

664018  
DP-1015

AEC RESEARCH AND DEVELOPMENT REPORT

# **RADIOISOTOPE PRODUCTION CAPABILITIES OF U. S. POWER REACTORS**

E. J. HENNELLY and R. R. HOOD

**RECORD  
COPY**

DO NOT RELEASE  
FROM FILE



*Savannah River Laboratory*  
*Aiken, South Carolina*

## LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Printed in USA. Price \$2.00

Available from the Clearinghouse for Federal Scientific  
and Technical Information, National Bureau of Standards,  
U. S. Department of Commerce, Springfield, Virginia

664018

DP-1015

Reactor Technology  
(TID-4500, 46th Ed.)

# **RADIOISOTOPE PRODUCTION CAPABILITIES OF U. S. POWER REACTORS**

by

Edward J. Hennelly  
and  
Ramon R. Hood

Approved by

D. F. Babcock, Director  
Reactor Engineering Section  
Wilmington, Delaware

November 1965

**E. I. DU PONT DE NEMOURS & COMPANY  
SAVANNAH RIVER LABORATORY  
AIKEN, SOUTH CAROLINA**

**CONTRACT AT(07-2)-1 WITH THE  
UNITED STATES ATOMIC ENERGY COMMISSION**

### ABSTRACT

The future production capability of the U. S. nuclear power complex is estimated for four radioisotopes that are of interest as heat sources, namely,  $^{147}\text{Pm}$ ,  $^{210}\text{Po}$ ,  $^{238}\text{Pu}$ , and  $^{244}\text{Cm}$ . Production estimates for the next 15 years are derived from forecasts of the growth rate of nuclear power and published data on isotopic yields. The estimated total possible production by power reactors in the decade 1971-1980 ranges from 100 kwt (thermal kilowatts) of  $^{244}\text{Cm}$  to 240 kwt of  $^{210}\text{Po}$ ; the estimated possible annual rates in 1980 range from 30 to 50 kwt. Substantial increases in the production of  $^{238}\text{Pu}$  and  $^{244}\text{Cm}$  can be achieved by irradiating  $^{237}\text{Np}$  and  $^{243}\text{Am}$  from power reactors in special reactors that operate at a neutron flux of  $10^{14}$  n/(cm<sup>2</sup>)(sec) or higher. Recycling of plutonium in power reactors will also increase production of  $^{244}\text{Cm}$ .

## CONTENTS

	<u>Page</u>
List of Tables and Figures . . . . .	4
INTRODUCTION . . . . .	5
SUMMARY . . . . .	7
DISCUSSION . . . . .	9
The NASA Requirements . . . . .	9
Growth of Nuclear Power in the U. S. . . . .	9
Data on Radioisotope Yields . . . . .	10
Estimated Production Capacities . . . . .	12
Promethium-147 . . . . .	12
Polonium-210 . . . . .	13
Plutonium-238 . . . . .	13
Curium-244 . . . . .	16
Direct Production of $^{244}\text{Cm}$ . . . . .	16
Production of $^{244}\text{Cm}$ by Plutonium Irradiations . . . . .	16
Production of $^{244}\text{Cm}$ by Irradiating Americium . . . . .	17
Effects of Fuel Recycle . . . . .	18
Chemical Processing . . . . .	19
Economic Factors . . . . .	19
APPENDIX - Analysis of Yankee Production Data . . . . .	21
REFERENCES . . . . .	24

## LIST OF TABLES AND FIGURES

<u>Table</u>	<u>Page</u>
I      Composition of Irradiated Yankee Fuel . .	11
II     Radioisotopes in Power Reactor Fuels . .	17

<u>Figure</u>		
1	Predicted Growth of Nuclear Power in the U. S. . . . .	9
2	Estimated Capacities for Radioisotope Production in U. S. Power Reactors . . .	12
3	Estimated Production of $^{237}\text{Np}$ in U. S. Power Reactors . . . . .	14
4	Estimated Production of $^{238}\text{Pu}$ and $^{244}\text{Cm}$ in High-Flux Irradiations . . . . .	15
5	Effect of Plutonium and Uranium Recycle on $^{238}\text{Pu}$ and $^{244}\text{Cm}$ Production . . . . .	18

## INTRODUCTION

NASA and other agencies project increasing demands for radioisotopes as heat sources.<sup>(1)</sup> The growing nuclear power industry is a potential supplier.

