^\circ\text{K}$	3500 Ref. 16	4140(j) Ref. 21
Specific activity	$\frac{\text{dis/min}}{\text{microgram}}$	3.72 $\times 10^7$ Ref. 22	

## Key to Table XXIII

- (a) Value for Pu.
- (b) Value at 400 $^\circ\text{C}$ , table of values in Table XXIV.
- (c) These are the values for UO<sub>2</sub> but since UO<sub>2</sub> and PuO<sub>2</sub> show such strong physical and chemical properties the UO<sub>2</sub> values can be used.
- (d) This is the value calculated using a power density of 0.55 watts/gm and a half life of 87.5 y.
- (e) This is a representative value at 407 $^\circ\text{C}$ . A complete table of heat capacity vs temperature can be found in Table XXV.



- (f) A representative of the values of heat capacity  $C_p$  for  $\text{PuO}_2$  can be found by using the following equation:

$$C_p = 19.2 + 1.62 \times 10^{-3} T - 3.957 \times 10^{-5} T^2 \left[ \frac{\text{cal}}{\text{mole } ^\circ\text{C}} \right]$$

- (g) Heat of fusion for the to liquid phase. The latent heat required to go through all the phases of Pu can be found from Table XXV.
- (h) Value for Pu at  $21^\circ\text{C}$ . The densities of the other phases of Pu can be found by referring to Table XXV, Structure Data on the Crystal Phases of Pu Metal.
- (i) This is obtained from the vapor pressure  $\log p$  (atm) =  $\frac{(29240 \pm 530)}{T} + (8.072 \pm 0.239)$ .
- (j)  $\text{PuO}_2$  heat in an inert atmosphere releases oxygen to yield a mixture of  $\text{PuO}_2$  with lower oxides and the resulting values are lower than the true melting point. The values referred to above were heated in a helium atmosphere.

There is little or no thermal conductivity data available for refractory plutonium compounds. However, since the uranium compounds show strong physical and chemical similarity to the compounds of plutonium, the thermal conductivity of  $\text{UO}_2$  has been cited. These are shown in Table XXIV as follows:

Table XXIV (Ref. 13)

Thermal conductivity  $\left[ \frac{\text{cal cm}}{\text{sec } ^\circ\text{C cm}^2} \right]$  for  $\text{PuO}_2$   
vs Temperature  $^\circ\text{C}$

Temperature	100 $^\circ\text{C}$	400 $^\circ\text{C}$	1000 $^\circ\text{C}$
Thermal Conductivity	0.023	0.014	0.008

Table XXV is a table of the specific heat and heat of transformation of plutonium between the temperature of  $-62^\circ\text{C}$  to  $546^\circ\text{C}$ . Table XXVI indicates the crystal phases of Pu metal as well as its density as a function of temperature.

Table XXV (Ref. 16)

The Specific Heat and Heats of Transformation  
of Plutonium

Temperature (°C)	Specific Heat (cal / gm x 10 <sup>3</sup> )	Temperature (°C)	Specific Heat (cal / gm x 10 <sup>3</sup> )
62.0	33.4	127 = superheat; the true transformation temperature = 121°C; the latent heat of the $\beta$ transition = 958 $\pm$ 10 cal/gm-atom.	
58.5	33.4		
50	34.2		
44	35.2		
32	37.5		
25	38.9		
21	38.7		
13	38.8	127	161.9
1.5	35.7	130	99.0
1.0	35.5	132	72.2
5.0	32.4	133	57.9
13	35.5	135	50.9
15	33.3	140	43.1
20	33.8	145	40.5
29	34.1	152	39.9
38	34.5	158	41.1
44	34.9	167	41.9
52	36.6	171	42.0
62	36.6	176	42.5
65	35.0	184	41.6
70	38.7	190	41.9
79	38.6	192	43.3
81	41.3	197	43.4
85	41.5	200	42.3
90	41.3	204	42.8
96	42.5	208	43.4
104	44.9	210	49.7
111	44.9	211	90.3
113	47.8	$\beta \rightarrow \nu$ transition; latent heat = 140 $\pm$ 15 cal/gm-atom.	
118	52.8		
121	56.0		
122	60.9	212	66.5
124	76.7	212	59.9
126	101.7	213	42.6
126	129.2	218	40.8

Table XXV (continued)

Temperature (°C)	Specific Heat (cal/gm x 10 <sup>3</sup> )	Temperature (°C)	Specific Heat (cal/gm x 10 <sup>3</sup> )
226	42.3	407	45.6
230	44.0	412	47.8
236	42.0	420	48.6
242	44.8	426	48.6
243	44.4	433	45.0
250	45.0	442	47.0
254	46.3	449	49.7
260	45.2	450	50.2
297	47.0	452	51.0
303	49.9	454	53.9
305	50.5	456	56.6
307	60.7	457	64.1
312	89.6	459	86.8
315	128.1		
$\gamma\delta'$ transition; latent heat = 156 $\pm$ cal/gm-atom			
		$\delta\delta'$ transition; latent heat = 17 $\pm$ 10 cal/gm-atom	
		461	62.9
318	87.3	464	58.6
320	82.0	468	65.8
322	73.9	470	75.0
328	62.3	473	79.4
326	56.8	473	100.6
327	52.8	477	141.6
332	48.5	480	155.2
334	47.1	481	157.8
343	44.6	483	268.1
346	45.5		
350	45.5	$\delta'\epsilon$ transition; latent $\delta'$ heat = 470 $\pm$ cal/gm-atom	
356	48.1		
361	46.5		
368	46.9	486	55.6
374	46.4	488	43.8
376	46.9	491	43.4
379	47.1	498	43.1
383	46.4	505	44.0
389	48.3	513	42.2
397	45.7	518	39.4

Table XXV (continued)

Temperature (°C)	Specific Heat (cal/gm x 10 <sup>3</sup> )
537	41.7
533	35.4
538	36.9
546	37.6

ℰ liquid transition at ℰ 639°C; latent heat =  $940 \pm 150$  cal/gm-atom; total heat of plutonium in solid state =  $11.7 \pm 0.5$  Kcal/gm-atom.

Table XXVI (Ref. 23)

Structure Data on the Crystal Phases of Plutonium Metal

Reference	Phase	Symmetry and Cell Content	Cell Dimensions in.	Density in g/cm <sup>3</sup>
5	ℳ	Monoclinic Z = 16 Space Group: P2 <sub>1</sub> /m	t = 21°C a = 6.183 b = 4.822 c = 10.963 8 = 101.79°	19.86
8	β	Monoclinic Z = 34 Space Group: I2/m	t = 190°C a = 9.284 b = 10.463 c = 7.859 8 = 92.13	17.70
6	γ	Orthorhombic Z = 8 Space Group: Fddd	t = 235°C a = 3.159 b = 5.768 c = 10.162	17.14
7	ℳ	Cubic Z = 4 Al Structure	t = 320°C a = 4.637	15.92

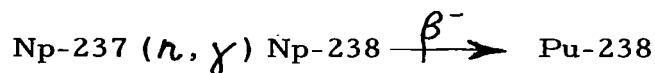
Table XXV (continued)

Reference	Phase	Symmetry and Cell Content	Cell Dimensions in.	Density in g/cm <sup>3</sup>
7	$\gamma'$	Tetragonal $Z_1 = 1$ A1 Structure variant	$t = 465^\circ\text{C}$ $a = 3.327$ $c = 4.482$	16.00
7	$\xi$	Cubic $Z = 2$ A2 Structure	$t = 490^\circ\text{C}$ $a = 3.636$	16.51

### 3.7.4 Isotope Production, Availability and Cost

#### 3.7.4.1 Production Method

Although Pu-238 can be produced in a number of ways the most economical at present is through the production of Np-237 by the extended irradiation in a reactor of U-235. The Np-237 is further irradiated to produce Pu-238



#### 3.7.4.2 Availability and Cost

Prospective production levels of plutonium for use in auxiliary power devices - as published by the Division of Isotope Development, USAEC in January 1963 - are presented below:

	<u>1964</u>	<u>1965</u>	<u>1966</u>	<u>1967</u>	<u>1968</u>	<u>1969</u>	<u>1970</u>	<u>1980</u>
Kgm	6.0	13	18	24	32	36	42	52
Kw(t)	2.8	6.25	8.6	11.0	15.3	17.3	20	25

The above values are based primarily on Government production facilities. In the near future private power reactor production is expected to increase the available plutonium considerably. In 1970 the quantity of plutonium is expected to be 26 Kw(t) and in 1980 this will rise to 73 Kw(t) (reference 24).

Plutonium costs are quite high. Present costs are \$1000/gram, but are expected to decrease to \$500/gram (894 \$/w(t)) at a yearly production rate of 50 Kg.

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### 3. 7. 5      Annotated Bibliography

3. 7. 5. 1      FISSION CROSS SECTION MEASUREMENTS ON Pu-239, Pu-241, U-232 AND A SEARCH FOR FISSION COMPONENTS IN Pu-238 RESONANCES. G. D. James (Atomic Energy Research Establishment, Harwell, Berks, Eng.). Vienna, International Atomic Energy Agency, 1965, Preprint No. SM-60/15, 13p. (CONF. -650301-18)

From Symposium on the Physics and Chemistry of Fission, Salzburg, Austria.

Fission cross section measurements on Pu-239 and Pu-241 were carried out from sub-thermal neutron energy to the kev region. Resonance parameters are given and errors in the average values of the cross section are discussed. The discovery of large fission components in the U-232 neutrons resonance stimulated a search for fission components in Pu-238. The results of these experiments are given. All the measurements were performed on the Harwell time-of-flight spectrometer.

3. 7. 5. 2      (HW-68600-43) PLUTONIUM ABSTRACTS, Volume 4, Number 9. (General Electric Co., Hanford Atomic Products Operation, Richland, Washington). Oct. 1, 1964. Contract AT(45-1)-1350. 20 p.

A total of 32 references is presented on new developments in plutonium technology. The references are arranged into the following subject fields: Biology and Medicine, Chemistry, Health and Safety, Metallurgy and Ceramics, Physics, and Miscellaneous.

3. 7. 5. 3      RADIOCHEMICAL DETERMINATION OF PLUTONIUM ISOTOPES AND AMERICIUM-241 IN A PLUTONIUM SAMPLE. Kenju Watanabe, Eiji Sakai, and Kentaro Minami (Japan Atomic Energy Research Inst., Tokyo). J. Nucl. Sci. Technol. (Tokyo), 1: 197-202 (Sept. 1964)

Isotopic ratios of Pu-238, Pu-239, Pu-240, and Am-241 in a plutonium sample were approximately determined by means of combined alpha and gamma spectrometry without chemical separation of americium from plutonium. The intensity of alpha ray followed by internal conversion was determined by measuring the intensity of LX-rays. The ratio of Pu-240 to Pu-239 was obtained from the ratio between the alpha ray intensity thus determined and the total alpha ray intensity from the two nuclides.

- 3.7.5.4 (HW-68600-42) PLUTONIUM ABSTRACTS. Volume 4, Number 8. (General Electric Co., Hanford Atomic Products Operation, Richland, Washington). Sept. 1, 1964 Contract AT(45-1)-1350. 18p.

A total of 21 abstracts is presented on new developments in plutonium technology. The abstracts are arranged under the subject headings: Biology and Medicine, Chemistry, Metallurgy and Ceramics, and Physics.

- 3.7.5.5 (HW-68600-41) PLUTONIUM ABSTRACTS. Volume 4, No. 7. (General Electric Co., Hanford Atomic Products Operation, Richland, Washington). Aug. 1, 1964. Contract AT(45-1)-1350, 34p.

Information concerning new developments in plutonium technology is presented. Subject fields reviewed include Biology and Medicine, Chemistry, Metallurgy and Ceramics, Physics, Reactor Technology, and Miscellaneous.

- 3.7.5.6 MLM-1196. MOUND LABORATORY PROGRESS REPORT FOR APRIL 1964. J. F. Eichelberger, G. R. Grove, and L. V. Jones (Mound Laboratory, Miamisburg, Ohio). July 10, 1964. 30p.

The half life of Pu-238 was determined by measuring the decay rate of two samples with Calorimeters 39 and 90. Each power measurement was corrected for other isotopes of plutonium, based upon the weight of the samples and analyses by mass spectrometry. By propagation of errors, using the external probable error because it is larger, the half life and probable error of Pu-238 from this work is  $87.48 \pm 0.08$  years\*.

\*Verified by private communication with Dr. G. R. Grove on 28 October 1964.

- 3.7.5.7 LOW-ENERGY GAMMA TRANSITIONS IN Pu-238 AND Pu-240. G. G. Akalaev, N. A. Vartanov, and P. S. Samoilov. At. Energ. (USSR). 16: 452-3 (May 1964) (In Russian)

A study was made of the electron and gamma spectra of a mixture of curium isotopes in a magnetic beta spectrometer with double focusing

and in a scintillation gamma spectrometer. Spectral line intensity, electron energy, conversion shell, transition energy, and relative line intensity are presented for lines in the electron spectrum of Pu-238 and Pu-240, formed in the alpha decay of Cm-242 and Cm-244. The ratios of coefficients of internal conversion in E2 transitions with energies 43 and 44 kev for  $Z = 94$  are derived and compared with those from two other experiments and from theoretical calculations.

- 3.7.5.8 ALPHA SOURCES FOR LOW THRUST TASKS IN SPACE. W. Diethorn (Nuclear Engineering Department, Pennsylvania State University, Pennsylvania). International Journal of Applied Radiation and Isotopes 15, 127-31, March 1964

The recoil thrust exerted on a flat plate alpha source is calculated for alpha particle escape from one side of the source. The thrust is low even for the high specific activity alpha emitters Po-210, Pu-238 and Cm-244 now receiving increased production emphasis. It is suggested that these three alpha emitters may find some future applications in space exploration requiring low thrust devices.

((This article presents a theoretical discussion on the maximum thrust capabilities of the subject alpha emitters. Information is also presented in Table 1, not reproduced here, of the chemical and physical properties of the isotopes. The information presented in the table is taken from several other articles.))

- 3.7.5.9 Refer to 3.6.5.5. (MLM-1200) GAMMA SHIELDING REQUIREMENTS FOR PLUTONIUM-238 AND POLONIUM-210. K. W. Foster (Mound Laboratory, Miamisburg, Ohio). May 15, 1964. Contract AT(33-1)-Gen.-53. 9p.
- 3.7.5.10 Refer to 3.10.12. (RSG-A-1) PRELIMINARY REPORT ON PUBLIC HEALTH HAZARD ANALYSES OF SPACE ISOTOPIC POWER SOURCES. J. J. Fitzgerald, Gordon L. Brownell, and N. Irving Sax - Jack J. Gabay, ed. (New York, State Department of Health, Albany). Mar. 1, 1964. Contract SAPH 76262, 87p.
- 3.7.5.11 PLUTONIUM, Mieczyslaw Taube. Translated by Eugeniusz Lepa and Zbigniew Nanowski. A Pergamon Press Book, New York, The Macmillan Company, 1964. 262 p.

Information is included on the nuclear properties of Pu, chemical properties of Pu, physiological effects of Pu and health physics, Pu technology, Pu nuclear fuel, and role of Pu in development of nuclear power. 372 references are included.

- 3.7.5.12 Refer to 3.6.5.7. RADIOISOTOPE POWER SYSTEMS FOR MANNED SPACE STATIONS. Alan J. Streb (Martin Company, Baltimore). New York, American Institute of Aeronautics and Astronautics, 1964. Paper No. 64-711, 31p. (CONF-640901-3)

From 3rd Biennial Aerospace Power Systems Conference, Philadelphia, Sept. 1964.

- 3.7.5.13 TERNARY FISSION OF PLUTONIUM. N. A. Perfilov, Z. I. Solov'eva, R. A. Filov, and G. I. Khlebnikov, J. Exptl. Theoret. Phys. USSR 44, 1832-6, June 1963. ((Translated in Soviet Physics JETP Vol. 17, No. 6, December 1963.))

The energy spectra of long range alpha particles produced in the spontaneous fission of Pu-238 and Pu-240 and in thermal neutron fission of Pu-239 are studied by the nuclear emulsion method. The spectrum shapes are discussed and are compared with the results for complex uranium fission.

In our study of spontaneous fission we used electrolytic films of Pu-238 and Pu-240 containing  $78.2 \pm 4$  and  $450 \pm 25$  micrograms.

- 3.7.5.14 Refer to 3.4.5.10. EVALUATION OF ISOTOPE THERMIONIC GENERATORS. Robert J. Harvey, Thomas C. Robinson, Joseph J. Fitzgerald and Gordon Brownell. Transactions of the American Nuclear Society, 6, 88-91, June 1963
- 3.7.5.15 DETERMINATION OF THE ENERGIES OF INTENSE ALPHA GROUPS EMITTED BY THE NUCLEI OF PLUTONIUM-238, 239, 240 AND NUCLEI OF AMERICIUM-241. Chin-Fan Leang (Centre de Spectrometrie Nucleaire et de Spectrometrie de Masse, Orsay, France). Compt. rend. 255: 3155-7 (Dec. 5, 1962) (In French)

The energies of the intense alpha rays of Pu-238, Pu-239, Pu-240, and Am-241 were determined by magnetic spectroscopy with respect to Bi-212 with a precision of 1/7000.

3. 7. 5. 16 Refer to 3. 8. 5. 11. A SURVEY OF RESEARCH ON THE TRANSURANIC ELEMENTS. Contemporary Physics 2, 385-402, June 1961.
3. 7. 5. 17 SPONTANEOUS FISSION OF NUCLEI. K. A. Petrzhak and G. N. Flerov, USP. Fiz. Nauk, 73, 655-683, April 1961. ((Translated in Soviet Physics USPEKHI, Vol. 4, No. 2, September-October 1961.))

((The subject article is more or less a primer on the subject of spontaneous fission of nuclei. It contains no specific experimental information but is of value as far as basic understanding of spontaneous fission is concerned. It contains a history of the study of spontaneous fission from the 1930s up through the date of the article.))

3. 7. 5. 18 SPONTANEOUS FISSION PERIODS OF Np-237, Pu-238 and Pu-242. V. A. Druin, V. P. Perelygin and G. I. Khlebnikov (Joint Institute for Nuclear Research). J. Exptl. Theoret. Phys. (USSR) 40, 1296-8, May 1961, translated in Soviet Physics JETP Vol. 13, No. 5, November 1961

An attempt has been made to determine the true spontaneous fission half period of Np-237 by employing nuclear photographic emulsions prior to development. The photographic plates were treated with potassium ferrocyanide to remove the background alpha particle tracks. The reliability of this method was checked by measuring the spontaneous fission half periods of Pu-238 and Pu-242 which were independently determined using a proportional counter. The results obtained for plutonium by the various methods are identical and agree with other available data. Only three fragment tracks were detected in the Np-237 photographic measurements. Thus only a lower limit of  $10^{18}$  years can be established for the half period as compared with the usually accepted value of  $4 \times 10^{16}$  years.

Seven samples were prepared, three of Np-237, two of Pu-238 (0.27 and 0.35 micrograms) and two consisting of a mixture of the isotopes Pu-238 (14%) and Pu-242 (86%) and containing 0.37 and 0.80 micrograms of Pu-238, respectively. As a result of the measurements,

the following values for the half periods were obtained:  $5.2 \times 10^{10}$  years for Pu-238 and  $6.7 \times 10^{10}$  years for Pu-242.

3.7.5.19 THE DECAY OF NEPTUNIUM-238. R. G. Albridge and J. M. Hollander (Lawrence Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California). Nuclear Physics 21, 438-449, 1960

A study has been made of the energy levels of Pu-238 which are postulated by Np-238 beta decay or by examination of the Np-238 conversion electron spectrum in high resolution beta spectrographs. The general features of the level scheme as previously given are unchanged but several new transitions are observed with energies of 119.8, 871, 943, 989, and 1034 keV and two new levels are postulated at 915 and 1034 keV which accommodate all but the 943 keV transition. A possible assignment of the 943 keV transition to the  $(0^+, 0)$  state of the beta vibrational band is discussed. In addition the weak 885 keV transition from the  $2^+$  state of the gamma vibrational band to the  $4^+$  state of the ground band is seen and its relative intensity determined. Comparisons are made of the experimental relative intensities of the three photons depopulating this band with those predicted from the rules of Alaga, et al. Only fair agreement is noted. A discussion is given of the beta decay branchings and log ft values of Np-238 decay in terms of the postulated characters of the Pu-238 states and the measured standard of Np-238.

An Np-238 sample was prepared by a five hour neutron irradiation of approximately 200 micrograms of Np-237 in the materials testing reactor at Arco, Idaho. The sample was dissolved in concentrated HCl and the neptunium precipitated with  $Zr_3(PO_4)_4$  after reduction to the  $+4$  state by  $++$ . Further purification was effected by cold precipitation with  $LaF_3$  and  $La(OH)_3$ . To separate the neptunium from lanthanum and other rare earths, the solution was passed through a Dowex A-1 anion column at high fluoride concentration and the neptunium subsequently elutriated with one molar HCl. This sample was electroplated onto a 0.25 mm platinum wire which served as a source for the spectrographs. ((The subject article is concerned primarily with the decay scheme of Np-238 ending in the Pu-238 level. It may be of interest to those wishing to have a complete picture of the decay scheme of Pu-238 to reference this article.))

