BNWL-45

UC-4, Chemistry
(TID-4500, 38th Ed.)

PROMETHIUM-146, FISSION PRODUCT, AND
TRANSURANIUM ISOTOPE CONTENT
OF POWER REACTOR FUELS
Comprehensive Chemical Analyses

By

F. P. Roberts, H. H. Van Tuyl

Chemical Research Section
Chemistry Department

FIRST UNRESTRICTED
DISTRIBUTION MADE

APR 9 '65

March, 1965

BATTELLE-NORTHWEST
RICHLAND, WASHINGTON

Printed by/for the U. S. Atomic Energy Commission

Printed in USA. Price \$1.00. Available from the
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards,
U. S. Department of Commerce,
Springfield, Virginia

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

PROMETHIUM-146, FISSION PRODUCT, AND TRANSURANIUM ISOTOPE CONTENT OF POWER REACTOR FUELS Comprehensive Chemical Analyses

INTRODUCTION

The work described here was directed principally toward determining the amount of Pm^{146} produced in power reactors and was extended to a number of other isotopes as well. Pm^{146} is an undesirable impurity in heat sources of fission product promethium because it decays by emission of moderately energetic gammas and thus increases the shielding requirements of the source. Very small amounts were discovered in highly purified promethium prepared at this laboratory. ^(1, 2) While the concentration of Pm^{146} in promethium from the Hanford and Savannah River production reactors (< 0.5 ppm) is too low to be of serious concern, other workers have predicted that in power reactors the Pm^{146} would be so high as to make this promethium useless as a heat source. ⁽³⁾ The results of this work show this not to be the case.

The nuclear power industry will be the long-term source of large quantities of many fission products and transuranium isotopes. The quantities and qualities of many of these isotopes generated in power reactors are presently being estimated solely by computer techniques. These techniques rely on input data that may be poorly known or extrapolated over wide ranges; gross errors may result.

Chemical analyses for these isotopes in typical reactors are necessary for verification of such computer predictions. Comprehensive analyses of the fission products and transuranium isotopes are reported for a typical pressurized water power reactor and a typical boiling water reactor.

SUMMARY

Four samples of fuel from the Yankee pressurized water reactor and three from the Vallecitos Boiling Water Reactor (VBWR) have been

analyzed radiochemically to determine the isotopic purity of the fission product promethium. Burnups were 13,700 to 31,800 MWd/tonne of uranium for the Yankee fuel and 6540 to 17,400 MWd/tonne* of uranium for the VBWR. Total quantities of a number of other potentially valuable fission products and transuranium isotopes were also determined.

The maximum ratio of Pm^{146} to Pm^{147} found in the Yankee Reactor was 4.7×10^{-6} on a curie basis. Surprisingly, there was little variation between samples representing widely different reactor exposure; the $\text{Pm}^{146}/\text{Pm}^{147}$ ratio evidently reaches an equilibrium value at rather low exposures.

The data obtained should relieve apprehensions concerning long-term availability of high quality (low gamma dose rate) promethium for isotopic heat source applications. Promethium sources containing 4 ppm Pm^{146} will require no more shielding than Pu^{238} for many applications.

The fuel from the VBWR contained significantly less Pm^{146} per curie of Pm^{147} than the Yankee samples. Because of this and the still rather high Pm^{148} content, precise $\text{Pm}^{146}/\text{Pm}^{147}$ measurements have not yet been made. Preliminary data indicate these values will be approximately 1×10^{-6} or about a factor of 4 lower than Yankee. Thus promethium from a VBWR type of reactor may be even more attractive as a heat source material and very nearly as good as that from the Hanford and Savannah River production reactors.

The amounts of Sr^{90} , Cs^{137} , Pm^{147} , Tc^{99} , total palladium, Am^{241} , Cm^{242} , Cm^{244} , Np^{237} as well as the uranium and plutonium isotopes found for these fuels are also reported. Because these isotopes represent truly valuable by-products of atomic power production, their rates of generation must be factored into the design and operation of power reactors and fuel reprocessing facilities. The data presented here can be used for testing the validity of computer techniques used for calculating these rates of generation.

*One tonne = 10^3 kg.

EXPERIMENTAL

SAMPLES

The reactor fuels chosen for analysis were from the Yankee pressurized water reactor and the Vallecitos Boiling Water Reactor. Yankee is typical of pressurized water power reactors and VBWR of boiling water power reactors. The fuel rods of both reactors consist of small pellets of UO_2 . The analytical samples used in this work were single pellets from carefully selected positions in the reactor cores.