This report is an evaluation of the potential of the nuclear power industry to produce  $^{147}\text{Pm}$ ,  $^{210}\text{Po}$ ,  $^{238}\text{Pu}$ , and  $^{244}\text{Cm}$  by 1980. Such an evaluation is needed to indicate the industry's possible potential for isotope production and to encourage application of the necessary effort to define the potential more accurately. In making this evaluation, the simplifying bases listed below were assumed, even though they introduce uncertainties, many of which may never be resolved completely:

- The nuclear power industry will grow smoothly and continuously.

In the earlier years a smooth growth is obviously impossible because each new reactor adds significantly to the size of the industry. These unpredictable deviations from smooth growth can have a significant effect on the production of radioisotopes that require long lead times, such as  $^{238}\text{Pu}$ .

- The nuclear power industry will grow at an optimistic rate.

An optimistic projection was selected from available forecasts<sup>(2-5)</sup> because of the recent increase in orders for large nuclear power plants. Even if all estimates of the size of the nuclear industry in 1980 were the same, the rate of growth achieved by the industry may differ from any predicted rate, and deviations from the predicted growth will affect the rate of production of radioisotopes. Particularly, a more rapid rate of growth in the earlier years would increase significantly the production of radioisotopes requiring long lead times, such as  $^{238}\text{Pu}$ .

- Yields of  $^{237}\text{Np}$  and  $^{243}\text{Am}$  (the precursors of  $^{238}\text{Pu}$  and  $^{244}\text{Cm}$ , respectively) per unit of power generated will be equal to those in the Yankee lattice.

Yields in this study are based on chemical analyses of Yankee fuel, the only such data available. The yield of these radioisotopes per unit of power generated in the reactors expected by 1980 may be lower than for Yankee, because the yield is closely related to the reactor lattice design and the fuel exposure, and calculations on other reactor designs are only now becoming available. Thus, present production estimates of nonfission product isotopes are

largely a matter of technical judgment, whereas the yields of fission product isotopes can be estimated more accurately from published data.

- Chemical separation technology and large-scale capacity will be available when needed in the 1970's.
- Plutonium will be available for recycle in fuel assemblies, and there will be a technology and an economic incentive for its use.
- Radioisotopes produced in U.S.-designed reactors built in foreign countries are not included.



## SUMMARY

As much as 500 kwt (thermal kilowatts) of  $^{238}\text{Pu}$  may be required for space missions before 1980; as much as 250 kwt per year of combinations of  $^{238}\text{Pu}$ ,  $^{244}\text{Cm}$ ,  $^{210}\text{Po}$ , and  $^{147}\text{Pm}$  may be needed in the 1970's.<sup>(1)</sup>

For comparison, the estimated production capacities of the U. S. power reactor complex are summarized below for three situations:

- I. Only the power reactors with fluxes less than  $4 \times 10^{13} \text{ n}/(\text{cm}^2)(\text{sec})$  are used.
- II. Actinide production is increased by converting  $^{237}\text{Np}$  and  $^{243}\text{Am}$  from power reactors to  $^{238}\text{Pu}$  and  $^{244}\text{Cm}$  in reactors operating at a flux of  $10^{14} \text{ n}/(\text{cm}^2)(\text{sec})$  or more.
- III. Actinide production is further increased by recycling fuel in one-fourth of the power reactor capacity; the neptunium and americium from all power reactors are converted as in II.

The production of the fission product  $^{147}\text{Pm}$  is the same in all cases. However, in Case I,  $^{210}\text{Po}$ ,  $^{238}\text{Pu}$ , and  $^{244}\text{Cm}$  will compete for reactor space; therefore, combinations of these nuclides will be produced in quantities less than the sum of the tabulated numbers.