- 3.7.5.20 ENERGY LEVELS OF Pu-238 and Pu-239 NUCLEI.  
S. A. Baranov and K. N. Shlyagin. p. 393-407 of  
"Soviet Research on the Lanthanide and Actinide Elements,  
1949-1957. PART III. Nuclear Chemistry and Nuclear  
Properties". (In English Translation). New York,  
Consultants Bureau, Inc. (1959)

The beta and gamma transitions in the decay of Np-238, Np-239, and Cm-242 were investigated using beta spectrometer, scintillation spectrometer, and proportional counter techniques. Possible spin values of the Pu-239 nuclear levels were determined and a hypothesis established as to the possibility of the existence of rotational levels for this even-odd nucleus. Possible energy level schemes for Pu-238 and Pu-239 are presented.

- 3.7.5.21 SPECTRUM OF THE INTERNAL CONVERSION ELECTRONS  
ACCOMPANYING DECAY OF Pu-238 and Pu-240. E. F.  
Tretyakov, L. N. Kondrat'ev, G. I. Khlebnikov and L. L.  
Gol'din, J. Exptl. Theoret. Phys, USSR 36, 362-366,  
February 1959. ((Translated in Soviet Physics JETP  
Vo. 36, No. 2, August 1959.))

The spectrum of conversion electrons accompanying the alpha decay of Pu-238 and Pu-240 was studied by means of a high transmission magnetic spectrometer with toroidal field shape which measured alpha-electron coincidences. Transitions from the 6+ excited levels were detected and the multi-polarity and more precise energy values were determined for transitions from the 4+ and 2+ levels.

- 3.7.5.22 NUCLEAR SPECTROSCOPY. I. Perlman (Lawrence  
Radiation Laboratory, University of California, Berkeley).  
Proceedings of the National Academy of Science, U.S.,  
45, 461-70, April 1959

((This paper deals with nuclear states in the heavy element region and how information is obtained from the study of alpha radioactive substances. The article is entirely theoretical in nature and is in the form of a basic study of nuclear spectroscopy. It contains no particular experimental information.))

- 3.7.5.23 ELECTRIC MONOPOLE TRANSITIONS IN Pu-238 AND  
U-234. I. Perlman, Frank Asaro, E. G. Harvey and  
F. S. Stephens, Jr. (University of California, Berkeley).  
Bull. of the American Physical Society, Series II, Vol. 2,  
394 December 1957.

((The following is the entire reference presented in the subject document.))

Electric monopole transitions in Pu-238 and U-234 were seen following the alpha decay of Cm-242 and Pu-238, respectively. An anthracene crystal spectrometer was necessary to register the weak K and L + M lines which occurred in fewer than  $1 \times 10^6$  alpha disintegrations. The K/L + M ratios were 4.5 which agrees with the value 5 given by theory for an EO transition. Each EO transition was shown to lead to an excited 0+ state to the 0+ ground state and to compete with an E-2 gamma ray from the same excited level to the 2+ state of the ground state rotational band. This competition will be discussed in terms of the theory. The EO assignment was made by determining minimum values for the conversion coefficients and showing that the high values so obtained could not arise from high multi-polarity (M4 or above) transitions. The excited 0+ state in Pu-238 is at 935 kev and that for U-234 at 810 kev. The U-234 state is undoubtedly the same as that previously seen following the beta decay of UX<sub>2</sub>.

- 3.7.5.24 ALPHA DECAY OF PLUTONIUM-238. L. N. Kondrat'ev, G. I. Novikova, V. B. Dedov, and L. L. Gol'din, Izvestia Akad Nauk, USSR, 21, 907-8, July 1957

Knowing the alpha decay intensities to successive levels of one rotational band, we can draw certain conclusions regarding the shape of the daughter nucleus. In view of this, accumulation of experimental material on alpha decays is of considerable interest. In the present work we investigated the highest excited rotational states of U-234 evidenced in alpha decay of Pu-238. The Pu-238 was obtained as a product of alpha decay of Cm-242 formed by slow neutron bombardment of Am-241. The elements were separated by chromatographic method. The separation of the plutonium from the americium was so thorough that no trace of Am-241 alpha lines appeared in the recorded spectra.

- 3.7.5.25 HALF LIFE OF Pu-238. Darleane C. Hoffman, George P. Ford and Francine O. Lawrence (University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico). Journal of Inorganic and Nuclear Chemistry 5, 6-11, 1957

The half life of Pu-238 has been measured by determining its rate of growth from Cm-242. The weighted average of two determinations is 86.41 years with an estimated error of 0.3 years.



The Cm-242 used in the experiment was produced by thermal neutron irradiation of Am-241 and initial hot lactate cation resin column was used to separate the bulk of the Am from the Cm. The alpha activity from Cm-243 and Cm-244 corrected to 12 July 1955, the time of standardization of the curium solutions, was calculated to be 0.88% of the Cm-242 alpha activity. ((A substantial portion of this article is devoted to the preparation and analysis of the purities of samples and is too extensive to be covered in this annotated bibliography.))

3. 7. 5. 26 ENERGY LEVELS OF Pu-238 and Pu-239 NUCLEI.  
S. A. Baranov and K. N. Shlyagin. Soviet J. Atomic  
Energy, No. 1, 51-65 (1956)

A double-focusing magnetic beta spectrometer, a gamma scintillation spectrometer, and a proportional counter were used to study the radiations from Np-238, Np-239, and Cm-242. Resultant spectra are shown, and the level schemes of the Pu-238 and Pu-239 nuclei are given.

3. 7. 5. 27 GAMMA RADIATION FROM THE DECAY OF PLUTONIUM-238, CURIUM-242, CURIUM-243. J. O. Neuton, D. Rose and J. Milsted (Atomic Energy Research Establishment, Harwell). Philosophical Magazine 1981-1002, November 1956

Accurate measurements on the gamma rays from the decay of Pu-238 and Cm-242 have been made with proportional counters. Values of  $43.49 \pm 0.08$  and  $99.8 \pm 0.4$  and  $153.1 \pm 0.6$  kev for Pu-238 and  $44.03 \pm 0.06$ ,  $101.80 \pm 0.17$  and  $157.61 \pm 0.3$  kev for Cm-242 were obtained. The ratios of these energies for each nuclide agree well with those predicted in the strong coupling limit of the unified model. Gamma rays having energies of  $210 \pm 1.5$ ,  $228 \pm 2$ , and  $277 \pm 2$  kev and K-X radiation were found from the decay of Cm-243. These gamma rays are shown to be magnetic dipole with a 30% mixture of electric quadrupole. The magnetic dipole component is  $10^4$  times slower than the single particle estimate.

Preparation of Cm-242 and Pu-238 sources: two samples of curium were used in the present work, one from an irradiation in BEPO at Harwell and the other from a much more intense irradiation in the NRX reactor at Chalk River. The integrated neutron doses for these irradiations were approximately  $2 \times 10^{18}$  and  $10^{20}$  n/cm<sup>2</sup>, respectively. It is unlikely that an appreciable amount of Cm-243 was

formed in the BEPO irradiation but a fractional percentage by weight of this isotope could have been formed in the NRX irradiation by further neutron capture in Cm-242. In each case the curium was separated from the irradiated americium by the following chemical steps - precipitation as a fluoride from a solution in which the americium has been oxidized to the fluoride soluble hexavalent state, separation from rare earth by elutriation from cation exchange column with 12 normal hydrochloric acid, separation of curium from residual americium by elutriation from a cation exchange column with ammonium citrate solution at 85° centigrade.

Pu-238, the alpha daughter of 162 day curium-242, was extracted from purified curium fractions which had been standing several months by a further ion exchange step. The plutonium fraction was purified chemically by conventional solvent extraction and precipitation reactions on an ultra micro scale. The Pu-238 used in the present work was shown by pulse analysis to contain negligible amounts of Cm-242 and other alpha emitters. ((The decay schemes for Pu-238 and Cm-242 are presented as Figure 9 in this article.))

((Cited as best values for gamma ray energies in Rev. Mod. Phys. Vol. 30, #2, pt II, April 1958.))

3. 7. 5. 28 FIRST EXCITED STATES OF HEAVY EVEN-EVEN NUCLEI. Jack M. Hollander (Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California). Physical Review 103, 1590-1, September 1956

The systematic behavior of first excited states of even-even nuclei is well known, and indeed the pronounced maxima of the first excited state energies at the "magic numbers" are among the most striking manifestations of nuclear shell structure. Between the closed shells, in the regions  $155 < A < 185$  and  $A > 225$ , rather flat minima are developed whose constancy and low energy point to the collective nature of the excitations. It is the purpose of this letter to assemble some recent data which illustrates the "second-order" systematics of these excited states. Most of the energies quoted here were obtained from precision measurements of conversion electron spectra in the Berkeley permanent-magnet beta spectrographs. These data are given in Table I. ((The information contained in Table I as it pertains to this bibliography is presented as follows:))

	<u>Isotope</u>	<u>Energy (kev)</u>	<u>Lines Seen</u>	<u>Measured from</u>
	Pu-238	$44.11 \pm 0.05$	$L_{II}, L_{III}, M_{II},$ $M_{III}, N_{III}, O$	Cm-242
1.	Cm-242	$42.12 \pm 0.06$	"	Am-242m
2.	Cm-242	$42.18 \pm 0.1$	"	Am-242m
3.7.5.29	PROBABILITIES OF PROMPT NEUTRON EMISSION FROM SPONTANEOUS FISSION. Donald A. Hicks, John Ise, Jr., and Robert V. Pyle (Radiation Laboratory, University of California, Berkeley, California). Physical Review 101, 1016-1020, Jan. 1956			

The neutron number distributions from the spontaneous fission of seven isotopes have been measured by the use of a cadmium plated liquid scintillation tank. The experimental distributions can be roughly approximated by binomial distributions. The average number of neutrons per spontaneous fission have been found to be  $2.30 \pm 0.19$  for Pu-236,  $2.33 \pm 0.08$  for Pu-238,  $2.257 \pm 0.046$  for Pu-240,  $2.18 \pm 0.09$  for Pu-242,  $2.65 \pm 0.09$  for Cm-242 and  $2.84 \pm 0.09$  for Cm-244.

The nuclide to be investigated was mounted as a very thin sample upon a platinum foil which served as the cathode in a 3" diameter parallel plate fission chamber.

- 3.7.5.30 ALPHA-SPECTRA OF HEAVY ELEMENTS. L. L. Gol'din, E. F. Tret'yakov, and G. I. Novikova. p.226-50 in Meetings of the Division of Physical-Mathematical Sciences. Session of the Academy of Sciences of the USSR on the Peaceful Use of Atomic Energy. July 1-5 1955. Moscow, Publishing House of the Academy of Sciences of the USSR, 1955. 376 p. (In Russian)

The alpha particle spectra were studied, utilizing a 50 cm radius precision double-focusing magnetic alpha spectrometer with  $2 \times 10^{-4}$  of 4 pi transmission and 7.5 kev resolution, determined as half-maximum width. The automatic stabilization of coil current secured 0.01% long-time stability. The spectrometer construction was preceded by an extensive study of the theory of particle focusing in magnetic fields with cylindrical symmetry. The spectra of U-233,

U-234, Pu-238, Pu-239, Pu-240, Am-241 and Pa-231 were studied. The alpha spectrum of Pu-238 contains 2 groups: 5490.9 kev (60%) and 5449.9 kev (31%).

- 3.7.5.31 BETA EMITTER Np-238, II SCINTILLATION SPECTROSCOPY IN COINCIDENCE STUDIES. John O. Rasmussen, Frank S. Stephens, Jr., Donald Strominger (Department of Chemistry and Radiation Laboratory, University of California, Berkeley, California). Physical Review 99, 47-55, July 1955

The relative intensities of electromagnetic radiation from Np-238, determined by scintillation spectroscopy set an upper limit for K capture of K/beta as less than 1%. On the basis of extensive scintillation counter coincidence measurements together with the beta spectroscopic results of the preceding article two alternate decay schemes differing only in minor detail are proposed for Np-238. The levels of Pu-238 involved are three close line ground rotational band members and a cluster of three levels near 1.0 Mev. Two of which appear to belong to the same rotational band. Conversion coefficient determinations permit multi-polarity assignments for a number of the gamma transitions and consequent spin and parity assignments for a number of excited states. Selection rules and other intensity rules involving the Bohr-Mottelson K quantum numbers are tested providing K assignments for levels. There is a high degree of K purity of these states where tests were possible. The question of possible "vibrational" character of the band near 1 Mev is discussed speculatively.

((Figure 3 and Figure 4 presented as a part of this article give possible decay schemes for Np-238 which, in part, include partial decay schemes for Pu-238. It may be of some interest to refer to the original article for examination of these figures.))

- 3.7.5.32 ANL-5420. DECAY SCHEMES OF THE TRANSURANIC ELEMENTS. Eugene L. Church. Argonne National Laboratory, Physics Division, Summary Report, August 1954 - February 1955. Page 35, Section 37.6

Plutonium-238 is a 90 year even-even alpha emitter decaying to U-234. The conversion electrons spectrum of this activity has been previously examined and transitions of  $43.6 \pm 0.3$  and  $100.0 \pm 0.4$  kev recorded in uranium. These transitions are below the threshold for K conversion

and convert only to the L, M and higher shells. In the present experiments conversion lines corresponding to the various L and M subshells were resolved and both transitions found to convert almost entirely to the II and III subshells.

3. 7. 5. 33    NUCLEAR PROPERTIES OF THE PLUTONIUM ISOTOPES  
G. T. Seaborg book, The Actinide Elements, McGraw-Hill Book Company, Inc., 1954

((Chapter 7, under table 7. 1, radioactive properties of plutonium isotopes, the following information is given for Pu-238.)) Half life,  $89.59 \pm 0.37$  years; mode of disintegration radiation and energies, alpha 5.495 (76%) 5.454 (24%) 5.351 (0.15%), gamma 0.044, 0.101, 0.149 and conversion electrons; modes of formation: Np-238 beta-decay, U-238 (alpha, 4n), U-235 (alpha, n), Np-237 (d, n), Cm-242 alpha decay.

Seaborg, James and Ghiorso found that the alpha emitting isotope Cm-242 was produced from the Pu-239 (alpha, n) Cm-242 reaction, first using 32 Mev helium ions in this bombardment. Soon after the former method of production was discovered, these investigators found that the same isotope could be produced as a result of high order neutron reactions during the neutron irradiation of Pu-239. ((The balance of this article presents a short history on the half life determinations, the spontaneous fission rate, etc., of Cm-242.))

Curium-244, Reynolds, Hulet, and Street have identified Cm-244 in a mass spectrographic examination of the curium fraction from a highly neutron irradiated americium sample. ((Article goes on to present a short history of the determination of the various properties of Cm-244.))

3. 7. 5. 34    CORRELATION OF SPONTANEOUS FISSION HALF LIVES.  
M. H. Studier and J. R. Huizenga (Argonne National Laboratory, Lemont, Illinois). Physical Review 96, 545-6, October 1954

Empirical correlations which can be used in predicting the properties of undiscovered elements and isotopes are of great practical value. Many such relationships involving alpha disintegration and spontaneous fission rates have been published. Recently alpha disintegration and spontaneous fission half lives of a number of newly discovered nuclides have been measured. We have observed that linear lines connecting even-even nuclides differing by two Z units and six A units give better extrapolated values of R than linear lines connecting the alpha decay

products. In general alpha disintegration half lives of even-even isotopes of a given element beyond the double closed shell decrease with increasing  $Z^2/A$  values, whereas the spontaneous fission half lives of even-even isotopes go through a maximum with increasing  $Z^2/A$  values. However, it is interesting to note that R values increase with increasing  $Z^2/A$  values for the even-even isotopes for the given element (Table 1 entitled "Alpha Disintegration in Spontaneous Fission Half Lives" give references for these properties of 29 isotopes. Included in these are the alpha disintegration and spontaneous fission half lives for Pu-238, Cm-242 and Cm-244. These are reproduced as follows: Pu-238, alpha half life in years 90, fission half life in years  $4.9 \times 10^{10}$ , fission half life in years/alpha half life in years  $5.4 \times 10^8$ ; Cm-242 alpha half life in years 0.445, fission half life in years  $7.2 \times 10^6$ , fission half life in years/alpha half life in years  $1.6 \times 10^7$ ; Cm-244 alpha half life in years 18.4, fission half life in years  $1.4 \times 10^7$ , fission half life in years/alpha half life in years  $7.6 \times 10^5$ ).

Chapter 20, Slow Neutron and Spontaneous Fission Properties of Heavy Nuclei, by J. R. Huizenga, W. M. Manning, and G. T. Seaborg ((this article is concerned with a description of the spontaneous fission of the various transuranic elements)). The following information is given for Pu-238, Cm-242, and Cm-244. Nuclide Pu-238 fissions per gram hour, 36, half life in years  $5.5 \times 10^{15}$ ; Cm-242 fissions per gram hour  $3 \times 10^{10}$ , half life in years  $6.5 \times 10^6$ ; Cm-244, fissions per gram hour  $(1.4 \pm 0.2) \times 10^{10}$ , half life in years  $1.4 \times 10^7$ .

- 3.7.5.35 THE ALPHA- AND GAMMA-RAY SPECTRA OF Pu-238.  
Frank Asaro and I. Perlman (Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California). Physical Review 94, 381-4, April 1954. ((Also published as UCRL-2419.))

The alpha and gamma spectra of Pu-238 have been studied with an alpha-particle spectrograph and gamma-ray scintillation and proportional counters. Alpha groups of 5.495 (72 per cent), 5.452 (28 per cent), and 5.352 Mev (0.09 per cent) and electromagnetic radiations of 17 (13 per cent),  $43.8 \pm 0.5$  (0.038 per cent),  $99 \pm 2$  (0.008 per cent), and  $150 \pm 2$  kev (0.001 per cent) were observed. Spins and parities are assigned to the energy levels, and the results are evaluated with respect to the developing theory and systematics of complex alpha spectra and excited states of even-even nuclei.

The samples of Pu-238 used in the measurement were made by prolonged neutron irradiation of Am-241. The primary objective of the irradiation was to make Pu-242 through the electron capture branching of 16 hour Am-242m so sizable amounts of this isotope were present. ((Three samples in all were prepared.))

All samples were mounted by vacuum sublimation of plutonium chloride onto a platinum plate which was masked to present a band 1 in x 1/8 in. The alpha particles were caught on a photographic plate and the track count plotted according to position on the plate in order to reproduce the spectrum.

Gamma-ray spectra were measured for the most part with a sodium iodide scintillation counter coupled with either a single channel or multichannel pulse-height analyzer. In some experiments a xenon-filled proportional counter was used to produce the pulse for the analyzer.

((Subjects discussed in detail in this article include the principal alpha groups of Pu-238, the low intensity alpha groups of Pu-238, the gamma rays of Pu-238, the 43.8 kev gamma ray, the L X-rays, the 99 kev gamma rays and the 150 kev gamma rays. Also presented in this article are the decay schemes for Cm-242 and Pu-238. Cited as best value for decay scheme in Rev. Mod. Phys. Vol. 30, #2, pt II, April 1958.))

3.7.5.36 THE RADIATIONS OF Np-238 AND THE HALF LIFE OF Pu-238. Paper 14.2 by A. H. Jaffey and L. B. Magnusson, 1003-5, National Nuclear Energy Series, Division IV, Plutonium Project Record, Vol. 14B, pt II, 1951, paper 6.40 to 22.80

((The half life of Pu-238 as determined by these authors equals 77 years. Authors point out the fact that the half life of Pu-238 had been previously determined as  $89 \pm 9$  years and  $92 \pm 2$  years. These experiments indicate that the Pu-238 half life is probably close to 90 years. If either of these values is correct, then the experimental value ((as determined by these authors)) must be too small.))

3.7.5.37 AECD-3149. SPONTANEOUS FISSION. Emilio Segre. Los Alamos Scientific Laboratory, May 8, 1951. Page 30. ((Also published in Physical Review 86, 21-28, 1952))

This substance was prepared by a (d, 2n) reaction on U-238 and it has a half life of about 60 years. The Pu-238 was evaporated on platinum discs and the effective amount present was measured by observing the slow neutron fission of the contaminated Pu-239 and using the ratio of the alpha activities quoted above. In each of our three samples, we had approximately  $10^{-5}$  grams of Pu-239 and  $2 \times 10^{-8}$  grams of Pu-238 effective. The samples were observed for 830 hours total corresponding to  $18.8 \times 10^{-6}$  gram hours for Pu-238 and  $8.6 \times 10^{-3}$  gram hours for Pu-239. 144 fissions were counted. From this we deduce a spontaneous fission decay constant of  $2.1 \times 10^3$  fissions per gram per second. The possible contribution of other plutonium isotopes to spontaneous fission is negligible, being at the most on the order of 1% of the total observed.

((Note: The half life mentioned above, 60 years, is incorrect and was probably the reference cited in ANL-4286.))

- 3.7.5.38 ENERGY LEVELS OF Pu-238 AND Pu-239 NUCLEI.  
S. A. Baranov, K. N. Shlyagin, Atomic Energy USSR 2,  
50, 1950

((In this article the decay schemes of Cm-242 and Pu-238 are discussed. This article is in Russian, however, and no attempt at translation has been made.))

- 3.7.5.39 HALF LIFE FOR DOUBLE BETA DECAY. C. A. Levine,  
A. Ghiorso and G. T. Seaborg (Department of Chemistry  
and Radiation Laboratory, University of California,  
Berkeley, California). Physical Review 77, 296, 1950.  
(Also published as AECD-2741)

((The subject experiment)) consists of examining uranium samples for 90 year Pu-238 which would come from U-238 by the double beta particle mechanisms since Np-238 is heavier than U-238, which, in turn, is substantially heavier than Pu-238, in the isobaric triplate U-238, Np-238, Pu-238. This chemical method of investigation is particularly applicable to this isobaric triplate because there appears to be no other mechanisms to account for the Pu-238 should it be found.