The samples from Yankee were from Fuel Element E5, which was irradiated with Core I only, and Fuel Element F5 which was irradiated with both Core I and II. To obtain maximum difference in fission spectrum neutron flux to thermal neutron flux, one sample from a centrally located fuel rod and one sample from a corner rod were taken from each fuel element. All samples were taken from near the midpoint of the fuel rod. Sample designations are as follows:

<u>Sample</u>	<u>Core Location</u>	<u>Fuel Rod Number</u>	<u>Shutdown Date</u>	<u>Approximate Burnup, MWd/tonne</u>
5	E5-C-a6	7992	5/18/62	13,700
2	E5-SW-a5	15025	5/18/62	17,700
3	F5-C-f6	541	9/2/63	24,300
1	F5-SE-f5	8717	9/2/63	31,800

Details of the fuel element locations and irradiation conditions for Core I are given by McGaugh and Chastain. ⁽⁴⁾

Samples of VBWR fuel were selected from a single corner rod, T2D28, on the Fuel Assembly 11J. This rod contained 3.49% U^{235} initially. The position of the fuel element had been changed periodically during the irradiation, but it was always near the center of the core. The irradiation began on 7/16/61 and ended on 12/9/63. Samples taken 12 5/8, 25 3/4, and 30 1/4 in. from the bottom of the rod gave burnups of approximately 17,400, 9900, and 6600 MWd/tonne, respectively. Details of the fuel elements are given by Baroch, Hoffman, and Rous. ⁽⁵⁾

The samples, which consisted of ~ 10 g each of UO_2 , were dissolved under total reflux in concentrated HNO_3 containing a small amount of fluoride ion and then were diluted to known volumes. Analyses for Cs^{137} , uranium, and plutonium were performed immediately to minimize any effects of sample concentration by evaporation. Since the work was to extend over several months, subsequent analyses were accompanied by a redetermination of Cs^{137} , and the sample volume was corrected accordingly.

ISOTOPIC ANALYSIS OF PROMETHIUM

The primary purpose of this work was to determine $\text{Pm}^{146}/\text{Pm}^{147}$ ratios. The Pm^{146} content was expected to be a few parts per million of the Pm^{147} , and therefore its measurement would require a highly efficient decontamination from all other gamma emitting isotopes. Persistent contaminants expected were Ce^{144} , Zr^{95} , Nb^{95} , Eu^{152} , Eu^{154} , Eu^{155} , and Ru^{106} . The procedure used to remove these and other contaminants consisted of:

- (1) An aliquot containing about $1 \text{ mCi}^* \text{ Pm}^{147}$ in 8M HNO_3 was passed through a 1 ml ion-exchange column containing Dowex 1 X-4 in the nitrate form mixed with 50 mg lead dioxide. In this technique⁽⁶⁾ the cerium is absorbed, and the trivalent rare earths are washed through the column with 8M HNO_3 .
- (2) The effluent from the column was diluted to $\sim 1\text{M HNO}_3$ with water. Zirconium, niobium, and lanthanum carriers were added, and the rare earths were precipitated with HF . After being washed, the fluoride precipitate was metathesized to hydroxide and dissolved in a small amount of HNO_3 .
- (3) The promethium was then isolated by cation exchange using a modified ethylenediaminetetraacetic acid elution technique.⁽⁷⁾ The dissolved fluoride precipitate with added neodymium and samarium was absorbed on a 1.87 cm^2 by 20 cm bed of Dowex 50W X-8 (100 to 200 mesh). A second bed of the same resin (0.775 cm^2 by 100 cm) was saturated with yttrium. The columns were eluted in series

*Ci is the recently adopted abbreviation for curie.

at 58 C and 0.7 ml/min flow rate using 0.015 M ethylenediaminetetraacetic acid buffered at pH 8.6 with NH_4OH . Samarium and neodymium separated into individual bands with the promethium "sandwiched" between.

- (4) The Pm^{147} activity was determined by beta proportional counting, and the promethium was then precipitated along with the neodymium and samarium as the fluoride for gamma spectrometry. This procedure resulted in excellent decontamination of the promethium. No gamma emitters could be detected other than Pm^{146} and Pm^{148} .

The Pm^{146} activity was measured by a 5 by 5 in. NaI(Tl) well crystal provided with anticoincidence shielding⁽⁸⁾ and a Model 3412 RIDL 400 channel analyzer. The 0.45 and 0.75 MeV photopeaks of Pm^{146} were well resolved from each other and from the bremsstrahlung originating from the 0.225 MeV beta of Pm^{147} . The disintegration rate of Pm^{146} was calculated using the known detector efficiencies. Appropriate corrections were made for the Pm^{148} activity. Figure 1 shows typical gamma spectra obtained from promethium at various cooling times.

The Pm^{148} activity was determined for those samples that contained sufficient residual activity by a 3 by 3 in. NaI(Tl) solid crystal at a geometry of ~5% and a Model 3412 RIDL 400 channel analyzer.

URANIUM, PLUTONIUM, AND NEPTUNIUM ANALYSIS

Total uranium, plutonium and neptunium analyses were done using standard coulometric techniques with no separation step. For the neptunium analysis it was necessary to evaporate the sample to fumes of H_2SO_4 before titration to get reproducible results.

The isotopic analyses of uranium and plutonium, except Pu^{238} , were done (after separation of plutonium from uranium by a thenoyltrifluoroacetone extraction) using a Model Su-8 Nuclide Analysis Associates, 60°, 12 in. radius, thermal emission mass spectrometer. The Pu^{238} was obtained by alpha pulse height analysis of the freshly separated plutonium using a Frisch-gridded ion chamber.