### Radioisotopes from Power Complex (thermal kilowatts)

	<u>Cumulative Production Capacity for 1971-1980</u>			<u>Annual Rate of Production in 1980</u>		
	<u>I</u>	<u>II</u>	<u>III</u>	<u>I</u>	<u>II</u>	<u>III</u>
$^{238}\text{Pu}$	160	700	800	50	190	220
$^{244}\text{Cm}$	100	600	1700	30	170	480
$^{147}\text{Pm}$	130	-	-	40	-	-
$^{210}\text{Po}$	240	-	-	50	-	-
NASA Objective	500	500	500	250	250	250

Thus, the nuclear industry will be able to contribute significantly to the supply of radioisotopic heat sources. In time, with the help of reactors operating at fluxes of  $10^{14} \text{ n}/(\text{cm}^2)(\text{sec})$  or more, it can meet the demand. Interim needs will presumably be filled by the AEC's production complex.

## DISCUSSION

### THE NASA REQUIREMENTS

Published data on NASA requirements<sup>(1)</sup> indicate a need for 500 kwt of  $^{238}\text{Pu}$  by 1980, and for larger quantities if certain missions materialize. NASA also wants  $^{147}\text{Pm}$ ,  $^{210}\text{Po}$ , and  $^{244}\text{Cm}$ . During the decade 1971 to 1980, annual requirements may rise to 250 kwt for any one of the four radioisotopes.

### GROWTH OF NUCLEAR POWER IN THE U. S.

The estimates in this report are based on a 1964 forecast by the General Electric Company of the growth of central-station nuclear power in the U. S.<sup>(2)</sup> According to this forecast, nuclear capacity will increase at the following annual rates, which are for the first year of operation of the new equipment on the utility systems:

1968	1,400 MWe
1970	2,800 MWe
1975	7,500 MWe
1980	10,000 MWe

The cumulative capacities corresponding to these addition rates are represented by the top curve in Figure 1. The Hanford N-Reactor (800 MWe), not included in the forecast, will add 7% to the 1970 capacity and 2% to the 1975 capacity.

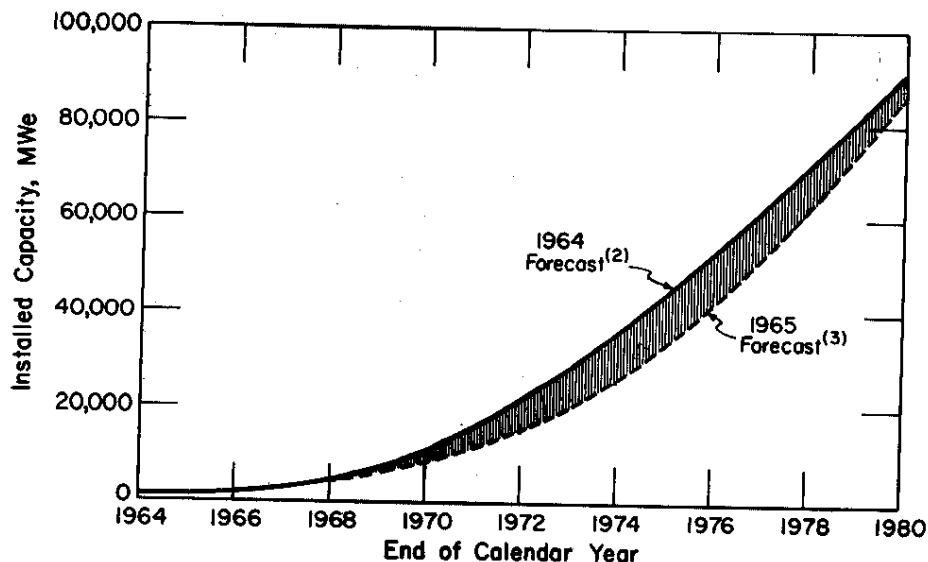


FIG. 1 PREDICTED GROWTH OF NUCLEAR POWER IN THE U. S.

Other recent forecasts are in reasonable agreement with the General Electric prediction, as follows:

Source of Forecast	Installed Capacity in 1980, MWe
General Electric Co. <sup>(2)</sup>	92,000
Westinghouse Electric Co. <sup>(3)</sup>	43,000 to 84,000
U. S. Atomic Energy Commission <sup>(4)</sup>	60,000 to 90,000
Federal Power Commission <sup>(5)</sup>	70,000

A slightly revised forecast was reported by General Electric in September 1965.<sup>(3)</sup> This forecast is also shown in Figure 1, but was not used as a basis for calculations of radioisotope production.