- 3.7.5.40 LAMS-991. SPONTANEOUS FISSION OF PLUTONIUM-238. John Jungerman. December 19, 1949, Decl.  
Dec. 10, 1955.



A 0.25 microgram sample of Pu-238 was obtained from Dr. J. G. Hamilton. Two samples were used of about 5 million alpha disintegrations per minute. The exact sample strength was  $5.17 \times 10^6$  disintegrations per minute and  $4.93 \times 10^6$  disintegrations per minute. The number of spontaneous fissions per gram times hours is less than  $4.79 \times 10^8$ /half life of Pu-238. Where the half life of Pu-238 is the half life of Pu-238 in years with a probable error of 10%. If the half life of Pu-238 for alpha activity is taken as 60 years, then this gives  $7.96 \times 10^6$  F/gram hours or  $2.22 \times 10^3$  F/gram seconds\*. This result agrees within the error of the experiment with the value obtained at Los Alamos which was  $2.1 \times 10^3$  F/gram seconds if the half life of Pu-238 is 60 years\*. ((The term F stands for fissions.))

((\*The 60 year value presented here is invalid. The presently accepted value for the half life of Pu-238 is  $86.41 \pm 0.3$  years.))

3.7.5.41 ANL-4286. SPONTANEOUS FISSION COUNTING OF URANIUM-236 AND PLUTONIUM-238. A. H. Jaffey and A. Hirsch. Argonne National Laboratory Summary Report January, February, March, 1949. Chemistry Division Section C, May 12, 1949. Decl with deletions Feb. 12, 1957, 45 pgs. Page 42.

A sample containing  $3.22 \times 10^6$  disintegrations per minute of Pu-238 was counted for 202 hours in which 72 spontaneous fission counts were observed. Using the value of 92 years for the Pu-238 half life, this counting rate corresponds to a sample content of  $0.089 \pm 0.003$  micrograms. Since counts due to alpha pile up and neutron fission are negligible, this determination yields a value of  $4.0 \pm 0.34 \times 10^6$  fissions per gram hour for Pu-238. The indicated errors are probable errors which are lower than the hitherto accepted value of  $7.7 \pm 1.2 \times 10^6$  fissions per gram hours. The latter value was based upon an inaccurate value for Pu-238 half life of 60 years and should be corrected to the presently accepted value of 92 years. The correction gives forth the value of  $5.0 \pm 0.8 \times 10^6$  fissions per gram hour which checks within experimental error with the value reported here. The plutonium was prepared by the reaction  $\text{Np-237} (n, \gamma) \text{Np-238}$  and probably contained less than 10% Pu-239 by weight and a negligible amount of Pu-240. Since the Pu-239 spontaneous fission rate was negligible no correction was necessary.

3. 7. 5. 42 UCRL-1243. THE SPECTROMETRIC DETERMINATION OF SOME BETA PARTICLE AND CONVERSION ELECTRON ENERGIES. Grover Davis O'Kelley, A. B. Howard College, 1948. Dissertation. Submitted in partial satisfaction of the requirements for the degree of Doctor of Philosophy in Chemistry in the Graduate Division of the University of California.

Section E. Radiations from Pu-243 and Pu-238. The electron capture in Am-242m has been shown to result in the long lived plutonium isotope Pu-242 while the beta decay of Am-242m forms Cm-242. If the plutonium fraction is separated from a sample of the Am-241 which has been subjected to long neutron radiation, both Pu-242 and Pu-238 are present in about equal amounts by weight, the latter plutonium isotope resulting from alpha decay of Cm-242. Alpha pulse analysis in mass spectrographic measurements of these plutonium samples showed the alpha particles energy of Pu-242 to be 4.88 Mev and the alpha particle half life to be approximately  $5 \times 10^5$  years, in good agreement with the alpha decay scheme systematics. Energy surface considerations in this region indicate beta stability for both Pu-242 and Pu-238. Recently the half life of Pu-238 has been measured as  $89.59 \pm 0.37$  years. The alpha particle energy is 5.47 Mev.

3. 7. 5. 43 RANGE MEASUREMENTS OF ALPHA PARTICLES FROM THE  $94^{239}$  and  $94^{238}$ . O Chamberlain, J. W. Gofman, E. Segre and A. C. Wahal (Department of Chemistry and Physics, University of California, Berkeley, California). Physical Review 71, 529-30, April 1947

The ranges of alpha particles from  $94^{239}$  and  $94^{238}$  have been measured by comparison with polonium alphas and are found to be 3.68 centimeters and 4.08 centimeters in air.

To determine the ranges of alpha particles from  $94^{239}$  and  $94^{238}$  with greater precision than previously reported a direct comparison of these ranges with the ranges of alpha particles from polonium was carried out. The samples of 94 were deposited on platinum by evaporation. They were separated from the bombarded uranium with a very small amount of carrier. The uranium sample was electroplated on a copper disc. The thinness of all these samples is borne out by the small values of the straggling coefficient observed.

### 3.8 CURIUM-242

#### 3.8.1 Nuclear Characteristics

3.8.1.1 Atomic Number - 96

3.8.1.2 Atomic Weight - 242

3.8.1.3 Half Life

(alpha)	162.5 days	Reference 1
(S. F.)	$7.2 \times 10^6$ years	Reference 2

3.8.1.4 Neutrons/spontaneous fission

$2.65 \pm 0.09$	Reference 3
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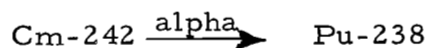
3.8.1.5 Neutrons from spontaneous fission

$2.0 \times 10^7$ neutrons/sec gram	Reference 4
-------------------------------------	-------------

3.8.1.6 Neutrons by ( $\alpha$ , n) with  $\text{Cm}_2\text{O}_3$

$9.5 \times 10^5$ neutrons/sec gram	Reference 4
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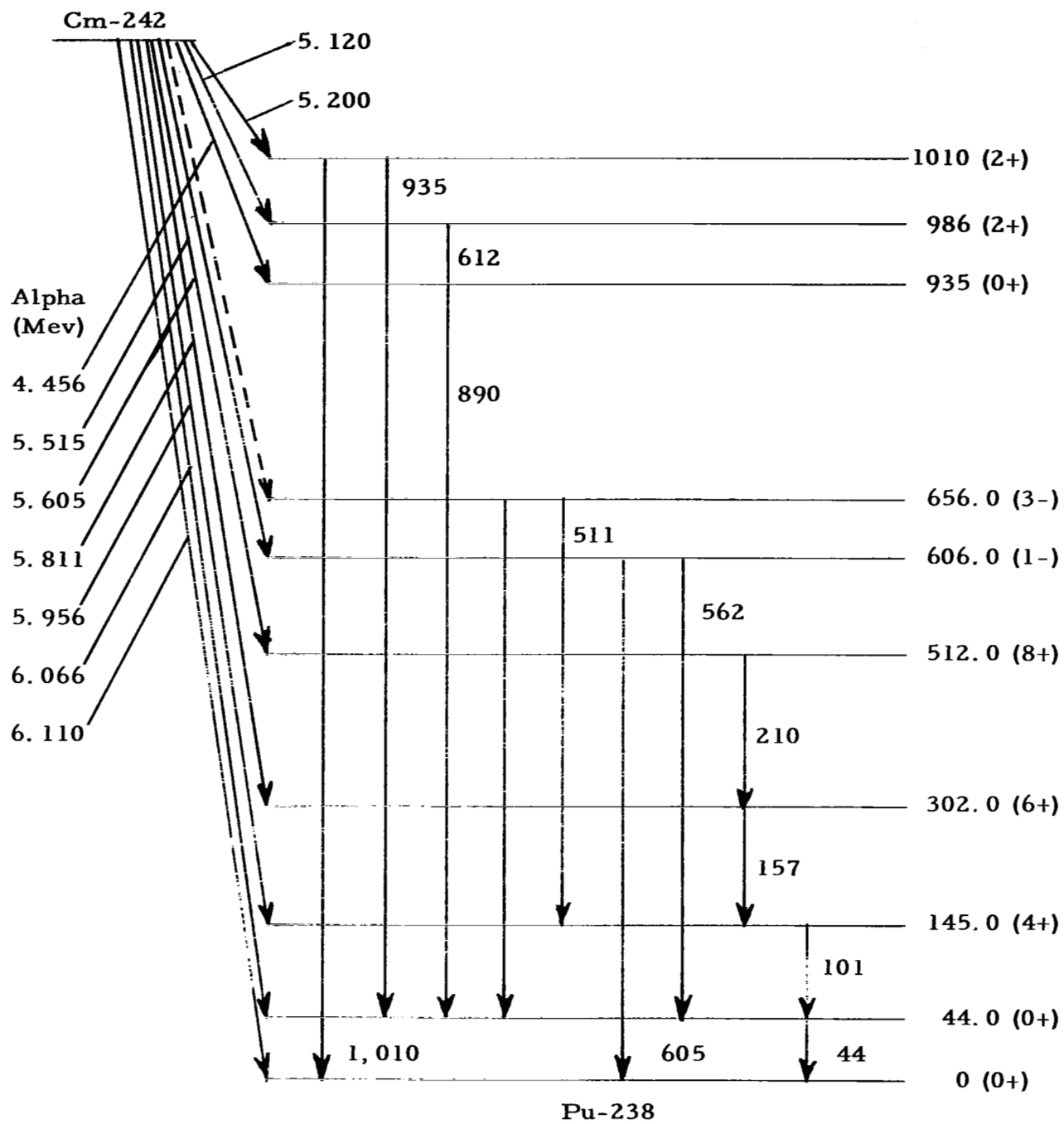
3.8.1.7 Energy Levels and Decay Scheme



Alpha Energies (Mev)	Alpha Particles per Dist. of Isotope (%)	
6.110	73.7	
6.066	26.3	
5.965	$3.5 \times 10^{-2}$	Alpha Energies
5.811	$6 \times 10^{-3}$	Reference 5
5.605	$3 \times 10^{-5}$	
5.515	$1 \times 10^{-4}$	Alphas/
4.465	Unknown	disintegration
5.200	$4 \times 10^{-6}$	Reference 6
5.120	$3 \times 10^{-5}$	

Gamma Energies* (Kev)	Percentage	
1.010	-	
935	$10^{-5}$	Gamma Energies
890	$8.6 \times 10^{-7}$	Reference 5
605	$1.4 \times 10^{-4}$	
562	$1.8 \times 10^{-4}$	Percentages
210	$1.5 \times 10^{-7}$	Reference 6
157	$2 \times 10^{-3}$	
101	$6 \times 10^{-3}$	
44	0.041	

\*It is probable that some of the gamma radiation attributed to decay gamma rays are prompt gamma rays from spontaneous fission. ( $\sim 10$  prompt gamma rays/fission). Private communication, R. S. Rimshaw, Oak Ridge National Laboratories, Sept. 1965.



Refs. 5, 6

### 3.8.1.8 Other Sources of Radiation

In addition to the gamma radiation resulting directly from the decay of Cm-242, other inherent sources of gamma radiation result from spontaneous fissioning. These are (1) prompt gammas associated with spontaneous fissioning, and (2) fission - product gammas. The fission - product gammas in turn come from two distinct sources: (1) from fission products created by spontaneous fissioning of curium, and (2) from fission products which were formed during the irradiation of americium but not removed during the purification process. Other sources of gamma radiation are the isotopes of americium which may amount to 50% of the isotopic mixture and plutonium which remain after purification, and the daughters in the decay chains of curium and other transuranic elements. The latter sources produce insignificant amounts of radiation when compared to the principal sources. The daughters in the Cm-242 chain do not become prominent until after several hundred years of decay.

As with plutonium, a survey of the literature revealed no values for the prompt fission gammas. Therefore, the information from U-235 fissioning (Ref. 7) was adapted to Cm-242 and is presented as Table XXVII.

Values of the fission gammas are also given by reference 8 and are also presented in Table XXVII.

Table XXVII

#### Gamma Radiation Associated with Cm-242

<u>Prompt Fission</u>		<u>Fission Products</u>	
<u>Energy</u> <u>(Mev)</u>	<u>Gammas per</u> <u>Sec. gram</u>	<u>Energy</u> <u>(Mev)</u>	<u>Gammas per</u> <u>Sec. gram</u> (Ref. 7)
0.5	$2.36 \times 10^7$	0.15	$2.05 \times 10^6$
1.0	$1.44 \times 10^7$	0.75	$2.16 \times 10^7$
1.5	$6.78 \times 10^6$	1.20	$4.61 \times 10^6$
2.0	$4.18 \times 10^6$	1.60	$3.05 \times 10^6$
2.5	$2.20 \times 10^6$	2.20	$7.1 \times 10^5$
3.0 up	$2.58 \times 10^6$	2.40	$4.8 \times 10^5$
		2.70	$2.8 \times 10^5$

Table XXVII (continued)

<u>Prompt Fission</u>		<u>Fission Products</u>	
<u>Energy</u> <u>(Mev)</u>	<u>Gammas per</u> <u>Sec. gram</u>	<u>Energy</u> <u>(Mev)</u>	<u>Gammas per</u> <u>Sec. gram</u> (Ref. 8)
1.0	$2.03 \times 10^5$	0.63	$1.8 \times 10^5$
1.5	$5.08 \times 10^4$	1.1	$6.66 \times 10^8$
2.3	$5.38 \times 10^4$	1.55	$7.80 \times 10^4$
3.0	$9.55 \times 10^3$	2.38	$1.50 \times 10^4$
5.0	$1.27 \times 10^4$	2.75	$2.22 \times 10^4$

### 3.8.2 Chemical Characteristics

#### 3.8.2.1 Fuel Forms

The properties of several curium compounds that have been considered for use as isotopic heat sources are presented in Table XXVIII. Where possible the melting points of the compounds are listed. Because the specific power and thermal output for the various compounds depend on the compound density and on the percentage of curium to the total compound weight, these figures are included for each fuel form.

Because the specific power of both Cm-242 and Cm<sub>2</sub>O<sub>3</sub> are so large ( $\sim 1641 \frac{\text{W}}{\text{cc}}$  for Cm-242 and  $\sim 1286 \frac{\text{W}}{\text{cc}}$  for Cm<sub>2</sub>O<sub>3</sub>) they are mixed with "filler" material when used as heat sources to lower their power densities. The preferred diluent is a chemically inert material with a high melting point and high thermal conductivity. The diluent must also be a nuclear inert material in the sense that as small a number as possible of secondary particles such as neutrons or gammas are generated due to nuclear reactions.

Since the maximum allowable specific power is in the vicinity of  $150 \frac{\text{watts}}{\text{cc}}$  a large percentage of diluent must be added and so the Cm-242 or the Cm<sub>2</sub>O<sub>3</sub> will take on the heat transfer characteristics of the matrix material formed. The thermal and physical properties of possible filler materials may be found in Table XXIX.

Table XXVIII(Ref. 9)

## Properties of Curium-242 Compounds

Compound	Molecular Weight	Wt. % Metal	Density g/cm <sup>3</sup>	Sp. Power w/gm	Power Density w/cm <sup>3</sup>	Melting Point °C
Cm	242.0	100.0	13.67	120.1	1641.0	*1340±40
CmO <sub>2</sub>	274.0	88.32	11.68	106.1	1239.0	
Cm <sub>2</sub> O <sub>3</sub>	532.0	91.0	11.77	109.2	1286.0	**9950°
CmB <sub>6</sub>	306.87	78.86	7.31	94.7	692.0	
CmB <sub>4</sub>	285.24	84.84	9.36	101.9	954.0	
CmC	254.01	95.27	13.99	114.4	1600.0	2000°
Cm <sub>2</sub> C <sub>3</sub>	520.0	93.1	12.7	111.8	1419.0	
CmN	256.01	94.53	14.23	113.5	1615.0	
CmSi <sub>2</sub>	298.17	81.2	9.18	97.5	895.0	
CmP	272.97	88.7	10.18	106.4	1084.0	
CmS	274.06	88.3	10.60	106.0	1124.0	
Cm <sub>2</sub> O <sub>2</sub> S	548.06	88.31	9.95	106.03	1055.0	
Cm <sub>2</sub> S <sub>3</sub>	580.19	83.42	8.41	100.2	842.0	
CmF <sub>3</sub>	-	-	-	97.2	943.0	+1410±20

\*B. B. Cunningham and J. C. Wallman. J. Inorg. & Nucl. Chem. 26, 271-5 (1964, also UCRL-11, 213 pg. 90.

\*\*R. E. McHenry and P. Kuehm, Trans. Am. Nucl. Soc. 8 (#1) 75-6, 1965.

+Personal communication with J. Burnett, Oak Ridge National Laboratories, June 1965.

Table XXIX

Fuel Matrix Materials for Cm-242 Compounds  
(Ref. 10 for Columns 1, 4, 5 and Ref. 11 for  
Columns 2, 3)

Material	M. P. (°C)	Specific Heat (Btu/lb°F)	Heat of Fusion (Btu/lb)	Thermal Conductivity (Btu/hr-ft°F)	Density (gm/cc)
Molybdenum	2,620	0.061	126	84.6 ( 68°F)	10.20
Rhodium	1,985	0.059	-	50.0 ( 68°F)	12.50
Zirconium	1,900	0.066	-	15.5 (1200°C)	6.40



Table XXIX (continued)

<u>Material</u>	<u>M. P.</u> <u>(°C)</u>	<u>Specific</u> <u>Heat</u> <u>(Btu/lb°F)</u>	<u>Heat of</u> <u>Fusion</u> <u>(Btu/lb)</u>	<u>Thermal</u> <u>Conductivity</u> <u>(Btu/hr-ft°F)</u>	<u>Density</u> <u>(gm/cc)</u>
Platinum	1,773	0.032	49	42.0 ( 68°F)	21.37
Titanium	1,800	0.126	-	8.1	4.5
Vanadium	1,710	0.120	-	21.3 ( 500°C)	5.96
Palladium	1,553	0.058 (32°F)	61.6	41.0	12.36
Iron (pure)	1,535	0.11	117	46 (1200°C)	7.85
Nickel	1,455	0.105	133	14 (1200°C)	8.9
Copper	1,083	0.092	91.9	210	8.94
Gold	1,063	0.031	129	170 ( 68°F)	19.32
Graphite	3,550			59 - 78.8	2.25
Gadolinium	-	-	-	-	7.95

### 3.8.2.2 Material Compatibility

Although very little unclassified work has been performed in the area of containment materials for curium heat sources, a series of tests has been conducted at the Martin Company (Ref. 12) on gadolinium which exhibits chemical behavior somewhat similar to curium. Data of this type should, of course, be used with caution due to the fact that curium is more reactive than gadolinium.

Tests were conducted at 2000°F in argon with both gadolinium oxide ( $Gd_2O_3$ ) and gadolinium carbide ( $GdC_2$ ). Except for the tests which included iron and nickel, studies were conducted in which pellets of the compound were placed in direct contact with the various metals under investigation. Iron and nickel were tested in cermet form. Compatibility was determined metallographically. Table XXX presents the results of these tests.

Table XXX (ref. 12)

Compatibility of  $Gd_2O_3$  and  $GdC_2$  (substitutes for curium compounds) with metals during 150-hr tests (2000°F in an argon atmosphere)

<u>Material</u>	<u><math>Gd_2O_3</math> Compatibility</u>	<u><math>GdC_2</math> Compatibility</u>
Haynes 25	No attack	No attack
Hastelloy C	No attack	No attack

Table XXX (continued)

<u>Material</u>	<u>Gd<sub>2</sub>O<sub>3</sub> Compatibility</u>	<u>GdC<sub>2</sub> Compatibility</u>
Iconel-X	No attack	No attack
Type 316 Stainless Steel	No attack	No attack
Nickel	No attack	No attack
Iron	Discoloration of oxide	Discoloration of carbide
Columbium	No attack	
Tantalum	No attack	
Molybdenum	No attack	
Tungsten	No attack	

To eliminate side reactions, pellets of gadolinium compound were placed in tubes of the metals (tantalum columbium, and platinum) to be tested; the tubes were evacuated, crimped shut, and seal welded before exposure to elevated temperatures. The result of these experiments are given in Table XXXI.

Table XXXI (Ref. 12)

Compatibility of Gd<sub>2</sub>O<sub>3</sub> and GdC<sub>2</sub> (substitutes for curium compounds) with metals in vacuum environment

<u>Material</u>	<u>Time (hr)</u>	<u>Gd<sub>2</sub>O<sub>3</sub> Compatibility</u>	<u>GdC<sub>2</sub> Compatibility</u>
Molybdenum (arc cast)	64	No attack	No attack
Tungsten (arc cast)	64	No attack	Questionable
Tantalum	64	No attack	Attack
Columbium	112	No attack	Attack
Platinum	112	Pt melted*	Pt melted
Zirconium	64	Badly attacked	Badly attacked

\*Experiments with Gd<sub>2</sub>O<sub>3</sub> in a platinum cermet at 2950°F revealed a dendritic growth of the oxide in the platinum. No serious deterioration of the pellets occurred.

No other information on curium compatibility is reported in the unclassified literature. However, work in this area is presently being undertaken at the Mound Laboratories.

If gadolinium oxide is a valid substitute for the curium fuel form, then Haynes 25, Hastelloy C, Inconel-X, Type 316 stainless steel, nickel, columbium, tantalum, tungsten, molybdenum will be compatible with curium oxide at temperatures up to 2000°F (for at least 150 hrs in an argon atmosphere). At 2950°F arc cast molybdenum, arc cast tungsten, tantalum, columbium, and platinum are compatible with the oxide (for short time periods in a vacuum).