FIGURE 1

Gamma Spectra of Promethium at Various Cooling Times
 (Source: 0.2 mCi Pm^{147} separated from Yankee Reactor fuel.
 Detector: 5 by 5 in. NaI(Tl) well crystal with anticoincidence shielding.)

AMERICIUM, CURIUM, AND TOTAL PROMETHIUM

Americium, curium, and promethium were separated by elution from a cation exchange column using 0.35M alphahydroxyisobutyric acid at pH 4.5 as the eluant. The Am^{241} , Cm^{242} , and Cm^{244} were determined by alpha pulse height analysis and total alpha counting. The promethium was measured by beta proportional counting. It should be noted that the determination of total promethium requires much less purification than the isotopic analysis.

CESIUM-137

Cs^{137} was measured by the cesium tetraphenylborate extraction and gamma energy analysis which is the standard method for burnup determination in this laboratory. ⁽⁹⁾

STRONTIUM-90

Sr^{90} was determined by loading an aliquot on a cation exchanger and selectively eluting the other nuclides including Y^{90} with 0.7M alphahydroxyisobutyric acid at pH 4. After a suitable growth period, the Y^{90} was again eluted and counted. From this the Sr^{90} content was calculated.

PALLADIUM

The palladium was separated from interferences by anion exchange and measured by flame photometry. An aliquot of the dissolved sample was evaporated to dryness after addition of concentrated HCl and was taken up with 3M HCl. This solution was passed through a column of Dowex 1 X-4. After being washed with 5 column volumes of 3M HNO_3 , the palladium was eluted with 6 column volumes of 8M HNO_3 . The eluate containing the palladium was fumed with HClO_4 containing a small amount of KIO_4 to remove ruthenium and diluted to a known volume. The palladium was then measured with the flame photometer.

TECHNETIUM

The technetium was isolated from other nuclides by a combination of cation exchange and solvent extraction. An aliquot of the sample in 1M HNO_3 was passed through a column of Dowex 50 X-8 to remove uranium, plutonium,

and most of the other cationic constituents. Technetium, which is not absorbed, was then extracted into hexone as the tetraphenylarsonium pertechnetate from 5M NaOH. It was necessary to wash the organic phase with 5M NaOH containing silver oxide to remove $\text{Ag}^{110\text{m}}$ contamination. The Tc^{99} was then determined by beta proportional counting of an aliquot of the organic phase.

RESULTS AND DISCUSSION

PROMETHIUM ISOTOPIC ANALYSIS

The ratios $\text{Pm}^{146}/\text{Pm}^{147}$ and $\text{Pm}^{148}/\text{Pm}^{147}$ for the seven samples are presented in Table I. These ratios are normalized to January 1, 1965, using half-lives as 2.62 yr, 1.94 yr, and 41.8 days for Pm^{147} , Pm^{146} , and Pm^{148} , respectively. Included also in this table are the estimated burnup based on Cs^{137} production and the date the fuel was removed from the reactor. The two aged samples from Yankee did not contain a detectable amount of Pm^{148} . The Pm^{148} in the VBWR samples had not decayed sufficiently to allow a precise Pm^{146} measurement, and the results given are approximations of the upper limit that could be detected. These latter samples will be recounted in 3 mo and an accurate measurement then made.

TABLE I
ISOTOPIC COMPOSITION OF PROMETHIUM

	Sample	Burnup, MWd/tonne	Date of Reactor Shutdown	$\text{Pm}^{146}/\text{Pm}^{147(a)}$	$\text{Pm}^{148}/\text{Pm}^{147(a)}$
Yankee Reactor	Y-5	13,700	5/18/62	$4.1 \pm 1 \times 10^{-6}$	
	Y-2	17,700	5/18/62	$4.0 \pm 1 \times 10^{-6}$	
	Y-3	24,300	9/2/63	$4.7 \pm 1 \times 10^{-6}$	1.1×10^{-4}
	Y-1	31,800	9/2/63	$4.0 \pm 1 \times 10^{-6}$	7.3×10^{-5}
VBWR	V-60	6,500	12/9/63	$\sim 1 \times 10^{-6}$	1.3×10^{-4}
	V-57	9,900	12/9/63	$\sim 1 \times 10^{-6}$	1.5×10^{-4}
	V-55	17,400	12/9/63	$\sim 1 \times 10^{-6}$	1.1×10^{-4}

(a) Curie ratios

Isotope Evolution

Pm^{146} may be formed as a primary fission product or as the result of an (n, 2n) reaction on Pm^{147} . The fission yield of Pm^{146} is about 4×10^{-7} of the 146 chain, and the (n, 2n) cross section of Pm^{147} is ~ 5.9 millibarns. ⁽¹⁾