In converting the nuclear capacities to isotope production rates, we assumed that:

- The first products from spent fuel discharged from a new reactor are available four years after reactor startup. This period includes one year for cooling and chemical processing of the fuel.
- Present-day H<sub>2</sub>O-moderated converters with a net thermal efficiency of 32% are used.
- The average fuel exposure is 24,000 MWD/MTU\*.
- The over-all reactor operation is equivalent to the reactor operating 80% of the time at full power.

#### DATA ON RADIOISOTOPE YIELDS

Within the past few months, radioisotopic analyses of irradiated fuel from the Yankee reactor have been reported. In 1962, the Westinghouse Atomic Power Division (WAPD) began a continuing evaluation of this fuel and has reported the results quarterly.<sup>(6)</sup> More recently, some typical WAPD fuel samples were analyzed in detail at Battelle-Northwest.<sup>(7)</sup> Results of these analyses are summarized in Table I

\* Megawatt-days per metric ton of uranium.

and are discussed in the Appendix. Simplified calculations of fuel burnup and isotope production, normalized to these analytical results, form one of the bases for the estimates in this report.

TABLE I

Composition of Irradiated Yankee Fuel  
Based on analyses at Battelle-Northwest<sup>(7)</sup>

Fuel Exposure, MWD/MTU	Content <sup>(a)</sup>							
	Total Pu, g/MWD	Plutonium				<sup>237</sup> Np, mg/MWD	<sup>244</sup> Cm, mg/MWD	<sup>147</sup> Pm, mg/MWD <sup>(b)</sup>
		Each Isotope, isotopic %						
		<sup>239</sup> Pu	<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>242</sup> Pu			
8,500	0.600	85.3	10.0	4.3	0.35	(18)	-	(9.4)
13,700	0.524	80.0	12.8	6.5	0.76	17.7	0.04	(9.8)
24,300	0.433	71.4	16.2	10.5	1.9	18.7	0.325	(7.4)
24,300 with Fuel Recycle	-	[55.0]	[27.5]	[13.0]	[4.5]	[25.3]	[3.4]	[7.4]

(a) Analyzed content except where indicated as follows:

( ) interpolated

[ ] extrapolated

(b) At reactor discharge.

We assumed that the isotope yields in BWR's per unit of electrical capacity are equal to those in PWR's.\* This assumption cannot be tested now inasmuch as no comparable analyses of fuel from BWR's are available. However, the assumption appears reasonable because the following three differences in the operating characteristics of the two reactor types tend to equalize their productivities. Compared to PWR's, the BWR's have:

- A harder neutron spectrum. This favors actinide production.
- A lower (~30%) specific power. This delays delivery of the products.
- A lower <sup>235</sup>U enrichment and a higher plutonium burnup. This improves the yields of the higher actinides at a given fuel exposure.

\* BWR = boiling H<sub>2</sub>O reactor, PWR = pressurized H<sub>2</sub>O reactor.

## ESTIMATED PRODUCTION CAPACITIES

The estimated production capacities of the nuclear power complex for  $^{147}\text{Pm}$ ,  $^{210}\text{Po}$ ,  $^{238}\text{Pu}$ , and  $^{244}\text{Cm}$  are summarized in Figure 2. The estimates for each isotope are discussed below.

### Promethium-147

The production rates for  $^{147}\text{Pm}$  shown in Figure 2 were derived from the measured  $^{147}\text{Pm}$  content of Yankee fuel, which is  $1.03 \times 10^5$  curies/MTU at an exposure of 24,000 MWD/MTU.<sup>(7)</sup> When corrected back to the time of discharge, this rate becomes  $1.63 \times 10^5$  curies/MTU, or 54 w/MTU.

To allow unwanted  $^{148}\text{Pm}$  to decay, the promethium must be stored for about two years after it is discharged from the reactor. Since this delay is about one half-life of  $^{147}\text{Pm}$ , the production is cut in half. This reduced production is plotted in Figure 2.