### 3.8.2.3 Effects of Impurities on Nuclear Characteristics

The isotopic purity obtainable with Cm-242 is quite high and is given as 90% by reference 13. As mentioned previously, sources of radiation from americium and plutonium isotopes which remain after purification do contribute some amount to the radiation produced by Cm-242 but are neglected in most shielding calculations. However, fission products produced during the irradiation of americium and removed during the purification process cannot be neglected. This has been reflected in the values of fission product gammas presented in Table XXVII,

### 3.8.3 Thermal and Physical Properties

The thermal and physical properties of Cm-242 are found in Table XXXII. Because Cm-242 is not available in quantity for general use some of the physical and thermal properties of the metal and the oxide are not well known. The oxide (Cm<sub>2</sub>O<sub>3</sub>) and the metal thermal properties are not available and hence the thermal properties of UO<sub>2</sub> and U-238 have been quoted.

The fuel material that has been given most consideration to date has been the oxide Cm<sub>2</sub>O<sub>3</sub>. The metal has not been considered but its physical properties have been included.

Table XXXII

#### Thermal and Physical Properties of Cm-242

		<u>Cm Metal</u>	<u>Cm<sub>2</sub>O<sub>3</sub></u>
Thermal conductivity	$\frac{\text{cal cm}}{\text{sec } ^\circ\text{C Cm}^2}$	0.012(a) Ref. 14	0.014(b) Ref. 15

Table XXXII (continued)

		<u>Cm Metal</u>	<u>Cm<sub>2</sub>O<sub>3</sub></u>
Specific power	$\frac{\text{watts}}{\text{gm}}$	120 Ref. 9, 16	-
Thermal output	$\frac{\text{watts}}{\text{K curie}}$	36.0 Ref. 17	36.0 Ref. 17
Heat capacity	$\frac{\text{cal}}{\text{gm } ^\circ\text{C}}$	0.0456 (407°C)(c) Ref. 15	0.059 (400°C)(a) Ref. 14
Heat of fusion	$\frac{\text{cal}}{\text{mole}}$	--	16,000(d) Ref. 14
Weight density	$\frac{\text{gm}}{\text{cm}^3}$	13.5 Ref. 9	10.7(e) Ref. 18
Melting point	°C	1340±40 Ref. 18	1950 Ref. 9
Specific activity	$\frac{\text{dis/min}}{\text{microgram}}$	7.37x10 <sup>9</sup> Ref. 19	--

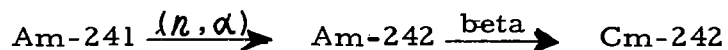
## Key to Table XXXII

- (a) Because the thermal properties of curium metal (oxide) are not presented in the unclassified literature the value for Pu (PuO<sub>2</sub>) has been used.
- (b) This is the value at 400°C for UO<sub>2</sub>, other values are .023 at 100°C and .008 at 1000°C.
- (c) This is the value for plutonium metal at 407°C.
- (d) Value for UO<sub>2</sub>.
- (e) Extrapolated values.

3.8.4 Isotope Production, Availability and Cost

#### 3.8.4.1 Production Method

The method used for the production of Cm-242 is the reactor irradiation of Am-241



The americium is obtained from the beta decay of Pu-241 and mixed with aluminum powder. Irradiation proceeds until a maximum conversion of Am-241 is achieved ( $\sim 40\%$ ). Chemical separation by solvent extraction removes all impurities except the americium. Since the power output per gram of pure curium is much too high for handling during the processing of fuel pellets, it is generally considered unnecessary to separate the americium.

#### 3.8.4.2 Availability and Cost

Prospective production levels of Cm-242 for use in auxiliary power devices have been published by the Division of Isotope Development, USAEC in January 1963. These values are presented below:

	FISCAL YEAR					
	<u>1964</u>	<u>1965</u>	<u>1966</u>	<u>1967</u>	<u>1968</u>	<u>1969</u>
Gm	12	80	1000	1000	1000	1000
Kw(t)	1.5	9.5	120	120	120	120

The cost of Cm-242 is estimated at 12,700 \$/gram at a production rate of 80 grams/year.

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18. B. B. Cunningham and J. C. Wallman. J. Inorg. & Nucl. Chem. 26, 271-5 (1964). Also UCRL 11, 213, pg. 90.
19. ANL-6042. Specific Activity of Nuclides Z 87.

### 3. 8. 5      Annotated Bibliography

3. 8. 5. 1      GAMMA EMISSION FROM Cm-243 AND Cm-242.  
I. A. Baranov, A. S. Krivokhatskii, and A. N. Silant'ev.  
Izv. Akad. Nauk SSSR, Ser. Fiz., 28: 1255-6 (July 1964).  
(In Russian).

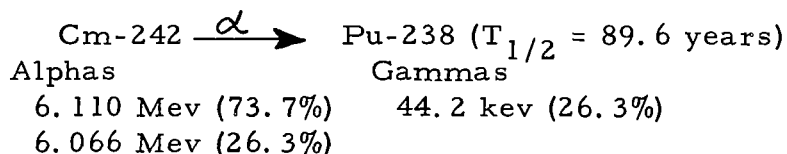
A gamma spectrometer with a resolving power of 10% for 662-keV gamma rays was used for determining the absolute intensity of Cm-243 gamma emission per alpha decay. The alpha-gamma coincidence method permitted reduction of the gamma background to a minimum. The absolute intensity of Cm-243 gamma emission per alpha decay at the gamma-ray energy of 100, 200, and 277 keV was  $(42 \pm 6)$ ,  $(11.3 \pm 1.7)$ , and  $(11.2 \pm 1.7)\%$ , respectively. Investigation of gamma spectra for Cm-242 indicated that the number of gamma quanta per alpha decay at  $E_{\text{gamma}} = 44 \text{ keV}$  was  $(2.7 \pm 0.5) \times 10^{-2}\%$ .

3. 8. 5. 2      Refer to 3. 1. 5. 1. (ORNL-3802) REVIEW OF RADIO-ISOTOPES PROGRAM, 1964. J. H. Gillette - Ingrid B. Lane, comp. (Oak Ridge National Laboratory, Tennessee). May 1965. Contract W-7405-eng-26. 71p.
3. 8. 5. 3      ISOTOPIC POWER DATA SHEETS. S. J. Rimshaw (Oak Ridge National Laboratory, Oak Ridge, Tennessee). For presentation at Industry Information Meeting on Isotopic Power Development and Applications, Washington, D. C. May 18-19, 1964

Source Material              Cm<sub>2</sub>O<sub>3</sub>

Half-Life                      162.5 days

Decay and Radiation  
Properties



Spontaneous fission half-life of Cm-242 =  
 $7.6 \times 10^6 \text{ years}$

Isotopic Composition      > 98% Cm-242.



Activity Concentration	3044 curies per gram of Cm-242 <sub>2</sub> O <sub>3</sub> ; 1218 curies per gram of product (0.4 grams of Cm-244 <sub>2</sub> O <sub>3</sub> + 0.6 grams of Am-241O <sub>2</sub> ).
Radiochemical Purity	>99%
Chemical Purity	40% or less. About 60% Am-241 (T <sub>1/2</sub> = 462 years) will be present.
Specific Power	44.1 watts per gram of Cm-Am oxides (approximate composition - 40% Cm-242 and 60% Am-241), or 36.1 watts per milo-curie of Cm.
Density	Theoretical density is 11.2 g/cm <sup>3</sup> . Practical density is 9.0 g/cm <sup>3</sup> (~ 80% of theoretical density).
Power Density	397 watts/cm <sup>3</sup> for Cm-242 <sub>2</sub> O <sub>3</sub> (40% Cm - 60% Am) with a density of 9 g/cm <sup>3</sup> . About 0.5% of the total power will be contributed b7 Pu-238 and Am-241.
Thermal Conductivity	0.028 watts/cm·°C at 125°C (based on Gd <sub>2</sub> O <sub>3</sub> )
Coefficient of Expansion	10.5 x 10 <sup>-6</sup> /°C (25° to 1000°C) (based on Gd <sub>2</sub> O <sub>3</sub> ).
Melting Point	1950°C in helium
Mechanical Properties	Modulus of elasticity (sonic) 14.5 x 10 <sup>6</sup> lb/in <sup>2</sup> at 25°C (based on Gd <sub>2</sub> O <sub>3</sub> )
Thermal and Radiation Stability	No data available
Radiation Attenuation	Shielding required for gamma radiation is small compared to the neutron shielding required. Neutron dose rate is 0.1 rad/hr at 50 cm for a 2000 thermal watt source shielded with 10 cm of LiH. Additional data are available in ORNL-TM-591 (Rev.) and in ORNL-3576.

Gas Evolution Due to Radioactive Decay Processes	Helium accumulates as a result of alpha decay. An original 1000-curie source of Cm-242 will produce 13.9 cm <sup>3</sup> of He in 162.5 days (1.0 half-life)
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Leach Rate	No data available
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Vapor Pressure	No data available
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Resistance to Thermal Shock	No data available
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Burnup Characteristics	Dispersibility Poor
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Capsule Compatibility	No data available
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- 3.8.5.4     ALPHA DECAY OF CURIUM ISOTOPES. B. S. Dzhelepov, R. B. Ivanov, E. G. Nedovesov, and V. P. Chechev, J. Exptl. Theoret. Phys, SSSR 45, 1360-1371, November 1963. ((Translated in Soviet Physics JETP, Vol. 18, No. 4, April 1964))

The alpha spectra of the Cm-242-246 isotopes are investigated with a pi  $\sqrt{2}$  magnetic alpha spectrometer. The energies and intensities of the alpha transitions are determined precisely. A possible interpretation of some excited Pu-239 and Pu-241 states resulting from the alpha decay of Cm-243 and Cm-244 are discussed on the basis of the Nilsson model. Decay schemes for Cm-242-246 are presented.

The sources used for measurement of the alpha spectra were prepared by vacuum deposition of active material on glass. In the different runs the dimensions of the sources varied from 0.1 x 10 to 1 x 15 millimeters depending upon the particular experiment. ((Table 1 of this work presents transition energy intensities, and energy levels, for Cm-242 through 246. This table is reproduced in part for Cm-242 and Cm-244.))

TABLE I

<u>Lines in Figures</u>	<u>Transition Energy (kev)</u>	<u>Energy Level (kev)</u>	<u>Transition Intensity (%)</u>
<u>CURIUM-242</u>			
1	$6115 \pm 1$	0	$74.0 \pm 2.0$
2	$6071 \pm 1$	44	$26.0 \pm 0.9$
5	$5971 \pm 1$	146	$0.035 \pm 0.002$
<u>CURIUM-244</u>			
8	$5906 \pm 3$	0	$76.2 \pm 2.0$
10	$5763 \pm 3$	43	$23.8 \pm 0.9$
13	$5666 \pm 3$	142	$0.021 \pm 0.002$
19	$5515 \pm 4$	292	$0.003 \pm 0.001$
3.8.5.5	Refer to 3.10.12. (RSG-A-1) PRELIMINARY REPORT ON PUBLIC HEALTH HAZARD ANALYSES OF SPACE ISOTOPIC POWER SOURCES. J. J. Fitzgerald, Gordon L. Brownell, and N. Irving Sax - Jack J. Gabay, ed. (New York, State Department of Health, Albany). Mar. 1, 1964. Contract SAPH 76262. 87p.		
3.8.5.6	(ORNL-TM-1047(p. 62-8)) THE OAK RIDGE NATIONAL LABORATORY CURIUM PROGRAM. E. Lamb (Oak Ridge National Laboratory, Tennessee).		

The recovery and purification of Cm-242 and Cm-244, fabrication of isotopic power sources containing curium, and their characterization with respect to normal operational and extreme conditions are discussed. The operations involved in the preparation of Cm-242 heat sources are Am-241 target fabrication, reactor irradiation, recovery, and purification of Cm-242-Am-241 from irradiated targets, and preparation of the source form. A flowsheet for the preparation of Cm-242 heat sources, target capsule assembly, irradiation assembly, Tramex process flow-sheet, source fabrication facility and Cm-242 power source program source fabrication operations are illustrated.

3. 8. 5. 7 (NP-14688) ISSLEDOVANIE RADIOAKTIVNOGO RASPADA KYURIYA-242 I KYURIYA-244. (A Study of the Radioactive Decay of Curium-242 and Curium-244). G. G. Akalaev, N. A. Vartanov, and P. S. Samoilov (USSR Sovet Ministrov. Gosudarstvennyi Komitet po Ispol'zovaniyu Atomnoi Energii). (1963). 7p. Dep. (mn).

Electron and gamma spectra of curium isotopes were studied using a 1.5 x 30 mm strip placed on a 0.1 u celluloid film which permitted the recording of electrons starting at 1 kev. The measurements were carried out with a beta spectrometer with double focusing at  $\pi\sqrt{2}$  angle. The general spectra for (0.8 to 120)-kev electrons exhibited 15 electron lines with a strong burst of Auger electrons appearing in  $M_i \rightarrow N_j X_k$  transitions. The Cm-242 isotopes were prepared by Am-241 thermal neutron bombardment in a reactor: Am-241 (n, gamma) Am-242m  $\xrightarrow[16 \text{ hr}]{\beta^-}$  Cm-242. The mean energy of the electron transitions was 44.09 kev. The L and M lines were found and estimated.

3. 8. 5. 8 LONG-RANGE ALPHA PARTICLES IN THE SPONTANEOUS FISSION OF TRANSURANIC ELEMENTS. N. A. Perfilov, Z. I. Solov'eva, R. A. Filov, and G. I. Khlebnikov. p. 145-50 of "Fizika Deleniya Atomnykh Yader". Moscow, Gosatomizdat, 1962.

Investigations were made of spontaneous fission in the transuranic region accompanied by the emission of long-range alpha particles. The samples used were Cm-242 and Pu-240. The alpha particles were detected using emulsions. Energy spectra were obtained for alphas whose energies were greater than 13 Mev. The maxima in these spectra occurred at  $15.5 \pm 1.0$  Mev for Cm and at  $17.0 \pm 0.5$  Mev for Pu. The spectra were extrapolated to lower alpha energies by assuming that the distributions were Gaussian. In this way ratios were determined for the probability of fission accompanied by alphas as compared to the probability for ordinary binary fission. These ratios were:  $1/280 \pm 50$  for Cm-242 and  $1/400 \pm 60$  for Pu-240. These numbers were compared with the ones published for Cf-252. It was found that the published Cf-252 values vary substantially among themselves.

3. 8. 5. 9 ENERGY OF ALPHA PARTICLES FROM SOME CURIUM ISOTOPES. R. B. Ivanov, A. S. Krivokhatskii and B. G. Nedovesov, Izvestia Akad Nauk, USSR 26, 976-8 August 1962

((In this effort the authors determine the energies of alpha particles from Cm-242, 243, 244 by comparing their energies with the energies of alpha groups from Bi-212.))

Bi-212. The measurements were carried out on the double focusing magnetic alpha spectrometer. For determining the energy of the alpha particles from Cm-242 we carried out four series of measurements. In each of these the photographic plate was exposed first to the curium activity and then to the Bi-212 source. The strength of the magnetic yield in each series of measurements was kept constant to within 0.01%. The values of the energy of the two alpha groups from Cm-242 together with the average values taken from references 2, 4, and 5 are shown in Tables I and II as follows:

TABLE I. ENERGY OF ALPHA TRANSITIONS  
IN Cm-242 (kev)

<u>Series #</u>	<u><math>\alpha_o</math></u>	<u><math>\alpha_i</math></u>
1	6114.0	6070.4
2	6115.0	6071.0
3	6115.4	6071.5
4	6114.5	6070.2
Mean	$6115 \pm 1$	$6071 \pm 1$
Data of other Ref.	$6110 \pm 4$	6066

TABLE II. ENERGY OF ALPHA TRANSITIONS  
IN Cm-244 (kev)

<u>Series #</u>	<u><math>\alpha_o</math></u>	<u><math>\alpha_i</math></u>
1	5805	5763
2	5806	5763
Mean	$5806 \pm 2$	$5763 \pm 2$
Data of other Ref.	$5801 \pm 2$	5759

((References 2, 4, and 5 refer to past work in this field.)) From the basis of our data on the alpha particle groups we were able to determine the energy of the transition with an intensity of 0.035% in Cm-242. Our calculations yielded a value of 5971 kev. The energy of the Cm-244 alpha-2 line was determined in the same way as for Cm-242. It proved to be equal to 5666 kev.

3. 8. 5. 10    LONG RANGE PARTICLES FROM NUCLEAR FISSION.  
Ralph A. Nobles (University of California, Los Alamos  
Scientific Laboratory, Los Alamos, New Mexico).  
Physical Review 116, 1508-1513, May 1962

A measurement of the probability of emission of long range particles which are known to be predominantly alpha particles made with a multiple ionization chamber gave the following rates for spontaneous fission ((only those of interest are presented)) Cm-242,  $257 \pm 17$ , Cm-244,  $314 \pm 20$ . ((The balance of this abstract is concerned with isotopes not of interest to this section and has been omitted.))

3. 8. 5. 11    A SURVEY OF RESEARCH ON THE TRANSURANIC  
ELEMENTS. M. B. Waldron (Metallurgy Division AERE,  
Harwell). Contemporary Physics 2, 385-402, June 1961

((This article presents an historical review of work done since 1940 on the transuranic elements. It reviews in part the properties of Cm-242, 244, and Pu-238. It presents half life values, disintegration products, etc., but does not present any detailed references on these figures. This article may be of some interest to those wishing to have a complete, or detailed picture on the history of the physics of these elements.))

3. 8. 5. 12    FLUORESCENCE AND OTHER YIELDS IN THE  $L_{II}$  SHELL  
IN PLUTONIUM. L. Salgueiro, J. G. Ferreira,  
J. J. H. Park, and M. A. S. Ross (Department of Natural  
Philosophy, University of Edinburgh). Proceedings of  
the Physical Society (London) 77, 657-64, March 1961

A study has been made of the L, X-radiations emitted after disintegration of Cm-242 to determine as accurately as possible the fluorescence and other yields of the  $L_{II}$  subshell in plutonium. Observations have been made on the total intensity per disintegration of the L, X-rays by observing the L, X-ray photons in coincidence with alpha particles from Cm-242. On the relative intensity of the L, X-rays from the  $L_{II}$  and  $L_{III}$  subshells the sensitivity of the curve crystal spectrograph used for the latter observations had previously been determined. The experimental results are: fluorescence yield,  $w_2 = 0.413 \pm 0.02$ , Coster-Kronig yield  $F_{2,3} = 0.22 \pm 0.08$ , and Auger yield  $A_2 = 0.37 \pm 0.08$ . The total fluorescence yield of the L shells is  $w = 0.486 \pm 0.01$ . The values of  $w_2$  and  $w$  are independent of previous work on fluorescence yields but for the distribution of yields between  $F_{2,3}$  and  $A_2$ , it was necessary to use an extrapolated value of  $w_3$ .

3. 8. 5. 13 AERE-R-3209. COLLECTED INDEPENDENT FISSION YIELDS FOR THERMAL NEUTRON FISSION OF U-233, Pu-239, and U-235, 14 MEV NEUTRON FISSION OF U-235, PILE NEUTRON FISSION OF U-238, Th-232, AND Am-241, AND SPONTANEOUS FISSION OF Cm-242. I. F. Croall, comp. Jan. 1960, 9p. BIS. (United Kingdom Atomic Energy Authority, Research Group. Atomic Energy Research Establishment, Harwell, Berks, England)
3. 8. 5. 14 Refer to 3. 7. 5. 20. ENERGY LEVELS OF Pu-238 AND Pu-239 NUCLEI-SOVIET RESEARCH ON THE LANTHANIDE AND ACTINIDE ELEMENTS 1949-1957. PART III. Nuclear Chemistry and Nuclear Properties, New York Consultants Bureau, Inc. 1959
3. 8. 5. 15 ZEEMAN INVESTIGATIONS OF CURIUM-242. J. C. Hubbs, R. Marrus and J. O. Winocur (Radiation Laboratory and Department of Physics, University of California, Berkeley, California). Physical Review 114, 586-9, April 1959

The atomic beam magnetic resonance method has been used to investigate 163-day Cm-242. The spin of this even-even nuclide is found to be zero. Four low lying electronic energy levels are found and the Lande factors are measured to be  $g_{J2} = 2.561 \pm 0.003$ ,  $g_{J3} = 2.000 \pm 0.003$ ,  $g_{J4} = 1.776 \pm 0.002$  and  $g_{J5} = 1.671 \pm 0.003$ . No direct measurement can be made of the angular momenta of these levels but other considerations contained in the text make probable the J values indicated in subscripts and arising from the electronic configuration  $(5f)^7 (6d)^1 (7s)^2$ .

3. 8. 5. 16 NUMERICAL SOLUTIONS OF THE CURIUM-242 ALPHA-DECAY WAVE EQUATION. John O. Rasmussen (Department of Chemistry and Radiation Laboratory, University of California, Berkeley, California), and Eldon R. Hansen (Radiation Laboratory, University of California, Berkeley, California). Physical Review 109, 1656, March 1958

Numerical integration of the previously derived alpha-decay wave equation, including electric quadrupole coupling terms, was carried out for Cm-242 including  $l = 0, 2, 4$ , and 6 partial waves. Eight integrations were carried inward in spherical polar coordinates on an

IBM-650 computer with different initial conditions, such that a complete set of eight linearly independent solutions to the system of coupled equations was generated. Eight different linear combinations of this base set were found which satisfy the boundary conditions imposed by experimental Cm-242 alpha-group intensities. Wave amplitudes on a spherical surface near the nucleus are given for all eight cases, and the radial variation throughout the barrier region is given for two cases. The matrix formalism of Froman is employed in another presentation of the results, and a comparison is made with the analogous Froman matrix. By using a modified Froman matrix together with our results, the alpha-wave distributions are calculated for a spheroidal nuclear interaction surface. A discussion is made of conditions imposed by alpha-group intensities, and the quadrupole phase shift problem is considered.