At high burnup both Pm^{147} and Pm^{146} approach equilibrium contents because of loss by both radioactive decay and neutron capture. Pm^{147} saturates more rapidly in Yankee than in VBWR because of the high resonance cross section of Pm^{147} and the harder spectrum of Yankee. The Pm^{147} content at saturation is higher than might be expected in Yankee because the selective flux depletion at 5.49 eV, caused by the resonance absorption of U^{236} , results in a somewhat lower burnout cross section for Pm^{147} . The half-life of Pm^{147} is 2.62 yr, ⁽¹⁰⁾ and that of Pm^{146} is reported variously as 1.94 ⁽¹¹⁾ and 4.4 yr. ⁽¹²⁾ The thermal neutron cross section of Pm^{147} is 235 barns, ⁽¹³⁾ and the resonance integral cross section is 3220 barns. The cross section of Pm^{146} for a reactor spectrum is 8400 barns. ⁽¹⁴⁾

These data are consistent with the measured promethium isotopic ratios if the ratio of uncollided fission spectrum flux to fission rate (fissions per cm^3) is taken as 11 for Yankee reactor and 3.3 for VBWR. The difference between the two reactors results from the larger amount of moderator in VBWR. In both reactors the amount of Pm^{146} formed by the (n, 2n) process is more than four times the amount formed by direct fission even for the lowest exposure. At high exposures in Yankee reactor more than 40 times as much is formed by the (n, 2n) process.

Shielding Implications

The amount of Pm^{146} in promethium from power reactors has been uncertain, but has been estimated to be as high as 837 ppm, ⁽³⁾ compared to only ~0.25 ppm for Hanford material. The 837 ppm promethium would require approximately the same shielding as Sr^{90} . The data given in Table I show that power reactors do produce a good grade of promethium. The additional shielding requirements over Hanford material are shown in Figure 2 for a 1000 W_t source. The increase amounts to only 0.8 cm for 1 ppm material (VBWR), and 1.6 cm for 4 ppm material (Yankee). Even with this additional requirement, promethium sources compare favorably with Pu^{238} sources.

FIGURE 2

Dose Rates from 1000 W Promethium Sources
with Different Pm¹⁴⁶ Contents

The calculated shielding requirements for promethium and Pu²³⁸ sources are compared in Table II on a shield weight and shield thickness basis. Both materials were considered as spherical oxide sources with a 1 cm

TABLE II

COMPARISON OF SHIELDING REQUIREMENTS FOR PROMETHIUM AND Pu²³⁸ SOURCES

Source Size, W	Dose Rate at 1 m, mR/hr	Shield Thickness, cm ^(a)					Shield Weight, kg ^(a)				
		Promethium			Man ^(e)	Pu ²³⁸ Transistors ^(e)	Promethium			Man ^(e)	Pu ²³⁸ Transistors ^(e)
		(b)	(c)	(d)			(b)	(c)	(d)		
1000	1000	<0.01	<0.01	0.14	0	23	0.02	0.02	1.2	0	72
	100	0.06	0.61	1.4	3.2	38	0.53	5.6	14	1.4	260
	10	1.1	1.9	2.7	23	53	11	21	33	72	640
	1	2.4	3.2	3.9	38	69	29	43	59	260	1330
100	1000	0	0	0	0	0.6	0	0	0	0	0.06
	100	<0.01	0.02	0.42	0	19	0.03	0.05	1.2	0	34
	10	0.17	0.91	1.7	0.6	36	0.46	3.0	6.9	0.06	190
	1	1.4	2.2	3.0	19	50	5.3	10	17	34	480
10	1000	0	0	0	0	0	0	0	0	0	0
	100	0	0	<0.01	0	0	0	0	<0.01	0	0
	10	0.03	0.07	0.60	0	18	0.03	0.07	0.79	0	26
	1	0.34	1.1	1.9	0	35	0.4	1.8	4.1	0	160
1	1000	0	0	0	0	0	0	0	0	0	0
	100	0	0	0	0	0	0	0	0	0	0
	10	<0.01	<0.01	<0.01	0	0	<0.01	<0.01	<0.01	0	0
	1	0.04	0.08	0.70	0	17	0.02	0.04	0.56	0	20

(a) Uranium shield for promethium; lithium hydride shield for plutonium

(b) 0.25 ppm Pm¹⁴⁶(c) 1 ppm Pm¹⁴⁶(d) 4 ppm Pm¹⁴⁶

(e) Radiation from the shielded source will cause the same damage to the receptor as does the listed dose rate.

space between the shield and the source material and with the shield completely surrounding the source.

Uranium shielding was used for the promethium source, except that the first 0.1 cm of the shield was tungsten or tantalum to avoid streaming behind the uranium K X-ray edge. Self-shielding and buildup of scattered radiation in the shield were included in the calculation.

The properties and the neutron shielding requirements of Pu^{238} were obtained from Arnold's report.⁽³⁾ The shield material was lithium hydride. Allowance was not made for the void volume required for helium collection. Neutron multiplication in the source due to fission of Pu^{238} was included, thus increasing the dose rate for the 1000 W source by a factor of 2. (For sources above 5000 W criticality may be reached.)