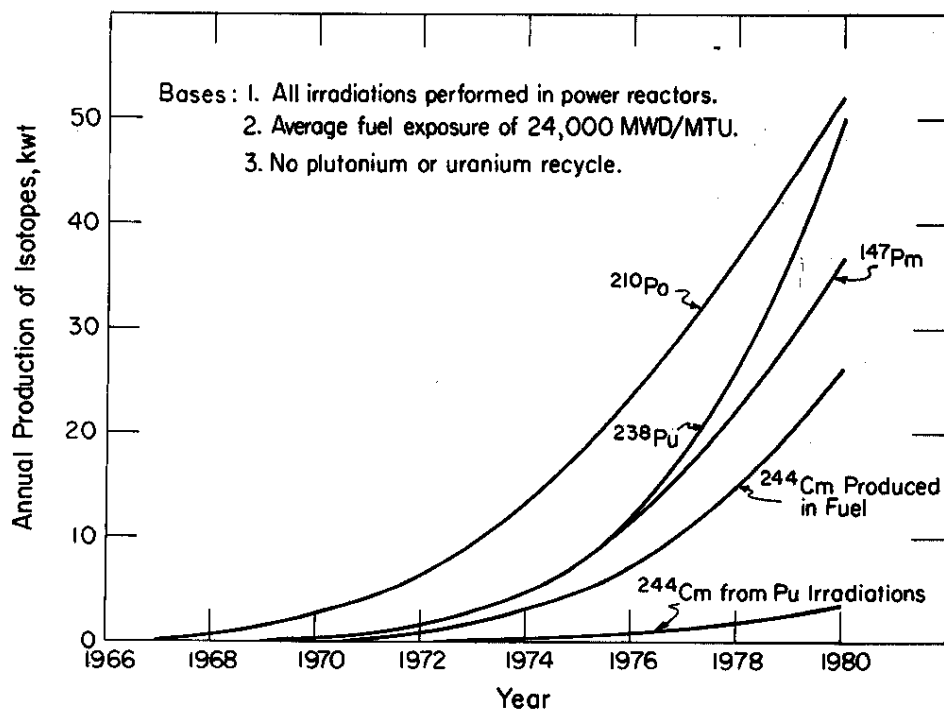


FIG. 2 ESTIMATED CAPACITIES FOR RADIOISOTOPE PRODUCTION IN U. S. POWER REACTORS

A plant to recover  $^{147}\text{Pm}$  and other fission products is to be built at Hanford. It will have a  $^{147}\text{Pm}$  capacity equivalent to 17 kwt/yr.<sup>(a)</sup> By 1980, a capacity of about 40 kwt/yr will be needed.

#### Polonium-210

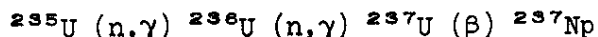
$^{210}\text{Po}$  is produced by neutron capture in  $^{209}\text{Bi}$  targets. The production rate depends on the neutron flux, the irradiation time, and the amount of reactor space that is occupied by bismuth targets. Estimates have been published of the possible  $^{210}\text{Po}$  production from each of 18 power reactors that are now in operation or under construction.<sup>(9)</sup> These estimates are based on assumptions that 2.4% of the fuel volume is available for target assemblies and that the thermal neutron flux in the target is the same as the average in the fuel. The average potential production of the five largest reactors studied (all 300 MWe or more) is  $1.65 \times 10^{-3}$  kwt of  $^{210}\text{Po}$  per year per MWe of capacity provided the net thermal efficiency of the reactors is 32%, the reactor operation is equivalent to operating 80% of the time at full power, and the irradiation time is one year. Production rates derived from these data are shown in Figure 2. The estimates include allowance for decay during a four-month period for cooling and processing. By 1980, the annual  $^{210}\text{Po}$  production can be about 50 kwt, which is one-fifth the NASA requirement.

The half-life of  $^{210}\text{Po}$  is short in comparison with the interval between refuelings of power reactors (4.6 months vs. 12 to 24 months). If the bismuth targets could be removed after about six months of irradiation (when the  $^{210}\text{Po}$  concentration is somewhat more than half the saturation value),  $^{210}\text{Po}$  production would be about one-third greater than shown in Figure 2.

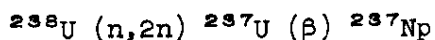
Power reactors will probably be a high-cost source of  $^{210}\text{Po}$ . In addition to the disparities between desirable irradiation intervals and refueling schedules, the  $^{210}\text{Po}$  is produced at low concentration, about 5 g per ton of bismuth. Costs incurred outside of the reactor for handling bismuth at this low concentration probably will be high.