3. 8. 5. 17    ALPHA DECAY OF Cm-242.    L. N. Kondrat'ev, V. B. Dedov, and L. L. Gol'din, *Izvestia Akad Nauk, USSR* 22, 99-100, February 1958

((The purpose of this experiment was the calculation of the intensity of the alpha decay of Cm-242 to the second excited level (4+) and the comparison of the calculated and experimental values. The results of the experiments purported to show that the pronounced divergence between the experimental and calculated values of the intensity of alpha decay to the (4+) level actually exist. Table 1, not reproduced here, presents excitation levels of alpha particles of Cm-242, intensities in per cent, and spin and parities.))

3. 8. 5. 18    Refer to 3. 7. 5. 25.    HALF LIFE OF Pu-238.    *Journal of Inorganic and Nucl. Chem.* 5, 6-11, 1957.
3. 8. 5. 19    THE ENERGY LEVELS OF Cm-242 NUCLEI.    S. A. Buranov and K. N. Sblyugin, *Soviet Journal of Atomic Energy* 1, 51-65, 1956

The alpha decay half life of the isotope Cm-242 is equal to 162.5 days. According to Asaro and others it seems that the alpha spectrum consists of four mono-energetic groups of alpha particle. There exists no information in the literature on the electron spectrum of Cm-242. ((The balance of the article is devoted to a detailed analysis of the decay scheme of Np-239 and Np-238. Included are several figures depicting the beta spectrum of neptunium, the spectrum of conversion electrons, nuclear energy levels, Curie plots, etc. As a part of the discussion on neptunium some information is given on the characteristics of Cm-242 and Pu-238.))



- 3. 8. 5. 20    Refer to 3. 7. 9. 27.    GAMMA RADIATION FROM THE  
DECAY OF PLUTONIUM-238 AND CURIUM-242.   Phil.  
Mag. 1, 981-1002, Nov. 1956
  
- 3. 8. 5. 21    Refer to 3. 7. 9. 28.    FIRST EXCITED STATES OF HEAVY  
EVEN-EVEN NUCLEI.   Phys. Rev. 103, 1590-1, Sept. 1956
  
- 3. 8. 5. 22    ALPHA DECAY OF SPHEROIDAL NUCLEI.   John O.  
Rasmussen (Radiation Laboratory and Department of  
Chemistry, University of California, Berkeley, California),  
and Benjamin Segall (Radiation Laboratory, University of  
California, Berkeley, California).   Physical Review 103,  
1298-1308, September 1956

The consequences of spheroidal deformation of nuclei on the barrier transmission in alpha decay are considered.   A set of coupled differential equations is derived relating the amplitudes of the various groups of alpha particles emitted from a nucleus described by the Bohr-Mottelson model.   The cases of the decay of Th-228 and Cm-242 were studied numerically and from them information regarding the probability distribution of alpha particles on the nuclear spheroidal surface is observed.   It is found that the one body model of an alpha particle in a well does not yield these distributions and it is thus concluded that alpha particle clusters have a short mean free path in nuclear matter.   The shift in the surface distributions of Th-228 and Cm-242 may be explained qualitatively in terms of the order of nucleon orbital filling.   The over-all penetration factors for the spheroidal case are compared with those for the spherical case.   It is found that the resultant enhancement due to the deformation is not nearly as large as that predicted by Hill and Wheeler on the basis of one dimensional approximation.   ((The entire article is devoted to theoretical work, no experimentation is performed.))

- 3. 8. 5. 23    UCRL-3456.   ALPHA DECAY STUDIES IN THE HEAVY  
ELEMENT REGION.   John Phillip Hummel (University of  
California, Radiation Laboratories, Berkeley, California).  
July 3, 1956

Under Table of Contents III.   Experimental Results, B. Gamma rays in the decay of Cm-242, page 42, and C. Alpha Decay of Cm-244, page 45.

Using primarily a 75 centimeter radius of curvature  $60^\circ$ , symmetrical electron magnetic analyzer, the study of the complexity of the following alpha spectra were made: E-253, Cf-246, Cm-244, Am-243, Pu-236, 242, Pa-231, Th-227, 230, Ae-225, At-209, Po-206. An investigation of the gamma rays associated with the following isotopes was also made, Cf-246, Cm-242, 244, Am-243, Pu-236, 242. The abundances of the three gamma rays of Cm-242 were measured. These abundances are as follows:  $2.9 \times 10^{-4}$  (44 kev gamma ray)  $4.1 \times 10^{-5}$  (100 kev gamma ray) and  $1.8 \times 10^{-5}$  (155 kev gamma ray). These abundances are all smaller than those given by Asaro. One notes, however, that the relative abundances of the three gamma rays are the same in both studies. These smaller intensities yield somewhat larger conversion coefficients for the 44 and 100 kev gamma rays than given by Asaro, et al. However, the previous interpretation of these conversion coefficients in terms of gamma ray multi-polarities is not altered by the new intensity.

3. 8. 5. 24    CONVERSION ELECTRON SPECTRA OF Cm-242 AND Cm-244.    W. G. Smith and J. M. Hollander (Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California).    Phys. Rev. 101, 746-50, Jan. 1956

The conversion electron spectra of Cm-242 and Cm-244 have been studied by means of two  $180^\circ$  photographic recording beta-ray spectrographs. Conversion coefficient ratios in the L and M subshells have been measured for several E2 transitions in the decay of these nuclides, and the values so obtained compared with theoretical ratios. Accurate energy determinations of the first three excited states in Pu-238 are reported, and the validity of the Bohr-Mottelson rotational energy formula, including the vibration-rotation interaction term to describe these states is discussed. A more accurate value is reported for the energy of the first excited state in Pu-240. The decay of I-131 is discussed briefly.

The spectrograph sources were prepared by electro deposition of the active materials upon 10 mil or 14 mil platinum wires using a procedure suggested by B. G. Harvey in which  $M(OH)_3$  (where M is the actinide cation) is deposited on the wire cathode of an electrolysis cell using  $NH_4HSO_4$  at pH of 3.6 as the electrolyte. A decay scheme for Cm-242 is presented in Figure 4 of this article.

3.8.5.25 Refer to 3.6.5.33. NUCLEAR PROPERTIES OF THE PLUTONIUM ISOTOPES. McGraw Hill Pb. Co. 1954

3.8.5.26 UCRL-2932. DECAY CHARACTERISTICS OF SOME HEAVY ISOTOPES. Isador Perlman, Frank Asaro, Frank S. Stephens, John P. Hummel, and Richard C. Pilger (University of California Radiation Laboratory Chemistry Division Quarterly Report December 1954-January, February 1955). page 59.

Curium-242. The energy of the E2 transition between the first excited state and ground state of Pu-238 has been determined to be  $44.11 \pm 0.04$  kev from measurement of 7 conversion lines. We have also observed the internal conversion coefficient ratios  $L_{II}/L_{III} = M_{II}/M_{III}$ ;  $M_{II}/M_{III} = 1.2$  in good agreement with the theoretical value of 1.2 calculated for the  $L_{II}/L_{III}$  ratio.

3.8.5.27 Refer to 3.6.5.34. CORRELATION OF SPONTANEOUS FISSION HALF LIVES. Phys. Rev. 96, 545-6, Oct. 1954

3.8.5.28 UCRL-2547. SOME CHEMICAL PROPERTIES OF CURIUM. Darrell Charles Feay (University of California Radiation Laboratory). Thesis, April 12, 1954. III Gamma Radiation from Cm-242, page 15

A sample of curium, see Table I, was prepared for the investigation of the electron spectra from gamma radiation using a double focusing beta ray spectrometer.

TABLE I

Spectrographic Analysis of Sample

Am	0.5% (by weight)
La	0.2
Fe	0.1
Al	0.4

The energies and  $L_1 + L_2 = L_3$  conversion ratios were determined for the two most abundant transitions of 44.1 and 102.0 kev, respectively. Previous work by F. Asaro on total conversion coefficients gives strong evidence that both are of E2 character. Since the  $L_1 + L_2 = L_3$  ratio narrows the possibilities to E1 or E2 for the 44.1 kev gamma

ray and to E2 only for the 102.0 kev gamma ray we may consider the E2 character confirmed. Thus, the decay scheme of Cm-242 is as shown in Figure 1. Some hint of the previously reported 157 kev radiation in this decay was found but remains to be confirmed with a sample of greater intensity.

3. 8. 5. 29    AERE C/R-1373.    THE HALF LIFE OF CURIUM-242.  
Mrs. K. M. Glover and J. Milsted (Atomic Energy  
Research Establishment, Department of Atomic Energy,  
Harwell, Berks).    Feb. 1954

The half life of Cm-242 has been redetermined by accurate alpha counting of a pure curium source over a period of 7 months, after allowing for the growth of the Pu-238 daughter. The value obtained, 162.46 days is in very close agreement with the accepted value of  $162.5 \pm 2$  days, but the precision has been greatly improved. The standard deviation of the half life from the present results;  $\pm 0.27$  days. The curium was produced by the neutron radiation of americium in a pile and was separated from the americium by the following steps: The precipitation as a fluoride with residual americium from a solution from which most of the americium has been oxidized to the fluoride soluble hexavalence state. This was repeated a second time with a reduction of volume. Elution from a zee-karb 225 cation exchange column with 11 normal hydrochloric acid to separate from rare earths. Elution from a similar cation exchange column from 0.25 molar curium nitrate solution adjusted to pH 3.50 carried out at 87° centigrade. The early fractions from the second elution showed no detectable activity, alpha type, other than that of Cm-242 (6.11 Mev). The later results obtained with a proportional gamma counter indicated a maximum americium 241/Cm-242 alpha activity ratio of  $10^{-4}$ . An aliquot containing about  $2 \times 10^7$  alpha disintegrations per minute was placed on a polished platinum disc, evaporated to dryness, and ignited to about 800° centigrade. The resulting source was practically weightless.

3. 8. 5. 30    AERE C/R-1365.    A CALORIMETRIC DETERMINATION  
OF THE HALF LIFE OF CURIUM-242.    W. P. Hutchinson  
and A. G. White (Atomic Research Establishment,  
Department of Atomic Energy, Harwell, Berks), Jan. 1954

Calorimetric measurements on a 2 microgram sample of Cm-242 have confirmed the value of 162.5 days previously published for the half life of this nuclide. The currently accepted value for the half life of

this nuclide is the value of  $162.5 \pm 2$  days, determined by Hanna using a counting procedure on samples from which the Am-241 used for radiation had not been separated.

- 3.8.5.31 AECU-2757. ANGULAR CORRELATION AND COINCIDENCE STUDIES OF ALPHA GAMMA CASCADES FROM Cm-242, Pa-231, and Am-241. Richard Lee Moore, A. B., MA, Dissertation presented in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the Graduate School of the Ohio State University (The Ohio State University 1953)

Under Table of Contents: Chapter 4, Cm-242 Experiments, page 107, 4.1 decay scheme. Cm-242 and Cm-243 have been studied by Asaro. His proposed decay scheme for Cm-242 is shown in Figure 36 ((of this article)). Cm-243 is known to be a prolific emitter of K, X-rays. In the present experiment there was a sufficient amount of Cm-243 present for its K, X-rays to obscure the 100 kev radiation from the second to the first excited state of Cm-242 4.2 source. The source used was evaporated from a 2 molar hydrochloric acid solution onto a thin nylon backing on a circular wire frame about 10 millimeters inside diameter. The diameter of the sources which could be judged from the area of discoloration was not greater than 3 millimeters. The decay rate was  $4.0 \times 10^7$  disintegrations per minute, about 18 microcuries. This rate included the Cm-243 disintegrations whose alphas were estimated to have a relative abundance of not greater than 1%. The source was not as thick for the alphas as the Pa source and gave an alpha particle energy resolution of 18%. However, this was poorer than that obtained from the thin Pu-239 source.

- 3.8.5.32 THE ALPHA SPECTRA OF Cm-242, Cm-243 AND Cm-244. Frank Asaro, S. G. Thompson and I. Perlman (Radiation Laboratory, Department of Chemistry, University of California, Berkeley, California). Physical Review 92, 694-702, November 1953. ((Also published as UCRL-2193, Ref. 3.8.5.35.))

The alpha and gamma spectra of Cm-242, Cm-243, and Cm-244 have been studied with an alpha particle spectrograph and gamma ray scintillation counter. Cm-242 has alpha groups of 6.110 (73.7%), 6.066 (26.3%) and 5.964 Mev (0.035%) and gamma rays of 44 (0.041%) 100 (0.006%) and 157 kev (0.0027%). Cm-243 has alpha groups of 5.985 (6%), 5.777 (81%) and 5.732 Mev (13%) and gamma rays of 104,

226 and 278 kev in coincidence with the 5.777 Mev alpha group. Cm-244 has alpha groups of 5.798 (75%) and 5.755 Mev (25%). The spectra are discussed relative to alpha decay theory and corresponding excited states reached by beta minus decay processes.

All samples which were used as sources in the spectrograph were prepared by vacuum sublimation. After chemical purification, each sample of curium present in solution as the chloride was evaporated to dryness on a tungsten ribbon. Under vacuum, current was passed through the tungsten ribbon and the curium sublimed onto the two mil thick platinum plate masked to a band 1" x 0.12". When placed in the spectrograph the sample was made to approximate a line source by placing before it a stainless steel plate with a devining slit 1" x 0.018" or 1" x 0.005". ((Article goes into fine detail in a description of the principal alpha groups of Cm-242, Cm-244 and the low energy alpha groups of Cm-242 and the alpha spectrum of Cm-243. Descriptions of decay schemes for Cm-242, 243, and 244 are described. The decay scheme for Cm-242 and Np-238 is presented as Figure 6.))

3.8.5.33 ANL-5024 REVIEW, RADIO CHEMICAL INVESTIGATION OF THE SPONTANEOUS FISSION OF CURIUM-242, Summary Report - October, November, and December 1952. Compiled by J. R. Gilbreath and O. C. Simpson, August 14, 1953. (Argonne National Laboratories, Chemistry Division.) Section 3, Radio Chemistry, page 55. L. E. Glendenin and E. P. Steinberg.

The measurements of the spontaneous fission yields of Cm-242 have been refined and extended and a relatively complete radiochemical yield mass curve has now been established. The data are presented in Table XIII and the total change yields are presented graphically in Figure 14, with the fission yield mass curve for pile neutron fission of Pu-239 shown for comparison and suggested by the previous results. ((Figure 14, not shown here, presents yield mass curves for spontaneous fission of 242.))

TABLE XIII

Spontaneous Fission Yield of Cm-242 (half lives are in hours unless otherwise indicated).

Sr-91, 9.7; Sr-92, 2.7; Mo-99, 67; Ru-103, 40 days;  
Ru-105, 4.4; Ru-104, 1.0 year; Pd-109, 13.1; Pd-112,

21; Cd-115, 2.33 days; Cd-117, 2.83; Sb-127, 93;  
Sb-129, 4.2; Te-131m, 30; Te-132, 77; I-131, 8.0 days;  
I-133, 21; I-134, 52.5 min; I-135, 6.7; Cs-136, 13.7 days;  
Ba-139, 85 min; Ba-140, 12.8 days.

- 3.8.5.34 THE AVERAGE NUMBER OF NEUTRONS EMITTED IN THE SPONTANEOUS FISSION OF Cm-242. F. R. Barclay and W. J. Whitehouse (Atomic Energy Research Establishment, Harwell, Didcot, Berks). Proceedings of the Physical Society (London) A66, 477-53, May 1953

The average number of neutrons emitted in the spontaneous fission of Cm-242 has been measured by a method depending upon the device of counting only those neutrons which are detected after the occurrence of a fission. In this way the effect of neutrons from sources other than spontaneous fission is eliminated. The value found for  $V$  is 3.0 neutrons per fission. The estimated standard error is 10-15%.

The curium used in the experiment was prepared in pile irradiation of Am-241 which was believed to be isotopically pure. The irradiation was comparatively short and only about  $2 \times 10^{-3}$  of the Am-241 was converted. The separation of curium from americium and plutonium was carried on in an ion exchange column and was not complete. Analysis of the product showed that only 92.2-99.7% of its alpha particle activity was due to Cm-242.

- 3.8.5.35 UCRL-2193. THE ALPHA SPECTRA OF CURIUM-242, CURIUM-243 AND CURIUM-244. Frank Asaro, S. G. Thompson, and I. Perlman. (University of California Radiation Laboratory) April 23, 1953

All samples which were used as sources in the spectrograph were prepared by vacuum sublimation. After chemical purification each sample of curium present in solution as the chloride was evaporated to dryness on a tungsten ribbon. Under vacuum, current was passed through the tungsten ribbon and the curium sublimated onto a 2 mil thick platinum plate masked to a band one inch by 0.12 inch. When placed in the spectrograph, the sample was made to approximate a line source by placing before it a stainless steel plate with a defining slit one inch by 0.018 inch or one inch by 0.005 inch.

3. 8. 5. 36 ANL-5000 (Del). MASS DISTRIBUTION IN THE SPONTANEOUS FISSION OF CURIUM-242. L. E. Glendenin, E. P. Steinberg, Summary Report for July, August and September 1952. Chemistry Division, Section C-11. Report compiled by J. R. Gilbreath, and O. C. Simpson (Argonne National Laboratory), page 55, January 21, 1953

Section III Radio Chemistry. The distribution of mass in the spontaneous fission of Cm-242 has been investigated by a radio-chemical determination of the fission yields of 14 nuclides ranging in mass from 91 to 140. The products of spontaneous fission were isolated by standard radiochemical procedures from solution of 1.0 and 1.5 milligrams of 162.5 days Cm-242, emitting  $7.37 \times 10^{12}$  alpha disintegrations/minutes/milligrams and giving  $4.66 \times 10^3$  spontaneous fissions/minutes/milligrams. Isolations of the elements of interest were carried out after appropriate periods of growth from known amounts of Cm-242. The results are shown in Table 10. ((Table 10 gives the following information))

Fission yields in the spontaneous fission of Cm-242:  
Sr-91,  $1.0 \pm 0.3$ ; Sr-92,  $1.1 \pm 0.3$ ; Mo-99,  $6.1 \pm 0.6$ ;  
Ru-105,  $9.8 \pm 0.6$ ; Pd-109,  $3.0 \pm 0.3$ ; Pd-112,  $1.0 \pm 0.1$ ;  
Cd-115,  $0.030 \pm 0.005$ ; Cd-117, less than 0.01; Sb-127,  
 $0.47 \pm 0.1$ ; Sb-129,  $1.2 \pm 0.2$ ; Te-131m,  $1.9 \pm 0.3$ ;  
Te-132,  $4.3 \pm 0.6$ ; Ba-139,  $6.8 \pm 0.4$ ; Ba-140,  $6.1 \pm 0.6$ ;  
Cs-136,  $0.8 \pm 0.1$ . ((The first symbol refers to the  
nuclide. The numbers refer to the fission yield in per  
cent.))

3. 8. 5. 37 THE COMPLEX ALPHA-SPECTRA OF Am-241 AND Cm-242. Frank Asaro, F. L. Reynolds, and I. Perlman (Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California). Physical Review 87, 277-85, July 1952

The alpha-particle spectra of Am-241 and Cm-242 was studied in detail utilizing a 75-cm radius of curvature  $60^\circ$  symmetrical electromagnetic analyzer with photographic plate detection. The radioactive sources containing up to  $3 \text{ ug/cm}^2$  of active atoms in the case of Am-241 were prepared by vacuum sublimation. The average geometry of the spectrograph is about 4 parts in  $10^5$ . The energy dispersion on the photographic plate is about 3.4 kev/mm for 5-Mev alpha-particles,



and the width at half-maximum of these alpha-particle groups is about 7 kev. Six alpha-particle groups were found in Am-241, and their energies and abundances are 5.546 Mev, 0.23 per cent; 5.35, 0.34 per cent; 5.503, 0.21 per cent; 5.476, 84.2 per cent; 5.433, 13.6 per cent; 5.379, 1.42 per cent. In Cm-242 two alpha-particle groups were found whose energies and abundances are 6.110 Mev, 73 per cent, and 6.064, 27 per cent. The alpha-decay scheme is correlated with gamma-rays and conversion electrons observed in this laboratory for both Am-241 and Cm-242. The various alpha-groups are evaluated with respect to the alpha-decay systematics and the degrees of hindrance of the various alpha-transitions are discussed with reference to normal trends in even-even nuclei. It is suggested that the totally different patterns of the spectra of the two nuclides are conditioned by the nuclear types.

((Samples were prepared by vacuum sublimation of curium-chloride from a hot tungsten filament. Average sample sizes were 2 micrograms/cm<sup>2</sup>. The complex structure, and the alpha decay theory of Cm-242 are discussed in some detail.))