The shielding comparison is presented both for effects on man and on transistors. The basis for damage to the receptor is that equivalent to a given dose of a pure gamma radiation. Thus, the allowable dose in rep of fast neutrons is one-tenth of the allowable dose in R when man is the receptor being protected and one-thousandth of the allowable dose in R when transistors are being protected.

The comparison, although only strictly valid for the assumptions stated, shows that promethium containing 0.25 ppm Pm^{146} is at least as good as Pu^{238} from a shielding standpoint for most source sizes and allowable dose rates. Promethium from power reactors is superior to Pu^{238} for large sources and low allowable dose rates; i. e., when a rather large amount of shielding is required for either. For small sources or high allowable dose rates, little or no shielding is required by either promethium or plutonium. For some intermediate conditions, plutonium is superior to promethium.

ACTINIDE AND FISSION PRODUCT ISOTOPES

The results of the uranium, neptunium, plutonium, americium, and curium determination and isotopic analyses are given in grams per tonne of uranium. The analyses are all normalized to January 1, 1965. The value used for tonnes of uranium is the amount present at the time of reactor loading. The data are presented in Table III.

TABLE III
ANALYSES^(a) OF ACTINIDE ELEMENTS IN YANKEE AND VBWR FUELS

Sample	Burnup, MWd/ tonne	U ²³⁶	U ²³⁵	U ²³⁴	Total Plutonium	Pu ²³⁸	Pu ²³⁹	Pu ²⁴⁰	Pu ²⁴¹	Pu ²⁴²	Np ²³⁷	Am ²⁴¹	Cm ²⁴²	Cm ²⁴⁴
Yankee Reactor														
Y-1	31,800	3690	11,100	190	10,200	113	6350	2160	1300	390	473	139	1.6	16
Y-3	24,300	3650	16,100	154	10,500	90	7490	1700	1110	200	455	119	1.0	7.9
Y-2	17,700	2230	19,900	234	7,560	32	5620	1240	599	96	205	85	0.033	0.89
Y-5	13,700	2520	21,800	166	7,190	33	5740	927	468	51	242	69	0.024	0.55
Vallecitos Reactor														
V-55	17,400	2890	19,000	165	4600	17.5	3330	880	329	57	140	45	0.39	0.48
V-57	9,900	1830	25,000	207	3280	5.1	2730	420	121	7.6	47	15	0.066	0.072
V-60	6,500	1230	28,500	240	2180	1.42	1961	182	35	1.9	22	5.6	0.018	0.012

(a) All analyses are given in grams per tonne of uranium.

Five fission products (Cs^{137} , Pm^{147} , Sr^{90} , Tc^{99} , and total palladium) were selected for analysis partly because of current interest in their recovery and partly because they represent typical problems in calculating amounts generated in the reactors. The quantities reported are in either curies per tonne of uranium or grams per tonne of uranium (according to conventional usage), and the uranium is the amount charged in the reactor. The data are shown in Table IV.

TABLE IV
ANALYSES OF SOME PERTINENT FISSION PRODUCTS IN YANKEE AND VBWR FUELS

Yankee Reactor						
Sample	Burnup MWd/tonne	Cs ¹³⁷ , Ci/tonne	Sr ⁹⁰ , Ci/tonne	Pm ¹⁴⁷ , Ci/tonne	Tc ⁹⁹ , g/tonne	Total Palladium, g/tonne
Y-1	31,800	1.02×10^5	6.57×10^4	1.25×10^5	770	---
Y-3	24,300	7.97×10^4	5.06×10^4	1.03×10^5	590	720
Y-2	17,700	5.28×10^4	4.06×10^4	7.23×10^4	430	410
Y-5	13,700	4.32×10^4	3.73×10^4	6.06×10^4	340	360
VBWR						
V-55	17,400	5.37×10^4	4.43×10^4	1.01×10^5	430	309
V-57	9,900	3.07×10^4	2.79×10^4	6.10×10^4	266	138
V-60	6,500	1.80×10^4	1.78×10^4	4.09×10^4	144	74

The data presented in Tables III and IV provide a firm basis for computations, such as Rohrmann's,⁽¹⁵⁾ of the economic value of "spent" reactor fuels. The data also supports the thesis that recovery of these by-products (particularly the transuranic isotopes) will have significant impact on the economics of atomic power and on the optimum integration

of reactor operation, fuel reprocessing, and waste management. The predicted long-term growth rate of atomic power production indicates that by 1980 nuclear power generation in the United States will exceed $2 \times 10^5 \text{ MW}_t$. Promethium production, based on the Yankee data at 25,000 MWd/tonne will then be 430 MCi/yr or equivalent to about 215 MCi after aging for one Pm¹⁴⁷ half-life (to allow for Pm¹⁴⁸ decay). As further examples, Np²³⁷, Am²⁴¹, and Cm²⁴⁴ annual productions will be 1330, 350, and 23 kg, respectively.