#### Plutonium-238

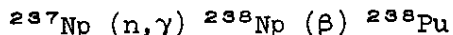
$^{238}\text{Pu}$  is produced by multiple neutron captures. The pertinent reactions in reactors that are fueled with uranium of low enrichment are the formation of  $^{237}\text{Np}$  by the reactions



and



followed by the reaction



To avoid dilution of the  $^{238}\text{Pu}$  with unwanted isotopes, the  $^{237}\text{Np}$  is separated from irradiated fuel and is then irradiated further. The production of  $^{238}\text{Pu}$  is therefore a two-step process.

A  $^{237}\text{Np}$  concentration of 455 g/MTU in Yankee fuel irradiated to 24,000 MWD/MTU, or 18.7 mg/MWD, has been measured.<sup>(7)</sup> The corresponding production rates are shown in Figure 3. The Yankee data indicate that fuel exposure has minor effect on the  $^{237}\text{Np}$  production rate; the concentration in fuel irradiated to 13,700 MWD/MTU is little more than half that at 24,000 MWD/MTU, but the higher fuel throughput for the lower exposure compensates to the extent that the annual production rates of  $^{237}\text{Np}$  at the two exposures differ by only 5%.

After the  $^{237}\text{Np}$  is recovered, it can be irradiated to make  $^{238}\text{Pu}$  either in power reactors or in special converter reactors that operate at a higher neutron flux. Figure 2 shows the estimated  $^{238}\text{Pu}$  production capability for the irradiation of  $^{237}\text{Np}$  in power reactors. For this estimate, the fractional conversion of  $^{237}\text{Np}$  for one year of irradiation was obtained from Reference 9 as the average for the five largest present-day reactors, 0.086 g of  $^{238}\text{Pu}$  per g of  $^{237}\text{Np}$  irradiated. The

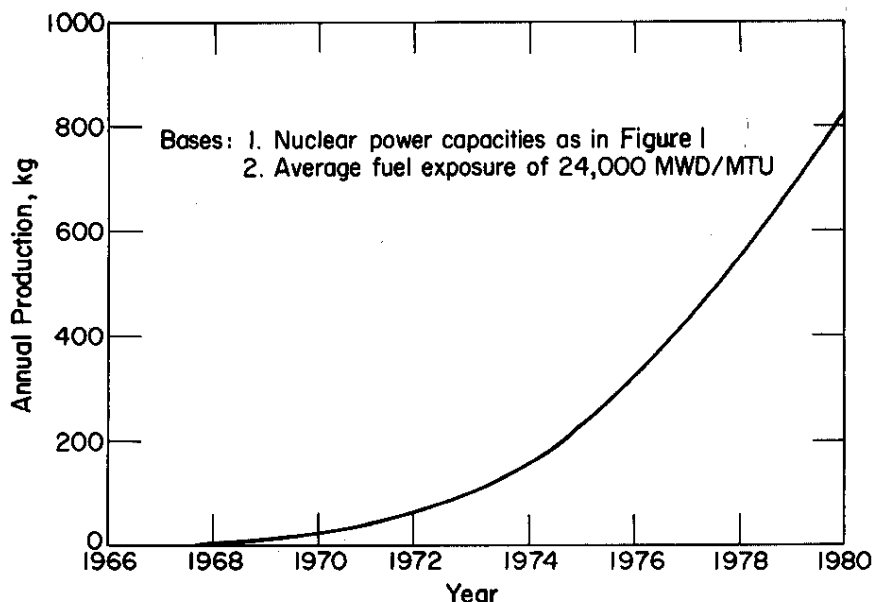


FIG. 3 ESTIMATED PRODUCTION OF  $^{237}\text{Np}$  IN U. S. POWER REACTORS

corresponding  $^{237}\text{Np}$  burnup (10.5%) was obtained from Reference 10, which shows the fractional conversion as a function of  $^{237}\text{Np}$  burnup. Recycle of unconverted  $^{237}\text{Np}$  was assumed, with a six-month delay to separate  $^{238}\text{Pu}$  and fabricate new  $^{237}\text{Np}$  targets. On this basis, approximately 160 kwt of  $^{238}\text{Pu}$  could be produced in power reactors by 1980.