- 3.8.5.38 UCRL-1632. INVESTIGATION OF THE ALPHA PARTICLES OF BISMUTH-203 AND CURIUM-242. Section 13, Nuclear Properties and Transformations. Chemistry Division Quarterly Report, September, October and November 1951. (University of California Radiation Laboratory) page 12, January 28, 1952

Curium-242. Nuclear emulsions were also used to determine the number of conversion electrons that accompanied the alpha particle emission of Cm-242. It was found that of 4100 alpha particles observed 942 or 23% were accompanied by conversion electrons. Hence, at least 23% of the alpha particle decay of Cm-242 leads to an excited level of Pu-238. This compares favorably with the presence of a low energy alpha particle group determined by alpha ray spectroscopy to the abundance of approximately 27%. The conversion coefficient of the gamma ray is indicated to be about 100%.

- 3.8.5.39 THE L X-RAY SPECTRA FROM RADIOACTIVE DECAY OF TRANSURANIUM ELEMENTS. G. W. Barton, Jr., H. P. Robinson, and I Perlman (Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California). Physical Review 81, 208-13, 1951 ((Also published as AECD-2494))

A bent crystal X-ray spectrometer is described and some results are given on the analysis of L series X-rays produced in radioactive decay processes of transuranium elements. There is generally good agreement between measured energy values of L series lines and those predicted by the Mosely relationship. The relative intensities of the various lines produced in this case from gamma ray internal conversion are compared with those from uranium excited by electron bombardment and values reported of internal conversion excited X-rays in the region of lead.

Plutonium X-rays from decay of Curium-242. The isotope Cm-242 is an alpha particle emitter with 162 days half life prepared for the present study by the neutron irradiation of the 475 year Am-241. The alpha decay of Cm-242 includes fine structure in which roughly 20% of disintegrations go to the excited state of Pu-238, about 50 kev above the ground state. The accompanying gamma ray transition is largely internally converted in the L shell and the X-rays measured in the study are those resulting from the refilling of these L orbit vacancies.

3. 8. 5. 40 NEW ISOTOPES OF BERKELIUM AND CALIFORNIUM.  
E. K. Hulet, S. G. Thompson, A. Ghiorso, and K. Street,  
Jr. (Radiation Laboratory and Department of Chemistry,  
University of California, Berkeley, California). Physical  
Review 83, 366-7, July 1951

In the course of work on transplutonium isotopes at Berkeley, it has been possible to prepare mixtures of Am-242, Cm-243, and Cm-244 by intensive neutron irradiation of samples originally consisting of the isotope Am-241. The heaviest curium isotopes are useful sources for the preparation of berkelium and californium isotopes heavier than have been previously observed from helium ion and deuteron bombardments of the isotopes Am-241 and Cm-242. A target containing approximately 100 ug of Cm-242,  $\sim$  5 ug Cm-243, and  $\sim$  2 ug of Cm-244, was bombarded with 35-Mev helium ions and 16-Mev deuterons. The resulting californium and berkelium isotopes were chemically separated from each other, from the target materials, and from fission products using the same combinations of precipitation and ion exchange methods as have been reported previously. The final chemical separations were completed approximately 9 hours after the end of the bombardment.

((The balance of this article is concerned with a discussion of the properties of Bk-243, 246, 247, Cm-243, 245 and Cf-245, 246.))

3. 8. 5. 41 UCRL-1365. THE DECAY SCHEMES OF PLUTONIUM-239 AND CURIUM-242. Charles Prohaska, Chemistry Division Quarterly Report: March, April, May 1951 (University of California Radiation Laboratory) June 26, 1951

No alpha gamma coincidences are observed. If any gamma rays are in coincidence with alpha particles they are in too low an abundance to be observed with this apparatus. A considerable number of alpha electron coincidences were observed in Cm-242. These may be resolved in two components, 37.5 kev electrons and 25 kev electrons. These correspond to M and L conversion electrons, respectively, of a 43 kev gamma, the abundance of these coincidences is as follows:

37.5 kev electrons  
0.09 coincidences per alpha  
25 kev electrons  
0.28 coincidences per alpha

This gives an abundance for the 43 kev transition of 0.37 per alpha and a ratio of L conversion/M conversion = 2.9.

3. 8. 5. 42 SPONTANEOUS FISSION IN Cm-242. G. C. Hanna, B. G. Harvey, N. Moss and P. R. Tunncliffe (Atomic Energy Project, National Research Council of Canada, Chalk River, Ontario, Canada). Physical Review 81, 466-7, February 1951

The spontaneous fission rate was measured as 6.2/minute per  $10^8$  Cm-242 alpha disintegrations/minute. The over-all limits of error in this figure should not exceed 2%. Using a value of 162.5 days for the alpha half life, the spontaneous fission half life becomes  $(7.2 \pm 0.2) 10^6$  years.

3. 8. 5. 43 MASS SPECTROGRAPHIC IDENTIFICATION OF Cm-242, AND Cm-244. F. L. Reynolds, F. K. Hulet and K. Street, Jr. (Radiation Laboratory, Department of Chemistry, University of California, Berkeley, California) Physical Review 80, 467, November 1950

((This reference deals entirely with the spectrographic lines identification of Cm-242 and Cm-244 and is largely of historical interest.))

3. 8. 5. 44 THE HALF LIFE OF CURIUM-242. G. C. Hanna, R. G. Harvey and N. Moss (Atomic Energy Project, National Research Council of Canada, Chalk River, Ontario, Canada). Physical Review 78, 617, June 1950

During recent work on the neutron irradiation of americium it was noted that the Cm-242 produced decay at a rate significantly different than that expected from the published half life of 150 days. Accordingly an attempt was made to measure this half life with reasonable precision. Three sources were prepared by evaporation from solution on a mirror finish platinum disc and ignited to red heat. ((As a result of the experiments a best value of 162.7 ± 0.2 days or 162.5 days ± 2 days was obtained for the half life of Cm-242.))

3. 8. 5. 45 THE NEW ELEMENT CURIUM ATOMIC NUMBER 96. G. T. Seaborg, R. A. James and A. Ghiorso, Paper 22.2, The Transuranium Elements Research Papers edited by Glen T. Seaborg, Joseph J. Katz, Winston M. Manning, Part II, Paper 6.40 to 22.80. First Edition, McGraw-Hill Book Co., Inc. 1949

((This article describes the work leading to the discovery of Cm-242 and so is mainly of historical interest.))

3. 8. 5. 46 UCRL-1243. THE SPECTROMETRIC DETERMINATION OF SOME BETA PARTICLE AND CONVERSION ELECTRON ENERGIES. Grover Davis O'Kelley, A.B., Howard College, 1948. Dissertation. Submitted in partial satisfaction of the requirements for the degree of Doctor of Philosophy in Chemistry in the Graduate Division of the University of California. 1948

Section C, Decay Scheme of Cm-242. The nuclide Cm-242, an alpha particle emitter of 163 days half life, can be prepared in microgram quantities by neutron irradiation of Am-241 and the subsequent decay of Am-242m. Absorption experiments indicated L X-rays and a gamma ray of about 50 kev. No hard electromagnetic radiation is detectable. Internal conversion lines can be assigned to the L, M and perhaps the N conversions of a 43 kev gamma ray.

3. 8. 5. 47 AECD-2376: NUCLEAR PROPERTIES OF TRANSPLUTONIUM ISOTOPES. G. T. Seaborg (Argonne National Laboratories, Lemont, Illinois) December 1947, 12 p. (ANL-JJK-14A-161)

Table I. Radioactive properties of transplutonium isotopes. Cm-242; half life  $162 \pm 3$  days, radiation energies, alpha 6.1 Mev (4.75 centimeters). The isotope Cm-242 also emits a small number of 65 kev gamma rays (about 1 per 500 alpha particles) and a small number of conversion electrons and corresponding L X-rays. The isotopic assignment of the radioactivity has been confirmed as a result of the proof that the isotope Pu-238 is its decay product.

3.8.5.48 ISOLATION OF CURIUM. L. B. Werner and I. Perlman, AECD-2148 (BC-74), August 9, 1947, declassified September 25, 1947 for oral presentation before the meeting of the Chemical Society in New York City September 1947

Curium, the fourth of the transuranium elements, has recently been isolated by L. D. Werner and I. Perlman. The isotopes involved use the intensely alpha active isotope Cm-242 of five months half life. The curium was prepared by the irradiation in a high flux pile of a small amount of 500-year Am-241 which had previously been isolated by B. B. Cunningham, L. B. Asprey and A. C. Stewart.

### 3.9 CURIUM-244

#### 3.9.1 Nuclear Characteristics

3.9.1.1 Atomic number - 96

3.9.1.2 Atomic weight - 244

3.9.1.3 Half Life

(alpha) 18.1 years  
(S. F.)  $(1.46 \pm 0.05) 10^7$  years

Reference 1

Reference 2

3.9.1.4 Alphas/spontaneous fission

$(7.43 \pm 0.01) 10^5$

Reference 3

3.9.1.5 Neutrons/spontaneous fission

$2.810 \pm 0.059$

Reference 4

3.9.1.6 Neutrons from spontaneous fission ( $n$  /  
sec gm)

$1.2 \times 10^7$

Reference 5

3.9.1.7 Neutrons by ( $\alpha$ ,  $n$ ) with  $\text{Cm}_2\text{O}_3$   
( $n$  /sec gm)

$4.2 \times 10^5$

Reference 5

3.9.1.8 Energy Levels and Decay Scheme

$\text{Cm-244} \xrightarrow{\alpha} \text{Pu-240}$

Alpha Energies  
(Mev)

Alpha Particles/  
Dist. (%)

5.798

76.7

5.756

23.3

5.658

0.016

5.511

0.004

Reference 6

Gamma Energies  
(kev)

Percentage

150

$1.3 \times 10^{-3}$

99

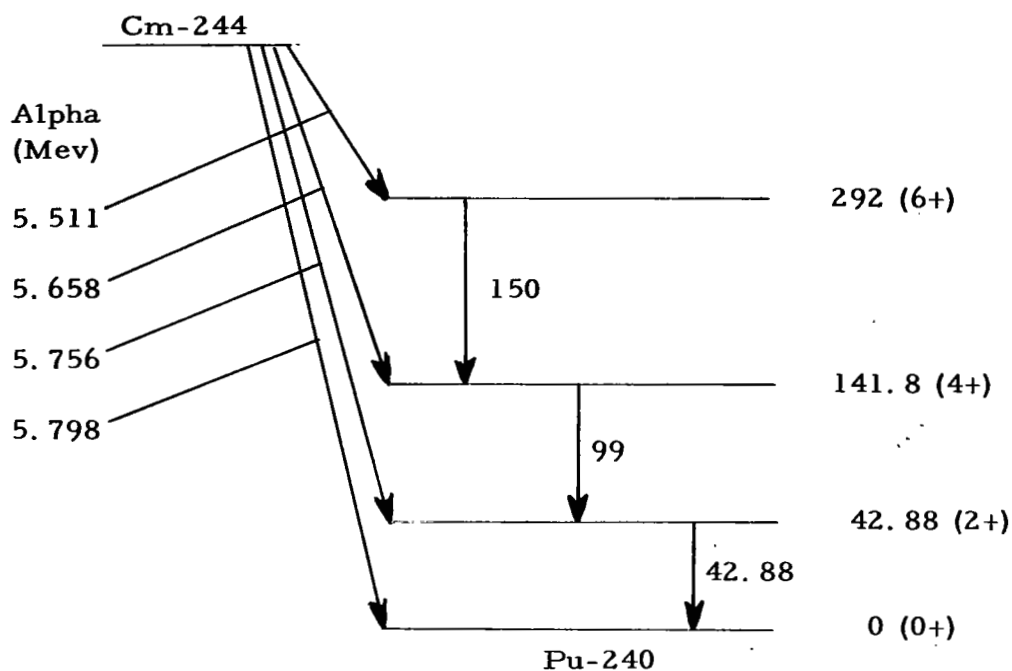
$1.5 \times 10^{-3}$

42.88

$2.1 \times 10^{-2}$

Reference 6

((NOTE)) Some references present a more complex decay scheme for Cm-244. However, gamma radiation attributed to the decay scheme may well be the result of spontaneous fission - Private communication with S. J. Rimshaw, ORNL, 27 June 1965.



Ref. 6

### 3.9.1.9 Other Sources of Radiation

To accurately assess the radiation emanating from Cm-244 all sources of radiation in a so-called Cm-244 product must be examined. The curium product is the material that would be obtained after long term reactor irradiation. Table XXXIII presents an analysis of such Cm-244 product. The composition of this product is taken at one year after reactor irradiation.

Table XXXIII(Ref. 7)

Nuclide	Mass Abundance Weight %
Cm-242	0.02
Cm-243	0.002
Cm-244	95.3
Cm-245	0.9
Cm-246	2.7
Cm-247	0.07
Cm-248	0.05
Am-243	0.5
Cf-252	$4 \times 10^{-5}$

In Table XXXIV the gamma radiation, emanating from the Cm-244 product is presented. This radiation is a result of: the decay of the nuclides, spontaneous fission, fission products, and impurities.

### 3.9.2 Chemical Properties

#### 3.9.2.1 Fuel Forms

The fuel forms of Cm-244 similar to those of Cm-242 (shown previously). The principal difference is in the thermal power. The thermal power of Cm-244 is 2.8 w/gram while for Cm-242 it is 120 w/gram. As with Cm-242 the most probable fuel form based on high melting point and chemical stability is the sesquioxide ( $\text{Cm}_2\text{O}_3$ ). The properties of Cm-244 heat source compounds are presented in Table XXXV. All other remarks made for Cm-242 apply equally to Cm-244.



Table XXXIV (Ref. 7)

## Significant Gamma Rays of Cm-244 Product

Energy Mev	Abundance, photons/(sec)(g Cm-244 product)									
	From Decay of Nuclides			From Spontaneous Fissions			From Fission Products of		From Impurities	Total
	Cm-242	Cm-243	Cm-244	Am-243	Cm-244	Cf-252	Cm-244	Cf-252		
0.0-0.5	$1.1 \times 10^7$	$2.5 \times 10^7$	$6.8 \times 10^8$	$2.8 \times 10^7$	$1.1 \times 10^7$	$1.3 \times 10^6$	$5.9 \times 10^6$	$5.2 \times 10^5$	$6 \times 10^7$	$8.2 \times 10^8$
0.5-1.0	$8 \times 10^4$	-	$5.9 \times 10^6$	-	$7 \times 10^6$	$6.8 \times 10^5$	$1.8 \times 10^7$	$1.6 \times 10^6$	$2 \times 10^7$	$5.3 \times 10^7$
1.0-2.0	$2.4 \times 10^3$	-	-	-	$5.2 \times 10^6$	$4.8 \times 10^5$	$4.1 \times 10^6$	$3.6 \times 10^5$	$3 \times 10^6$	$1.3 \times 10^7$
2.0-3.0	-	-	-	-	$1.6 \times 10^6$	$1.3 \times 10^5$	$1.6 \times 10^6$	$1.4 \times 10^3$	$1 \times 10^6$	$4.3 \times 10^6$
3.0-4.0	-	-	-	-	$4.7 \times 10^5$	$3.4 \times 10^4$	-	-	-	$5.0 \times 10^5$
4.0-5.0	-	-	-	-	$1.6 \times 10^5$	$1.0 \times 10^4$	-	-	-	$1.7 \times 10^5$
5.0-6.0	-	-	-	-	$8.9 \times 10^4$	$2.7 \times 10^3$	-	-	-	$8.9 \times 10^4$
6.0-7.0	-	-	-	-	$1.5 \times 10^4$	$4.0 \times 10^2$	-	-	-	$1.5 \times 10^4$

Table XXXV (Ref. 7)

## Properties of Cm-244 Compounds

	Molecular Weight	Specific Power w/gm	Power Density w/cm <sup>3</sup>
Cm	244.0	2.75	37.6
CmO <sub>2</sub>	278.0	2.42	28.3
Cm <sub>2</sub> O <sub>3</sub>	536.0	2.51	29.5
CmB <sub>6</sub>	308.87	2.17	15.9
CmB <sub>4</sub>	287.24	2.34	21.9
CmC	256.01	2.62	36.7
Cm <sub>2</sub> C <sub>3</sub>	524.03	2.56	32.5
CmN	258.01	2.60	37.0
CmSi <sub>2</sub>	300.17	2.24	20.6
CmP	274.97	2.44	24.8
CmS	276.06	2.43	25.8
Cm <sub>2</sub> O <sub>2</sub> S	552.06	2.43	24.2
Cm <sub>2</sub> S <sub>3</sub>	584.19	2.30	19.3

## 3.9.2.2 Material Compatibility

Chemical compatibility data for Cm-242 applies equally to Cm-244.

## 3.9.2.3 Effects of Impurities on Nuclear Characteristics

The isotopic purity obtainable with Cm-244 is approximately 95% (reference 7). As with Cm-242, minor sources of radiation from americium and plutonium isotopes which remain after purification, contribute only slightly to the radiation produced by Cm-244, and is completely neglected in shielding calculations. However, fission products produced during the irradiation of americium are not insignificant. Therefore, this contribution is included in the values of the fission product gammas presented in Table XXXIV.

3.9.3 Thermal and Physical Properties

The thermal and physical properties of Cm-244 are found in Table XXXVI. The isotope Cm-244 is not available in large quantities and hence some of the physical and thermal properties of

the metal and the oxide are not well known. The thermal properties have proven to be non-existent so the thermal properties of  $\text{PuO}_2$ ,  $\text{UO}_2$ , Pu and U have been quoted.

The only fuel material that has been given serious consideration to date has been the oxide  $\text{Cm}_2\text{O}_3$ . The metal has not been considered but its physical properties have been included.

Table XXXVI

		<u>Cm-244 Metal</u>	<u><math>\text{Cm}_2\text{O}_3</math></u>
Thermal conductivity	$\frac{\text{cal Cm}}{\text{sec } ^\circ\text{C Cm}^2}$	0.012(a) Ref. 8	0.014 Ref. 9
Specific power	$\frac{\text{watts}}{\text{gm (Cm-244)}}$	2.74 Ref. 10	2.3 Ref. 11
Thermal output	$\frac{\text{watts}}{\text{K curie}}$	33.0 Ref. 11	33.0 Ref. 11
Heat capacity at 400°C	$\frac{\text{cal}}{\text{gm } ^\circ\text{C}}$	0.0456(b) Ref. 9	0.059(a) Ref. 9
Heat of fusion	$\frac{\text{cal}}{\text{mole}}$	--	16,000(c) Ref. 9
Weight density	$\frac{\text{gm}}{\text{gm}^3}$	13.5 Ref. 12	10.6 Ref. 7
Melting point	°C	see Cm- 242 Ref. 18	see Cm-242 Ref. 9
Activity	$\frac{\text{dis/min}}{\text{microgram}}$	$1.85 \times 10^8$ Ref. 13	--

Key to Table XXXVI

- (a) Because the thermal properties of Curium metal (oxide) are not well known, the value for Plutonium ( $\text{PuO}_2$ ) has been used.
- (b) This is the value for Plutonium metal at 407°C.

(c) Value for UO<sub>2</sub>.

3.9.4 Isotope Production, Availability and Cost

3.9.4.1 Production Method

Cm-244 is produced by the extended irradiation of Am-241, an isotope resulting from extended irradiation of U-238, and its plutonium products. Because of the number of reactions that occur, the capsules containing the irradiated americium will contain a wide spectrum of materials ranging in atomic weight from about 85 to 245. After separation the curium fraction will contain Cm-242, Cm-243, Cm-244 and Cm-245.

3.9.4.2 Availability and Cost

The Division of Isotope Development, USAEC, published the following prospective production levels and costs in January of 1963.

	FISCAL YEAR							
	<u>1964</u>	<u>1965</u>	<u>1966</u>	<u>1967</u>	<u>1968</u>	<u>1969</u>	<u>1970</u>	<u>1980</u>
Kg	.05	0.1	6	18	40	56	56	58
Kw(t)	.115	.23	13.8	41.3	92	128	128	134

The projected cost of Cm-244 based on a yearly production rate of 50 Kg is 1000 \$/gram.

## REFERENCES

1. Average of best literature values. Private communication with S. J. Rimshaw, Oak Ridge National Laboratories, Sept. 1965.
2. Atomnaya Energiya 15, 3, 249-59, Sept. 1953.
3. Private communication with Mr. H. Diamond and W. C. Bentley, Argonne National Laboratories, Lemont, Illinois, Oct. 1964.
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5. Final Report, Radioisotope Evaluation Program. MNSP-MK-5002. Dec. 23, 1964, Contract NAS 5-3879.
6. Decay Schemes of Radioactive Nuclei. B. S. Dzhelepov and L. K. Peker, Pergamon Press, N. Y. 1961.
7. DPST-64-468. Radiation Properties of Cm-244 Produced for Isotopic Power Generators, D. H. Stoddard. Savanna River Laboratories, Aiken, South Carolina, Sept. 1964.
8. Material for Nuclear Engineers. A. B. McIntosh, T. J. Neal, Interscience Publishers, Inc. 1960.
9. The Metal Plutonium, Coffinberry & Miner. The University of Chicago Press. 1961.
10. ORNL-3576. Handbook of Shielding Requirements and Radiation Characteristics of Isotopic Power Sources for Terrestrial, Marine, and Space Applications. E. D. Arnold April 1964.
11. HW-76323. Rohrmann, Feb. 1963.
12. Nucleonics, Vol. 22 #2, Feb. 1964.
13. ANL-6042. Specific Activity of Nuclides, Z 87.