FUTURE WORK

The present work encompassed only two types of power reactors. Other typical reactors such as the fast breeders and plutonium fueled reactors should be studied. The analyses should also include such additional isotopes as Am²⁴³, Pu²³⁶, rhodium, etc. Reconciliation of experimental results with computer calculations will afford a firm basis for predicting availability of actinides and fission products.

ACKNOWLEDGEMENT

We wish to acknowledge the assistance of R. J. Elovich and T.R. Meyers who performed much of the laboratory work, E. J. Wheelwright who assisted with the promethium purification, and W. A. Emel and his associates who performed many of the separations and analyses. We also express appreciation to R. W. Perkins who made available the counting equipment needed and to R. L. Moore under whose direction this work was performed.

The samples were obtained with the cooperation of Lee Chajson of Westinghouse Electric Corporation and Sherman Naymark of the General Electric Company. George Jordy of the Division of Reactor Development, AEC, suggested the program and kindly made arrangements for obtaining Yankee fuel samples. The work was supported by the Division of Isotopes Development, AEC.

REFERENCES

1. F. P. Roberts, E. J. Wheelwright, and H. H. Van Tuyl. "Detection of Pm^{146} Among the Products of Uranium Fission," J. Inorg. Nucl. Chem., vol. 25, pp. 1298-1300. 1963.
2. H. H. Van Tuyl, F. P. Roberts, and E. J. Wheelwright. Yield of Pm^{146} in Uranium Fission, HW-76515 (General Electric Company, Richland, Washington). February, 1963. (SECRET)
3. E. D. Arnold. Handbook of Shielding Requirements and Radiation Characteristics of Isotopic Power Sources for Terrestrial, Marine, and Space Applications, ORNL-3576 (Oak Ridge National Laboratory). April, 1964.
4. J. D. McGaugh and R. H. Chastain. Power Density and Burnup Distributions in Yankee Core I, WCAP-6051 (Westinghouse Electric Corporation). January, 1963.
5. C. J. Baroch, J. P. Hoffman, and W. C. Rous. AEC Fuel Cycle Program. Design and Fabrication of the Basic Fuel Assemblies, GEAP-3653 (General Electric Atomic Products). March, 1963.
6. F. P. Roberts. Determination of Small Amounts of Cerium Activity in Strontium-90 Using Anion Exchange, HW-69737 (General Electric Company, Richland, Washington). 1961.
7. E. J. Wheelwright and F. P. Roberts. Development and Demonstration of an Ion-Exchange Process for Kilogram-Scale Production of High Purity Promethium, HW-78651 REV. (General Electric Company, Richland Washington). October, 1963.
8. R. W. Perkins, J. M. Nielsen, and R. N. Diebel. "Total Absorption Gamma-Ray Spectrometers Utilizing Anticoincidence Shielding," Rev. Sci. Inst., vol. 31, pp. 1344-1349. December, 1960.
9. A. C. Leaf. Application of the Cesium Tetrphenylborate Extraction to the Measurement of Uranium Burnup, HW-72199 (General Electric Company, Richland, Washington). April, 1962.
10. E. J. Wheelwright, D. M. Fleming, and F. P. Roberts. "The Mean Beta Energy and Half-Life of Pm^{147} ," To be published in April or May issue of J. Phys. Chem. 1965.
11. E. G. Funk, Jr., J. W. Miholich, and C. F. Schwerdtfeger. "The Radioactive Decay of Pm^{143} , Pm^{144} , and Pm^{146} ," Phys. Rev., vol. 120, p. 178. 1960.

12. I. M. H. Pagden, R. Jakeways, and F. C. Flack. "The Decays of Pm^{143} , Pm^{144} , and Pm^{146} ," Nucl. Phys., vol. 48, pp. 555-560. 1963.
13. R. P. Schuman and J. R. Berreth. "Neutron Activation Cross Sections of Pm^{147} , Pm^{148} , and Pm^{148m} ," Nucl. Sci. Eng., vol. 12, pp. 519-522. 1962.
14. Oak Ridge National Laboratory. Status and Progress Report, ORNL-3762. 1964.
15. C. A. Rohrmann. Values in Spent Fuel from Power Reactors, BNWL-25. March, 1965.