3.9.5        Annotated Bibliography

- 3.9.5.1      NUCLEAR CONSTANTS OF NINE TRANSPLUTONIUM  
NUCLIDES. D. Metta, H. Diamond, R. F. Barnes,  
J. Milsted, J. Gray, Jr., D. J. Henderson, and C. M.  
Stevens (Argonne National Laboratories, Illinois).  
J. Inorg. Nucl. Chem., 27: 33-9 (Jan. 1965)

The total half lives of Cf-252, Cf-250, Cf-253 are  $2.646 \pm 0.004$  years,  $13.2 \pm 0.5$  years, and  $17.6 \pm 0.2$  days. The ratios of alpha emission to spontaneous fission for Cf-252, Cf-250, Es-253, and Cm-244 are  $31.3 \pm 0.2$ ,  $1260 \pm 40$ ,  $(1.15 \pm 0.03) \times 10^7$ , and  $(7.43 \pm 0.01) \times 10^5$ . The partial spontaneous fission half life of Cm-246 =  $(1.66 \pm 0.10) \times 10^7$  years. The thermal neutron fission cross sections for Cf-249 and Cf-251 are  $1735 \pm 70$  and  $3000 \pm 260$  barns. A separate determination and a re-evaluation of data from the MIKE thermonuclear event confirmed the  $60.5 \pm 0.2$  day Cf-254 half life reported by others.

- 3.9.5.2      Refer to 3.8.5.6. (ORNL-TM-1047(p.62-8) THE OAK  
RIDGE NATIONAL LABORATORY CURIUM PROGRAM.  
E. Lamb (Oak Ridge National Lab., Tenn.)
- 3.9.5.3      DPST-64-468. RADIATION PROPERTIES OF Cm-244  
PRODUCED FOR ISOTOPIC POWER GENERATORS.  
Compiled by D. H. Stoddard (E. I. DuPont, Savannah River  
Lab., Aiken, South Carolina) Sept. 1964

Curium-244 is a candidate heat source for isotopic power generators. This isotope can be produced in large quantities at reasonable cost, and without excessively long lead times. A pilot production program for Cm-244 is underway at the Savannah River Plant. The program not only pilots the large-scale production of curium, but will also provide three kilograms of Cm-244 for the development and demonstration of power generators fueled with this isotope.

This report presents the radiation properties of Cm-244 that would be produced in a large-scale program at the Savannah River Plant. The information is intended for evaluation by users of Cm-244 as a heat source in isotopic power generators. These radiation properties are rather accurate estimates based on:

Measured radiation properties of a purified sample of  $\sim 95$  wt % Cm-244 produced in a Savannah River reactor for the Commission's Transuranium Program.

- Expected purification of the curium product in the chemical separations process being developed.
- Reported radiation properties of the nuclides in the curium product.

Measurements of the radiations of purified Cm-244 will be continued at the Savannah River Laboratory with more sophisticated instrumentation and techniques. These additional measurements are not expected to differ significantly from the present data; however, if discrepancies are noted, they will be reported promptly.

((This document goes into great detail concerning the nuclear properties of Cm-244 and all of its contaminants. Most of the nuclear properties presented in Section 3.9.1 were taken from this report. It does not, however, present any information on chemical or physical properties of Cm-244.))

3.9.5.4 CERTAIN DATA ON Cm-244 SPONTANEOUS FISSION.  
V. I. Bol'shov, L. I. Prokhorova, V. N. Okolovich,  
and G. N. Smirenkin. At. Energ (USSR), 17: 28-34  
(July 1964) (In Russian)

A determination is made of the average kinetic energy of fission fragments  $\overline{E_K} = 182.3 \pm 2.3$  Mev, the width of the distribution for the half height  $\Delta \overline{E_K} = 24.8 \pm 2.5$  Mev, and the average number of prompt neutrons for the spontaneous fission event  $\overline{\nu} = 2.71 \pm 0.04$  in Cm-244. In connection with the experimental results, the question of the dependence of  $\overline{E_K}$  and  $\overline{\nu}$  on the nucleon state of the fissioned nucleus is surveyed. Attention is turned to the correlation of anomalies in these variations with change of the most probable mass of fragments. An assumption of the connection of the observed effects with the change of elastic properties of the produced fragments is evaluated.

3.9.5.5 SPECIFIC IONIZATION ALONG THE TRACK OF U-235 AND Cm-244 FISSION FRAGMENTS. F. Nasyrov. At. Energ. (USSR), 16: 449-51 (May 1964) (In Russian)

A study was made of specific ionization in a mixture of Ar and 5% CH<sub>4</sub> by fission fragments from thermal-neutron fission of U-235 and spontaneous fission of Cm-244. Measurements were performed using a telescope of pulse ionization chambers. Ion pairs formed per unit path length and energy loss per unit length for light and heavy

fission fragments are given for both cases. The specific ionization energy loss dependence on velocity of fragments is presented for U-235 fission. The obtained value of W, energy per ion pair, coincides for both light and heavy groups of fragments.

- 3.9.5.6 SPECTRUM OF ALPHA PARTICLES IN THE Cm-244 SPONTANEOUS TERNARY FISSION. N. A. Perfilov, Z. Soloy'eva, and R. A. Filov. Zh. Eksperim, i. Teor. Fiz. 46: 2244-5 (June 1964) (In Russian)

The energy spectra of alpha particles in the Cm-244 spontaneous ternary fission were measured by means of photographic emulsions and photographic plates. The energy spectra have a Gaussian distribution with a maximum at  $\sim 15.5$  Mev.

- 3.9.5.7 ISOTOPIC POWER DATA SHEETS. S. J. Rimshaw (Oak Ridge National Laboratory, Oak Ridge, Tennessee). For presentation at Industry Information Meeting on Isotopic Power Development and Applications, Washington, D. C. May 18-19, 1964

Source Material	Cm <sub>2</sub> O <sub>3</sub>						
Half-Life	18.4 years						
Decay and Radiation Properties	<p>Cm-244 <math>\longrightarrow</math> Pu-240 (<math>T_{1/2} = 6.6 \times 10^3</math> years)</p> <table> <tr> <td>Alphas</td><td>Gammas</td></tr> <tr> <td>5.798 Mev (76.7%)</td><td>42.9 kev (23.3%)</td></tr> <tr> <td>5.756 Mev (23.3%)</td><td></td></tr> </table> <p>Spontaneous fission half-life = <math>1.4 \times 10^7</math> years</p>	Alphas	Gammas	5.798 Mev (76.7%)	42.9 kev (23.3%)	5.756 Mev (23.3%)	
Alphas	Gammas						
5.798 Mev (76.7%)	42.9 kev (23.3%)						
5.756 Mev (23.3%)							
Isotopic Composition	Mixture of Cm-244 and Cm-245						
Activity Concentration	72.6 curies per gram of Cm-244 <sub>2</sub> O <sub>3</sub>						
Radiochemical Purity	>99%						
Chemical Purity	$\sim 95\%$ with $\sim 5\%$ Am-243 (product specifications not established)						



Specific Power	2.5 watts per gram of curium oxide or 34.28 watts/kilocurie
Thermal Energy	29.2 curies per thermal watt
Density	Theoretical density is $11.2 \text{ g/cm}^3$ . Practical density is $\sim 9.0 \text{ g/cm}^3$ ( $\sim 80\%$ of theoretical density)
Power Density	$22.5 \text{ watts/cm}^3$ for $\text{Cm-244}_2\text{O}_3$ with a density of $9.0 \text{ g/cm}^3$ . About 0.5% of the total power will be contributed by the 5% Am-243
Thermal Conductivity	$0.028 \text{ watts/cm } ^\circ\text{C}$ at $125^\circ\text{C}$ (based on $\text{Gd}_2\text{O}_3$ )
Coefficient of Expansion	$10.5 \times 10^{-6}/^\circ\text{C}$ ( $25^\circ$ to $1000^\circ\text{C}$ ) (based on $\text{Gd}_2\text{O}_3$ )
Melting Point	$1950^\circ\text{C}$ in helium
Mechanical Properties	Modulus of elasticity (sonic) $14.5 \times 10^6$ $\text{lb/in}^2$ at $25^\circ\text{C}$ (based on $\text{Gd}_2\text{O}_3$ )
Thermal and Radiation Stability	No data available
Radiation Attenuation	Neutron shielding is required. See Shielding Handbook by E. D. Arwold, ORNL-3576 for shielding requirements, and also ORNL-TM-591 (Rev.)
Gas Evolution Due to Radioactive Decay Processes	Helium accumulates as a result of alpha decay. An original 1000-curie source of Cm-244 will produce $85.2 \text{ cm}^3$ of helium in two years (0.11 half-lives)
Leach Rate	No data available
Vapor Pressure	No data available
Resistance to Thermal Shock	No data available

Burnup Characteristics      Dispersibility poor

Capsule Compatibility      No data available

- 3.9.5.8      Refer to 3.10.12. (RSG-A-1) PRELIMINARY REPORT ON PUBLIC HEALTH HAZARD ANALYSES OF SPACE ISOTOPIC POWER SOURCES. J. J. Fitzgerald, Gordon L. Brownell, and N. Irving Sax - Jack J. Gabay, ed. (New York. State Department of Health, Albany). Mar. 1, 1964. Contract SAPH 76262, 87p.
- 3.9.5.9      Refer to 3.7.5.8. ALPHA SOURCES FOR LOW THRUST TASKS IN SPACE. International Journal of Applied Radiation and Isotopes. 15, 127-31, March 1964
- 3.9.5.10      SEARCH FOR A SPONTANEOUS FISSION BRANCH IN A METASTABLE STATE OF Cm-244. R. Vandenbosh, P. R. Fields, S. E. Vandenbosh and D. Metta (Argonne National Laboratory, Argonne, Illinois). Journal of Inorganic and Nuclear Chemistry 26, 219-234, 1964

The partial spontaneous fission half life of the 0.034 second ( $I = 6+$ ) metastable state of Cm-244 has been found to be greater than, or equal to,  $1.4 \times 10^2$  years. The life time is not reduced as much as one might expect from the 1.04 Mev excitation energy of the isomeric state. This hindrance may be due to the intrinsic structure of the metastable state.

- 3.9.5.11      Refer to 3.8.5.7. (NP-14688) ISSLEDOVANIE RADIO-KTIVNOGO RASPADA KYURIYA-242 I KYURIYA-244. (A Study of the Radioactive Decay of Curium-242 and Curium-244). G. G. Akalaev, N. A. Vartanov, and P. S. Samoilov (USSR Sovet Ministrov. Gosudarstvennyi Komitet po Ispol'zovaniyu Atomnoi Energii). (1963) 7p. Dep (mn)
- 3.9.5.12      THE ENERGY DISTRIBUTION OF Cm-244 SPONTANEOUS FISSION FRAGMENTS. L. Z. Malkin, I. D. Alkhazov, A. S. Krivoklatskii, K. A. Petrzhak and L. M. Belov, Atomnaya Energiya 15, 3, 249-259, September 1963 (Translated in Atomic Energy USSR 15, 249-50, 1963)

The working specimen spread on a platinum substrate contained 0.243 micrograms of Cm-244, 0.0059 micrograms of Cm-242 and not more

than 0.046 micrograms of Pu-238. When the energy distribution of the Cm-244 spontaneous fission fragments was plotted the contribution from spontaneous fission of Pu-238 was found to be negligible and was left out of the consideration but a correction was made for the contribution from the spontaneous fission of Cm-242. The spontaneous fission period for Cm-242 was assumed to be  $7.2 \times 10^6$  years. ((On the basis of the experiment described in this article the spontaneous fission period was found to be  $(1.46 \pm 0.05) \times 10^7$  years.))

- 3.9.5.13 DECAY OF AN ISOMERIC STATE IN Cm-244. P. G. Hansen and K. Wilsby, Chemistry Department (Research Establishment) Riso, Denmark, Nuclear Physics 45, 410-416, 1963

The 1042 keV state of Cm-244 which is postulated in the beta decay of 10.1 hour Am-244 has previously been assigned  $(K, \pi) = (6, 6+)$ . This assignment has now been confirmed through an angular correlation measurement and it was found that the 746 keV gamma ray is a mixture of 46% quadrupole (E2) and 54% dipole (M1). The half life of the 1042 keV state has been determined by the measurement of delayed coincidences between beta particles and conversion electrons. The result is  $34 \pm 2$  ms. Comparison with the single particle estimate shows that the E2 transitions from the delayed state are hindered by factors of about  $10^{10}$ . This reflects the fact that the transitions are 4 times forbidden in K. The relative transition probabilities for the E2's from the delayed state are shown to agree well with a recent theoretical estimate.

- 3.9.5.14 Refer to 3.4.5.10. EVALUATION OF ISOTOPE THERMIONIC GENERATORS. Robert J. Harvey, Thomas C. Robinson, Joseph J. Fitzgerald, and Gordon Brownell. Transactions of the American Nuclear Society. 6, 88-91, June 1963.
- 3.9.5.15 Refer to 3.8.5.4. ALPHA DECAY OF CURIUM ISOTOPES. J. Exptl. Phys. USSR 45, 1360-71. Nov. 1963
- 3.9.5.16 LIFETIME OF THE FIRST TWO PLUS LEVEL IN Cm-244. Rex Christensen (Institute for Theoretical Physics, University of Copenhagen, Denmark). Nuclear Physics 37, 482-485, 1962

The half life of the 42.9 kev 2+ level in Cm-244 has been measured using the delayed coincidence method. The exponential decay was directly observed in the time spectra. The results obtained in half life equals  $(0.97 \pm 0.05) \times 10^{-10}$  seconds.

- 3.9.5.17 Refer to 3.8.5.9. ENERGY OF ALPHA PARTICLES FROM SOME CURIUM ISOTOPES. Izvestia Akad Nauk, USSR 26, 976-8, Aug. 1962
- 3.9.5.18 Refer to 3.8.5.10. Cm-216 LONG RANGE PARTICLES FROM NUCLEAR FISSION. Phys. Rev. 116, 1508-1513, May 1962
- 3.9.5.19 THE ALPHA HALF LIVES OF Cm-244, Cm-245, and Cm-246. W. T. Carnall, S. Fried, and A. L. Harnkess, (Argonne National Laboratory, Argonne, Illinois). Journal of Inorganic and Nuclear Chemistry 17, 12-14, 1961

The alpha half life of Cm-244 determined from specific activity measurement and the alpha half lives of Cm-245 and 246 determined by mass spectrometric analysis of curium and its plutonium daughters have been determined to be  $17.59 \pm 0.06$  years,  $9320 \pm 280$  years and  $5480 \pm 170$  years, respectively.

- 3.9.5.20 Refer to 3.6.5.34. ABSOLUTE ENERGY MEASUREMENT OF ALPHA PARTICLES FROM Po-210. Phys. Rev. 109, 437-442, Jan. 1958
- 3.9.5.21 UCRL-3456. ALPHA DECAY STUDIES IN THE HEAVY ELEMENT REGION. John Phillip Hummel (University of California, Radiation Laboratories, Berkeley, California) July 3, 1956

#### Section C. Alpha Decay of Cm-244.

- 3.9.5.22 MULTIPLICITIES OF FISSION NEUTRONS. B. C. Diven, H. C. Martin, R. F. Taschek and J. Terrell (Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico). Physical Review, 101, 1012-15, February 1956

((The following is a representation of the abstract as it pertains to Cm-244. Several other fissionable isotopes are described in this article.))

A large liquid scintillator with approximately 80% efficiency for detection of neutrons has been used to obtain data on numbers of neutrons emitted per fission for several fissioning nuclides. Reported here are the average number of neutrons per fission and the respective probabilities of 0, 1, 2... neutrons per fission for spontaneous fission of Cm-244 (and several other isotopes). The value for Cm-244 is  $2.810 \pm 0.059$  neutrons per spontaneous fission. The probabilities of 0, 1, 2... neutrons per fission approach closely a binomial distribution with a maximum number of neutrons equal to 5, 6, or 7, depending upon  $\bar{\nu}$ . ((Table 2 contained in this article presents a summary of the results of the experiments on all of the fissionable materials studied. This table is not reproduced here.))

- 3. 9. 5. 23    Refer to 3. 7. 5. 29.    PROBABILITIES OF PROMPT  
NEUTRON EMISSION FROM SPONTANEOUS FISSION.  
Phys. Rev. 101, 746-50, Jan. 1956
  
- 3. 9. 5. 24    AVERAGE NUMBER OF NEUTRONS PER SPONTANEOUS  
FISSION OF Cm-244. J. M. Higgins, W. W. T. Crane,  
and S. R. Gunn (Radiation Laboratory, Livermore,  
California).    Physical Review 99, 183, July 1955

The average number of neutrons per fission for Cm-244 had been measured by the manganous sulphate moderator absorber system. The value  $2.60 \pm 0.11$  has been determined. A sample of Cm-244 was purified by ion exchange techniques and electroplated on a platinum foil of about 20 square centimeters area. The foil was rolled tightly and the heat from it was measured calorimetrically. The calorimeter used was of the steady state resistance bridge type used at the Mound Laboratory. The accuracy of the present measurement was to within about 1%.

- 3. 9. 5. 25    Refer to 3. 8. 5. 26.    CONVERSION ELECTRON SPECTRA  
OF Cm-242 AND Cm-244. Phys. Rev. 99, 47-55, July  
1955
  
- 3. 9. 5. 26    UCRL-2932.    DECAY CHARACTERISTICS OF SOME  
HEAVY ISOTOPES. (University of California Radiation  
Laboratory Chemistry Division Quarterly Report  
December 1954 - January, February 1955). Isador  
Perlman, Frank Asaro, Frank S. Stephens, John P.  
Hummel, and Richard C. Pilger

Curium-244. The abundances of  $\alpha_0$  and  $\alpha_{43}$  in Cm-244 decay were measured with an alpha particle spectrograph as 76.7 and 28.3, respectively. Their gamma energy separation was 43.0 kev. A new alpha group 142.7 kev lower in decay than the main group was found in an abundance of 0.014%. The ratio of the energies of the second excited state to the first excited state was 3.32 from the Bohr Mottelson theory. The 43- and 143-kev states have spins of 2+ and 4+. The conversion coefficient of the 43 kev gamma ray was measured and indicated the spin of 43 kev state was 2+.

3.9.5.27 Refer to 3.7.5.33. NUCLEAR PROPERTIES OF THE PLUTONIUM ISOTOPES. McGraw-Hill Publishing Co. 1954

3.9.5.28 Refer to 3.7.5.34. CORRELATION OF SPONTANEOUS FISSION HALF LIVES. Phys. Rev. 96, 545-6, October 1954

3.9.5.29 ALPHA HALF LIVES OF Cm-244, Cm-245, AND Cm-246. A. M. Friedman, A. L. Harkness, P. R. Fields, M. A. Studier and J. R. Huizenga, Physical Review 95, 1501, September 1954

The alpha half lives of Cm-244, Cm-245, and Cm-246 measured from mass spectrometric analysis of curium and its plutonium daughters are  $18.4 \pm 0.5$  years (weighted average of present results and those reported in reference 1)\*,  $(1.15 \pm 0.5) 10^4$  years and  $(4.0 \pm 0.6) 10^3$  years, respectively.

The alpha half lives of Cm-244, 245, and 246 have been determined by a technique which depends upon the growth of the plutonium daughters from a curium sample of known isotopic composition.

\*Reference 1 is Physical Review 94, 1083, 1954

3.9.5.30 CURIUM ISOTOPES 246 and 247 FROM PILE-IRRADIATED PLUTONIUM. C. M. Stevens, M. H. Studier, P. R. Fields, P. A. Sellers, A. M. Friedman, H. Diamond and J. R. Huizenga (Argonne National Laboratory, Lemont, Illinois). Physical Review 94, 974, May 1954

Mass spectrometric analyses show the presence of Cm-246 and Cm-247 in curium samples produced from neutron-irradiated plutonium. The

pile-neutron capture cross sections of Am-243, Cm-244, Cm-245, and Cm-246 are  $115 \pm 20$ ,  $25 \pm 10$ ,  $200 \pm 100$ ,  $15 \pm 10$  barns, respectively. The alpha-disintegration half-life of Cm-244 is calculated to be  $19.2 \pm 0.6$  years.

The curium produced by the irradiation of two plutonium samples (total integrated fluxes  $4 \times 10^{21}$  and  $8 \times 10^{21}$  neutrons) in the Materials Testing Reactor (MTR) was chemically purified from fission products and other actinide elements. The plutonium was removed by utilizing its multivalent character. The transplutonium elements were freed of fission products by standard cation resin column techniques. Finally, the curium was separated from americium and the transcurium elements by an ion-exchange column of Dowex 50 resin in the ammonium form eluted with 0.25M citrate solution at a pH of 3.3 at 98°C. The column was 20 cm long with a cross-sectional area of 0.1 square cm. The actinide elements elute at different rates and the various fractions were collected and repurified.

- 3.9.5.31 Refer to 3.8.5.34. THE ALPHA SPECTRA OF Cm-242, Cm-243, Cm-244. Phys. Rev. 92, 694-702, Nov. 1953
- 3.9.5.32 SPONTANEOUS FISSION OF U-234, Pu-236, Cm-240, and Cm-244. A. Ghiorso, G. H. Higgins, A. E. Larsh, G. T. Seaborg, and S. A. Thompson (Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California). Physical Review 87, 163-4, 1952. ((Identical to UCRL-1777.))

((This article consists of an experimental re-determination of the fission half lives of the four isotopes mentioned in the title of this article. The information presented for Cm-244 in the article is as follows: fissions per gram hour  $(1.4 \pm 0.2) 10^{10}$ ; half life in years is  $(1.4 \pm 0.2) 10^7$ .)

- 3.9.5.33 UCRL-1772. SPONTANEOUS FISSION OF URANIUM-234, PLUTONIUM-236, CURIUM-240, AND CURIUM-244. A. Ghiorso, G. H. Higgins, A. E. Larsh, G. T. Seaborg, and S. G. Thompson (University of California Radiation Laboratory). April 21, 1952

Spontaneous fission rates were measured by placing the chemically purified samples on one electrode of a parallel plate ionization

chamber filled with a mixture of argon and carbon dioxide which was connected with an amplifier followed by a register and a stylus recorder. The results are summarized (as follows): Spontaneous fission rates of Cm-240, and Cm-244; Cm-240 fissions per gram hour,  $1.0 \pm 0.2 \times 10^{11}$ , spontaneous fission half life in years,  $1.9 \pm 0.4 \times 10^6$ ; Cm-244 fissions per gram hour  $1.4 \pm 0.2 \times 10^{10}$ , spontaneous fission half life in years,  $1.4 \pm 0.2 \times 10^7$ .

3. 9. 5. 34 UCRL-1365. SPONTANEOUS FISSION OF CURIUM-244. A. E. Larsh, A. Ghiorso, and S. C. Thompson, Quarterly Progress Report Project 48. Nuclear Properties and Transformations. Chemistry Division Quarterly Report: March, April, May, 1951. (University of California Radiation Laboratory). Pages 9 and 10, June 26, 1951

The heaviest isotope of Cm-244 has been shown to undergo spontaneous fission with the extremely high ratio of  $1.3 \pm 0.2$  spontaneous fission per  $10^6$  alpha disintegrations. This value is about twenty times higher per alpha disintegrations than that for Cm-242 known from early work to be 0.062 spontaneous fissions per  $10^6$  alpha disintegrations. The measurements were made on almost pure Cm-244 produced in the Hanford pile by neutron bombardment of Pu-242 through the reaction  $\text{Pu-242} (n, \gamma) \text{Pu-243} \longrightarrow \text{Am-243} (n, \gamma) \text{Am-244} \longrightarrow \text{Cm-244}$ . A small amount of Cm-242 by weight was present in the sample, having been formed by other neutron reactions, but its contribution to the spontaneous fission rate was only about twenty per cent. Assuming a half-life of some forty years as discussed in other sections of this report the half-life of spontaneous fission of Cm-244 is approximately  $3 \times 10^7$  years.



3. 10        General Annotated Bibliography

3. 10. 1        RADIOISOTOPE-FUELED THERMOELECTRIC GENERATORS. William E. Kortier and Meyer Pobereskin, Battelle Tech. Rev., 13: No. 12, 3-8 (Dec. 1964)

The need for dependable, unattended sources of power is being filled increasingly by radioisotope-fueled thermoelectric generators. A discussion of problems involved in their design and some steps being taken for improving such devices is presented.

3. 10. 2        NUCLEAR POWER IN SPACE-RADIOISOTOPE SYSTEMS. J. G. Morse (Martin Co., Baltimore). Raumfahrtforschung, 8: 161-4 (Oct-Dec. 1964)

Radioisotope-fueled thermoelectric generators are today supplying dependable power to satellite systems. Although the technology is relatively new, it has already demonstrated long-term reliability, power-to-weight ratios that are competitive with solar cells, and independence from energy storage in batteries, natural and artificial radiation belts, and solar transients. The development of these power sources from the first unit used in space to those which promise widespread application for the next two or three years is reviewed. The work reported was performed by Martin under the USAEC SNAP Program.

3. 10. 3        ISOTOPIC PROPULSION. John S. Martinez (TRW-Space Technology Labs., Redondo Beach, Calif.). Space/Aeron., 42: No. 6, 77-82 (Nov. 1964)

The use of radioisotopes to heat propellants for space propulsion is discussed, with particular consideration of the Poodle engine now under development. The specific impulse available with gaseous, liquid, and subliming propellants is considered, together with safety and technical problems, thermal control, and isotope selection.

3. 10. 4        POWER SYSTEMS COMPARISON FOR MANNED SPACE STATION APPLICATIONS. Donald I. Southam (Thompson Ramo Wooldridge Inc., Cleveland). New York, American Inst. of Aeronautics and Astronautics, 1964, Paper No. 64-721, 26p. (CONF-640901-5)

From 3rd Biennial Aerospace Power Systems Conference, Philadelphia, Sept. 1964.

A comparison of solar cell, thermionic, mercury Rankine, biphenyl Rankine, steam and Brayton solar systems along with isotope Rankine systems are analyzed at 3, 8, and 20 Kw electric. Component performance, efficiency and weight estimates are provided for each system with consistent basic assumptions and analytical techniques in order to provide comparison validity. Criteria used for the comparisons include present development status, flight availability, system weight, collector area, radiator area, system cost, development cost, efficiency, and orientation requirements.

3. 10. 5      QUARTERLY PROGRESS REPORT, RADIOISOTOPE EVALUATION PROGRAM. V. Truscello, M. Kniedler, T. Bustard (Nuclear Division, Martin Company, Baltimore, Maryland). Report #MNSP-VT-4017. 15 September 1964

This report presents the results of the first three months of a six month study-experimental effort to investigate and evaluate several isotopes as heat sources for scientific satellite power supplies. Isotopes reviewed are Sr-90, Pu-238, Cm-244, Cm-242, Po-210, Ce-144, Pm-147, Cs-134, and Cs-137. Characteristics studied are nuclear and chemical properties, thermal and physical properties, and production and availability.

Experimentation to measure the external bremsstrahlung of Pm-147 in various materials also is described.

3. 10. 6      (BMI-1681(Del)) PROGRESS RELATING TO CIVILIAN APPLICATIONS DURING JULY, 1964. Russell W. Dayton and Ronald F. Dickerson (Battelle Memorial Inst., Columbus, Ohio). Aug. 1, 1964. Contract W-7405-eng-92, 45p.

Progress is reported on development of reactor materials and components, reactor fuel development, radioisotope and radiation applications, development of coated-particle fuel materials, corrosion studies of fluoride-volatility process, gas-cooled reactor development, and development of radioisotopic power sources.

3. 10. 7      (BMI-1674(Del.)) PROGRESS RELATING TO CIVILIAN APPLICATIONS DURING JUNE 1964. Russell W. Dayton and Ronald F. Dickerson (Battelle Memorial Inst., Columbus, Ohio). July 1, 1964. Contract W-7405-eng-92, 46p.

Research and development progress on reactor materials and components, fuel, fuel elements, radioisotope and radiation applications, coated-particle fuel materials, materials development program, fuel irradiation, corrosion studies of the fluoride volatility process, creep-rupture of SAP alloys, mechanical properties of zirconium alloys, fission-product deposition, gas-cooled reactors, and small radioisotope power sources is reported.

3. 10. 8 (TID-3561 (Rev. 4)). SYSTEMS FOR NUCLEAR AUXILIARY POWER (SNAP). A literature search, Henry D. Raleigh, comp. (Division of Technical Information Extension, AEC) June 1964. 80p.

A bibliography of 324 references to publications on the SNAP program and related nuclear-powered generators is presented. The references cover the period January 1956 through April 15, 1964. Author and report number indexes are included.

3. 10. 9 THE SAFETY OF AEROSPACE NUCLEAR SYSTEMS. Douglas G. Harvey (Martin Co., Baltimore). 23 p. (CONF-642-3)

From Health Physics Society, Cincinnati, June 1964.

The safety of nuclear devices for power and/or propulsion aboard space systems is examined with primary emphasis on radioisotope power systems. The procedures and techniques used to analyze the safety of a system are followed from launch phase aborts through re-entry burnup. It is concluded that under the analysis, design, and test process used the potential hazard to the public from aerospace nuclear systems is essentially nil.

3. 10. 10 (A/CONF.28/P/318). RADIOISOTOPE-FUELED THERMO-ELECTRIC GENERATORS. A. N. Voronin, I. G. Gverdsiteli, and R. S. Orinberg (USSR). May 1964. 20p.

The design of a thermoelectric generator operating in vacuum is presented. The description of the isotopic heat source of the thermoelectric generator and the high temperature thermoelectric material base is given. The test method of the device and the test stand description are given. The stand test results (volt-ampere characteristics, temperature distribution, the device activity, etc.) are presented.

3. 10. 11      ORNL-3576 (TID-4500)    HANDBOOK OF SHIELDING  
                         REQUIREMENTS AND RADIATION CHARACTERISTICS  
                         OF ISOTOPIC POWER SOURCES FOR TERRESTRIAL,  
                         MARINE, AND SPACE APPLICATIONS.    E. D. Arnold  
                         (Oak Ridge National Laboratories, Oak Ridge, Tennessee)  
                         April 1964

Isotopic power or radiation sources for use in terrestrial, marine, and space applications require shielding or a degree of isolation to prevent excessive radiation doses to personnel handling the source, to prevent excessive radiation damage to instrument systems associated with the mission or application, and to prevent interference with experimental measurements that use radiation detection instruments. This report is in essence a handbook of radiation properties and shielding requirements for isotopic power or radiation sources and is intended primarily for use in preparation of preliminary design estimates by design engineers in the field of isotopic source development and application. The calculated radiation intensities are probably slightly pessimistic in that they are over-estimated no more than 50%. It therefore may be necessary to optimize the shield for an actual source from experimental data on the source itself. Actual measurements, proving the integrity of a shield, are necessary for licensing by the federal government. Calculations have been made of the radiation intensities from shielded and unshielded sources fabricated from seventeen isotopes that show promise for use in isotopic power or radiation applications. Source sizes in the range of 100 to 20,000 thermal watts were evaluated. All shielded sources were assumed to be attenuated by iron, lead, and uranium; and in those cases where the source also emitted neutrons, neutron and gamma attenuation through water was determined.

The isotopes studied and their physical form are as follows: Co-60 (metal), Kr-85 (liquefied gas), Sr-90 (oxide and titanate), Zr-Nb-95 (oxide), Ru-106 (metal), Cs-137 (glass), Ce-144 (oxide), Tm-170 and Tm-171 (both as oxides), Tl-204 (metal), Po-210 (metal matrix with void space for gas collection), U-232 (oxide), Th-228 (oxide matrix with void space for gas collection), and Pu-238 (oxide), Cm-242 (oxide matrix with void space for gas collection), and Cm-244 (oxide).

For the reader's convenience, several samples of how the graphical results may be used to calculate separation distance, shield thickness, and shield weight are included.

- 3.10.12 (RSG-A-1) PRELIMINARY REPORT ON PUBLIC HEALTH HAZARD ANALYSES OF SPACE ISOTOPIC POWER SOURCES. J. J. Fitzgerald, Gordon L. Brownell, and N. Irving Sax - Jack J. Gabay, ed. (New York. State Department of Health, Albany). Mar. 1, 1964. Contract SAPH 76262. 87p.

A preliminary analysis was made of potential public health problems associated with the large quantities of radioisotopes required for space energy. The properties of likely radioisotopes are considered in some detail. A review was made of prospective NASA space missions to indicate extent to which specific isotopes may be used and the hazards associated therein. External and internal exposures from specific radioisotopes were calculated and specific credible incidents were evaluated. It was concluded that the planned space operations can be conducted at an acceptable level of safety if appropriate consideration is given to the selection of isotopes and if adequate safeguards are provided. Data are appended on the physical, chemical, radiological, and toxicological characteristics of space heat source materials including Sr-90, Y-90, Cs-137, Ce-144, Pr-144, Pm-147, Tm-170, Tm-171, Po-210, Th-228, Pu-236, Pu-238, Cm-242, and Cm-244; the bremsstrahlung production and spectra of Ce-144 and Tm-170; and the pressure build-up from He release. (63 references.)

- 3.10.13 RADIOISOTOPIC SPACE POWER-PROSPECTS AND LIMITATIONS. C. A. Rohrmann and E. D. Sayre (General Electric Company, Richland, Washington). New York, American Institute of Aeronautics and Astronautics, 1964. Paper No. 64-453, 9p. (CONF-600-28)

From American Institute of Aeronautics and Astronautics 1st Annual Meeting, Washington, D. C., June-July 1964. The properties of Co-60, Sr-90, Cs-137, Ce-144, Pm-147, Tm-170, Tl-204, Po-210, Th-228, U-232, Pu-238, Am-241, Cm-242 and Cm-244 are reviewed for use in radioisotope heat sources for direct conversion devices.

- 3.10.14 INDUSTRIAL PARTICIPATION IN NUCLEAR ENERGY. OPPORTUNITIES AND OBSTACLES. p. 72-96 of "Proceedings of the Tenth Anniversary Conference of the Atomic Industrial Forum." New York, Atomic Industrial Forum, 1964

The activities and development of the nuclear industry in 1963 are summarized, and some aspects of commercial production and distribution of radioisotopes are discussed. The performance of SNAP-7 generators for terrestrial uses is summarized, together with the predicted characteristics of future terrestrial SNAP generators. The Marine Products Development Irradiator is briefly described.

3. 10. 15 RADIOISOTOPIC POWER GENERATION. William R. Corliss and Douglas G. Harvey. Prentice-Hall International Series in Space Technology. Englewood Cliffs, N.J., Prentice-Hall, Inc. 1964. 312p.

A survey is presented of general design considerations (radioisotope fuels, safety, energy conversion characteristics, and generator design principles) and specific radioisotope generators (undersea and space units, nuclear batteries, and advanced concepts).

3. 10. 16 (AED-C-15-02). AUSGEWAHLTES SCHRIFTTUM NACH SACHGEBIETEN. KERNABATTERIEN. BIBLIOGRAPHISCHE ZUSAMMENSTELLUNG. (Selected literature according to subject field. Nuclear Batteries. Bibliographic Compilation). Battelle Institute, V., Frankfurtam Main). Sept. 1963. 127p. (In German)

Five hundred and thirty references, covering the period 1959 to 1963, on nuclear batteries are presented. Report number, author, and subject indexes are included.

3. 10. 17 HW-78658. ISOTOPIC SEPARATION AS A FACTOR IN THE SELECTION OF CANDIDATE HEAT SOURCE RADIOISOTOPE. Charles A. Rohrmann (General Electric Company, Hanford Atomic Products Operations, Richland, Washington). September 3, 1963

((This document presents no original information on the nuclear characteristics of radioisotopes.))

3. 10. 18 WASH-1044 REPORTS TO THE AEC NUCLEAR CROSS-SECTION ADVISORY GROUP. Meeting at the University of Colorado, August 13, 14, 1963

((Document is concerned mainly with nuclear cross-sections. However, no significant information is presented for the isotopes under study.))

3. 10. 19 NYO-10689. A BIBLIOGRAPHY ON RADIOISOTOPE POWER SUPPLIES. Daniel M. Azlerod and Joseph P. Novarro, August 1963. 30 pages.

((This article is mainly concerned with the economic aspects of isotope power supplies and the packaging of them.))

3. 10. 20 TID-3561 (rev. 3). DIRECT ENERGY CONVERSION AND SYSTEMS FOR NUCLEAR AUXILIARY POWER (SNAP) A LITERATURE SEARCH. Sidney F. Lanier, Henry D. Raleigh, January 1963

((This document gives references on many aspects of the SNAP program. These are (1) Radioisotope fueled units. (2) Reactor fueled units. (3) Direct energy conversion. (4) General topics on nuclear auxiliary power. For the most part, those references dealing directly with the nuclear characteristics of the various isotopes are contained elsewhere in the bibliography.))

3. 10. 21 NP-11455. DIRECT ENERGY CONVERSION LITERATURE ABSTRACTS. Compiled in the Library Branch Technical Information Division, April 1962. U. S. Naval Research Laboratory, Washington 25, D. C.

((This document contains no direct references concerning the nuclear properties of the isotopes of interest. It is mainly concerned with mechanical assemblies, chemical separation processes, physical preparation processes, and so on.))

3. 10. 22 TID-16939. RELATIVE CROSS-SECTIONS FOR FORMATION FOR THE SHIELDED ISOMERIC CESIUM-134m AND CESIUM-134 IN MEDIUM ENERGY FISSION. R. Vandenbosch and H. Warhanek (Argonne National Laboratories, Lemont, Illinois). July 1962, 18p.

((This article is concerned entirely with nuclear cross-sections.))

3. 10. 23 MND-P-2581-1. ISOTOPIC POWER SOURCES, PROPERTIES, AND PROCESSES REVIEW. A COMPENDIUM. E. H. Smith, William Bowes (The Martin Company, Baltimore, Maryland) June 1961

A selective bibliography has been compiled containing 2063 references to reports and published literature on isotopes usable as power sources. The source was NUCLEAR SCIENCE ABSTRACT, Volumes 1 through 14 (1948 to 1960). Nuclear, physical and chemical properties and separation processes have been emphasized; biological aspects have been excluded. The isotopes covered are Sr-90, Cs-137, Ce-144, Pm-147, Po-210, Ra-228, Ac-227, Th-228, U-232, Np-237, Pu-238, Pu-239, Pu-241, Am-241, Cm-242, Cm-243, and Cm-244. The thoroughness of the search was greatest for the transuranium isotopes, Pm-147, Po-210 and Ac-227 but was deliberately restricted to the most pertinent information about U-232, Th-228 and the fission product elements. The bibliography is arranged according to elements, modified by the number of references included.

((The unrestricted portion of this report is in three volumes. The first volume consists of reference titles and sources only. Volume two (entitled part I) covers the fission products and isotopes with  $(72 < Z < 93)$  and goes into detail concerning the properties described above. Volume three (entitled part II) is devoted to plutonium and the transplutonium elements and goes into the same detail as volume two. The restricted portion of this report was published as MND-P-2581-II.))

3. 10. 24 ARF-1122-27. ISOTOPIC SOURCES OF SECONDARY RADIATION. I. Filosofo. Feb. 1961, 131p.

((This article is primarily concerned with mechanical assemblies using isotopes, and so is not of primary importance to this program.))

3. 10. 25 DECAY SCHEMES OF RADIOACTIVE NUCLEI. Dzhelepov and P. Eker, Pergamon Press, 1961

3. 10. 26 WASH-1029, UC-34 REPORTS TO THE AEC NUCLEAR CROSS-SECTIONS ADVISORY GROUP; ARGONNE NATIONAL LABORATORY. Physics and Mathematics. September 19-21, 1960.

((There is no significant information in this article. Article is concerned mainly with nuclear cross-sections.))

3. 10. 27 MND-P-2374. 13 WATT CURIUM FUELED THERMO-ELECTRIC GENERATOR FOR HARD LUNAR IMPACT MISSION. Final Report sub task 5.8 by J. Bloom. (Physics and Mathematics, Martin Company, Baltimore, Maryland) August 1960.



3. 10. 28 IDO-17648 MTR-ETR. TECHNICAL BRANCHES  
QUARTERLY REPORT. April 1 - June 30, 1960

((This article is concerned for the most part with activation cross-section analysis and is not directly concerned with the physical properties or the half life of isotopes.))

3. 10. 29 IDO-16648 MTR-ETR. TECHNICAL BRANCHES  
QUARTERLY REPORT. April 1 - June 3, 1960.  
Phillips Petroleum Company, Atomic Energy Division  
National Reactor Testing Station, U. S. Atomic Energy  
Commission.

((This article is concerned in part with the half life of Cesium-134m, and with activation cross-sections for Pm-147.))

3. 10. 30 TID-3540. ISOTOPIC POWER AND THERMIONIC  
CONVERSION. A LITERATURE SEARCH. Raymond L.  
Scott, comp. Dec. 1959, 12p.

3. 10. 31 MND-LIB-1631. THERMOELECTRIC POWER SOURCES.  
AN ANNOTATED LITERATURE RESEARCH PART I.  
George E. Halpern and Elizabeth G. Sanford, Dec. 5,  
1958. 24p.

((All applicable references contained in this document are contained elsewhere in the subject report.))

3. 10. 32 TABLE OF ISOTOPES. D. Strominger, J. M. Hollander,  
G. T. Seaborg (Department of Chemistry and Radiation  
Laboratory, University of California, Berkeley, California)  
Reviews of Modern Physics, Vol. 30, Number 2, part II  
585-904, April 1958

((This document contains a listing of all of the radioactive and stable isotopes of the elements, together with a number of their salient features, as recorded in the literature or by private communication to February 1958. Included in the description of each isotope is the following information:

1. Half life
2. Type of decay (including spontaneous fissions)
3. Class and identification

4. Per cent abundance
5. Nuclear moments
6. Energy of radiation
7. Disintegration energy and scheme

The value of this document to the researcher cannot be over-stated.

Although many of the references contained in the Table of Isotopes are also contained in this bibliography, the possession of a copy of the Table of Isotopes by the researcher is almost mandatory.

((Editions of the Table of Isotopes also appeared in:

1. Reviews of Modern Physics 25, 469, 1953
2. Reviews of Modern Physics 20, 585, 1948
3. Reviews of Modern Physics 16, 1, 1944
4. Reviews of Modern Physics 12, 30, 1940

Because the Table of Isotopes appears as an accumulative document, these earlier editions are mentioned only for the sake of completeness.))

3. 10. 33 KAPL-1660. AEC RESEARCH AND DEVELOPMENT REPORT. Physics and Mathematics, M-3679, 18th edition. Knolls Atomic Laboratory, New York. Physics Report of the Research Operation, September, October, November 1956

((The isotopes reference in this document is to Cm-244. However, this isotope was used only as a standardizing isotope for a mass spectrometer that was being calibrated.))

3. 10. 34 NYO-3298. ALPHA GAMMA COINCIDENCES AND ANGULAR CORRELATION IN POLONIUM-210. G. H. Minton, Feb. 20, 1952, 50p.

((This article contains no original information on the nuclear characteristics of polonium as needed for the project.))