UC-4
CHEMISTRY

Ptd.	Standard Distribution	Ptd.	Standard Distribution
3	ABERDEEN PROVING GROUND	2	BABCOCK AND WILCOX COMPANY
1	*ACF INDUSTRIES, INC.	2	BATTELLE MEMORIAL INSTITUTE
2	*AEROJET-GENERAL CORPORATION	1	BERYLLIUM CORPORATION
1	AEROJET-GENERAL NUCLEONICS	4	BROOKHAVEN NATIONAL LABORATORY
2	AERONAUTICAL SYSTEMS DIVISION	1	BUREAU OF MINES, ALBANY
		1	BUREAU OF MINES, SALT LAKE CITY
		1	BUREAU OF SHIPS (CODE 1500)
1	AIR FORCE INSTITUTE OF TECHNOLOGY	1	CHICAGO PATENT GROUP
		1	COLUMBIA UNIVERSITY (CROPPER)
1	AIR FORCE SURGEON GENERAL	1	COMBUSTION ENGINEERING, INC.
2	AIR FORCE WEAPONS LABORATORY	1	COMBUSTION ENGINEERING, INC. (NRD)
1	ALBUQUERQUE OPERATIONS OFFICE	1	COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION
1	ALLIS-CHALMERS MANUFACTURING COMPANY		
1	ALLIS-CHALMERS MANUFACTURING COMPANY, BETHESDA	1	DEFENCE RESEARCH MEMBER
		1	DENVER RESEARCH INSTITUTE
1	ALLISON DIVISION-GMC	1	DOW CHEMICAL COMPANY, ROCKY FLATS
1	ARGONNE CANCER RESEARCH HOSPITAL	3	DU PONT COMPANY, AIKEN
10	ARGONNE NATIONAL LABORATORY	1	DU PONT COMPANY, WILMINGTON
1	ARMED FORCES RADIOBIOLOGY RESEARCH INSTITUTE	1	EDGERTON, GERMESHAUSEN AND GRIER, INC., GOLETA
2	ARMY CHEMICAL RESEARCH AND DEVELOPMENT LABORATORIES	1	EDGERTON, GERMESHAUSEN AND GRIER, INC., LAS VEGAS
1	ARMY ELECTRONICS RESEARCH AND DEVELOPMENT LABORATORIES	3	FRANKFORD ARSENAL
1	ARMY ENGINEER RESEARCH AND DEVELOPMENT LABORATORIES	1	FUNDAMENTAL METHODS ASSOCIATION
		1	GENERAL ATOMIC DIVISION
1	ARMY ENVIRONMENTAL HYGIENE AGENCY	2	GENERAL DYNAMICS/FORT WORTH
1	ARMY MATERIALS RESEARCH AGENCY	2	GENERAL ELECTRIC COMPANY, CINCINNATI
1	ARMY MISSILE COMMAND	4	GENERAL ELECTRIC COMPANY, RICHLAND
2	ARMY NUCLEAR DEFENSE LABORATORY	1	GENERAL ELECTRIC COMPANY, SAN JOSE
1	ARMY RESEARCH OFFICE, DURHAM	1	GOODYEAR ATOMIC CORPORATION
1	ARMY TANK-AUTOMOTIVE CENTER	1	HARVARD UNIVERSITY
1	ATOMIC BOMB CASUALTY COMMISSION	1	HAZLETON NUCLEAR SCIENCE CORPORATION
1	ATOMIC ENERGY COMMISSION, BETHESDA	1	HUGHES AIRCRAFT COMPANY
1	AEC SCIENTIFIC REPRESENTATIVE, FRANCE	2	IOWA STATE UNIVERSITY
1	AEC SCIENTIFIC REPRESENTATIVE, JAPAN	1	JET PROPULSION LABORATORY
3	ATOMIC ENERGY COMMISSION, WASHINGTON	1	JOHNS HOPKINS UNIVERSITY
4	ATOMIC ENERGY OF CANADA LIMITED	2	KNOLLS ATOMIC POWER LABORATORY
2	ATOMIC ENERGY OF CANADA LIMITED, WHITESHELL	2	LOS ALAMOS SCIENTIFIC LABORATORY
4	ATOMICS INTERNATIONAL	1	MALLINCKRODT CHEMICAL WORKS
1	AVCO CORPORATION	1	MARE ISLAND NAVAL SHIPYARD

Ptd.	Standard Distribution	Ptd.	Standard Distribution
1	MARITIME ADMINISTRATION		
1	MARTIN-MARIETTA CORPORATION	1	PUBLIC HEALTH SERVICE, LAS VEGAS
1	MASSACHUSETTS INSTITUTE OF TECHNOLOGY	1	PUBLIC HEALTH SERVICE, MONTGOMERY
1	MONSANTO RESEARCH CORPORATION	1	PURDUE UNIVERSITY
1	MOUND LABORATORY	1	RADIATION APPLICATIONS, INC.
		1	RADIOPTICS, INC.
1	NASA LEWIS RESEARCH CENTER	1	RAND CORPORATION
	NASA LEWIS RESEARCH CENTER, SANDUSKY	1	REYNOLDS ELECTRICAL AND ENGINEERING COMPANY, INC.
1	NASA MANNED SPACECRAFT CENTER	1	ROCKY MOUNTAIN ARSENAL
2	NASA SCIENTIFIC AND TECHNICAL INFORMATION FACILITY	1	SANDIA CORPORATION, ALBUQUERQUE
		1	SANDIA CORPORATION, LIVERMORE
1	NATIONAL AGRICULTURAL LIBRARY	1	SCHOOL OF AEROSPACE MEDICINE
1	NATIONAL BUREAU OF STANDARDS	1	SOUTHWEST RESEARCH INSTITUTE
1	NATIONAL BUREAU OF STANDARDS (LIBRARY)	1	TRACERLAB, INC., RICHMOND
		3	UNION CARBIDE CORPORATION (ORGDP)
1	NATIONAL LEAD COMPANY OF OHIO	4	UNION CARBIDE CORPORATION (ORNL)
1	NAVAL MEDICAL RESEARCH INSTITUTE		
1	NAVAL POSTGRADUATE SCHOOL	1	UNION CARBIDE CORPORATION (PADUCAH PLANT)
1	NAVAL RADIOLOGICAL DEFENSE LABORATORY	1	UNION CARBIDE CORPORATION (TUXEDO)
3	NAVAL RESEARCH LABORATORY	1	UNITED NUCLEAR CORPORATION (NDA)
1	NEW BRUNSWICK AREA OFFICE	1	U. S. GEOLOGICAL SURVEY (BAL)
1	NEW YORK OPERATIONS OFFICE	1	U. S. GEOLOGICAL SURVEY, DENVER
1	NRA, INC.	1	U. S. GEOLOGICAL SURVEY, MENLO PARK
1	NUCLEAR MATERIALS AND EQUIPMENT CORPORATION	1	U. S. GEOLOGICAL SURVEY, WASHINGTON
1	NUCLEAR METALS, INC.	1	U. S. PATENT OFFICE
1	NUCLEAR SCIENCE AND ENGINEERING CORPORATION	2	UNIVERSITY OF CALIFORNIA, BERKELEY
1	NUCLEAR UTILITY SERVICES, INC.	2	UNIVERSITY OF CALIFORNIA, LIVERMORE
1	OFFICE OF ASSISTANT GENERAL COUNSEL FOR PATENTS (AEC)	1	UNIVERSITY OF CALIFORNIA, LOS ANGELES
		1	UNIVERSITY OF HAWAII
6	OFFICE OF NAVAL RESEARCH	1	UNIVERSITY OF PUERTO RICO
1	OFFICE OF NAVAL RESEARCH (CODE 422)	1	UNIVERSITY OF ROCHESTER
1	OFFICE OF THE CHIEF OF RESEARCH AND DEVELOPMENT	1	UNIVERSITY OF UTAH
4	PHILLIPS PETROLEUM COMPANY (NRTS)	1	UNIVERSITY OF WASHINGTON
1	POWER REACTOR DEVELOPMENT COMPANY	1	WALTER REED ARMY MEDICAL CENTER
4	PRATT AND WHITNEY AIRCRAFT DIVISION	2	WESTINGHOUSE BETTIS ATOMIC POWER LABORATORY
1	PRINCETON UNIVERSITY (SHERR)	1	WESTINGHOUSE ELECTRIC CORPORATION
2	PUBLIC HEALTH SERVICE	1	WESTINGHOUSE ELECTRIC CORPORATION (NASA)

INTERNAL DISTRIBUTION

Copy Number

1	F W. Albaugh
2	R. T. Allemann
3	C. A. Bennett
4	E. A. Berreth
5	L. A. Bray
6	L. L. Burger
7	R. E. Burns
8	E W. Christopherson
9	D. E. Deonigi
10	R. F. Dickerson
11	K. M. Harmon
12	E. R. Irish
13	A. R. Keene
14	H. A. Kornberg
15	E. T. Merrill
16	R. L. Moore
17	J M. Nielsen
18	H. M. Parker
19	D. W. Pearce
20	A. M. Platt
21 - 170	F. P. Roberts
171	C. A. Rohrmann
172	L. C. Schwendiman
173	W. H. Swift
174	C. R. Tipton
175	H. H. Van Tuyl
176	E. E. Voiland
177	M. T. Walling
178	E. J. Wheelwright
179	Technical Publications
180 - 184	Technical Information Files

EXTERNAL DISTRIBUTION (SPECIAL)

Number of Copies

2	AEC, Division of Isotope Development Attn: J. L. Bloom W. K. Eister
1	AEC, Division of Reactor Development Attn: G. Y. Jordy
2	APED, San Jose P. O. Box 846 Attn: W. H. Reas Sherman Naymark

EXTERNAL DISTRIBUTION (SPECIAL) (contd.)

Number of Copies

9	General Electric Co. Attn: S. J. Beard M. K. Harmon O. F. Hill O. H. Greager M. C. Leverett M. Lewis H. C. Rathvon R. E. Tomlinson W. K. Woods
1	Martin-Marietta Corporation Attn: R. P. Wischow
5	RLOO Attn: R. K. Sharp (1) Technical Information Library C. L. Robinson N. T. Karagianes J. T. Christy
1	Westinghouse Electric Corporation Attn: L. Chajson

UC-4
CHEMISTRY

Ptd.

325

Standard Distribution

DIVISION OF TECHNICAL INFORMATION
EXTENSION

Ptd.

75

Standard Distribution

CLEARINGHOUSE FOR FEDERAL SCIENTIFIC
AND TECHNICAL INFORMATION

38th Ed.
TID-4500

*New listing or change in old listing.