The production of  $^{238}\text{Pu}$  can be accelerated by irradiating  $^{237}\text{Np}$  at a higher flux than is typical of power reactors. The higher the flux, the shorter the irradiation time required to convert a given fraction of the neptunium. A flux of about  $10^{14} \text{ n}/(\text{cm}^2)(\text{sec})$  would suffice for near-maximum production of  $^{238}\text{Pu}$ . Shorter irradiations at even higher fluxes do not help because (1) the time required for out-of-reactor processing then limits the production rate, and (2)  $^{238}\text{Np}$  fissions decrease the product yield.

Estimates of  $^{238}\text{Pu}$  production from  $^{237}\text{Np}$  irradiations at a flux of  $2 \times 10^{14} \text{ n}/(\text{cm}^2)(\text{sec})$  are shown in Figure 4. These estimates are based on the  $^{237}\text{Np}$  supply rates in Figure 3 and the conversion data in References 9 and 10. Three months' irradiation at  $2 \times 10^{14} \text{ n}/(\text{cm}^2)(\text{sec})$  converts 17% of the  $^{237}\text{Np}$  to  $^{238}\text{Pu}$ .<sup>(9)</sup> The corresponding  $^{237}\text{Np}$  burnup is 24%.<sup>(10)</sup> We assumed that 90 days are required to cool the neptunium targets, separate the unconverted neptunium, refabricate it into new targets, and put it back in the reactors. The production rates under these conditions are severalfold greater than those of power reactors.

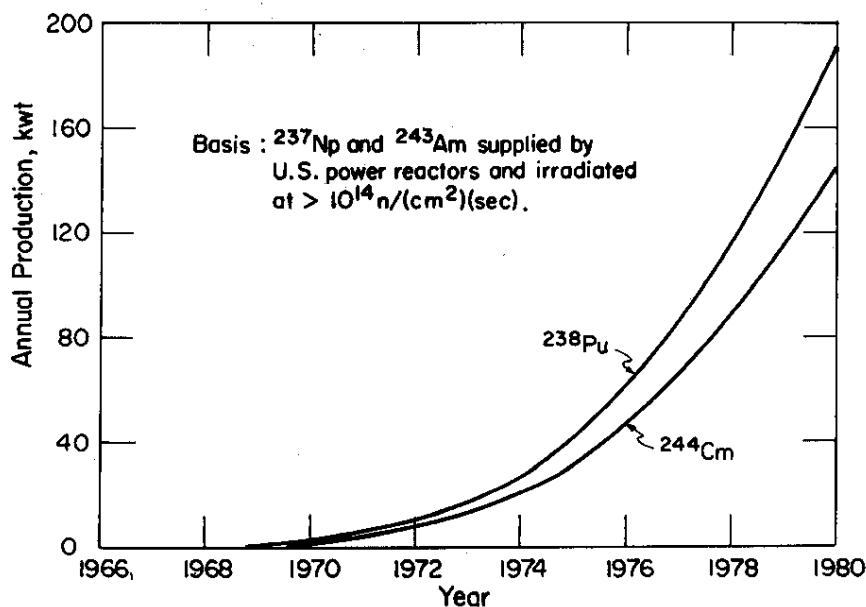


FIG. 4 ESTIMATED PRODUCTION OF  $^{238}\text{Pu}$  AND  $^{244}\text{Cm}$  IN HIGH-FLUX IRRADIATIONS



### Curium-244

Small amounts of  $^{244}\text{Cm}$  are present in irradiated fuel from power reactors. Additional quantities can be produced by irradiating either plutonium or  $^{243}\text{Am}$  from spent fuel.

Direct Production of  $^{244}\text{Cm}$ . The direct production of  $^{244}\text{Cm}$  in power reactor fuel, with no recycling of plutonium or americium, was estimated from the isotopic analyses of Yankee fuel.<sup>(7)</sup> At 24,000 MWD/MTU, this fuel contains 7.9 g of  $^{244}\text{Cm}$ /MTU. In translating this yield to annual production rates, we assumed that the  $^{244}\text{Cm}$  must be held for two years after discharge to permit  $^{242}\text{Cm}$  to decay. The aging period delays delivery and allows 5% of the product to decay. The estimated production rates are plotted in Figure 2; the total production through 1980 is estimated to be 90 kwt.

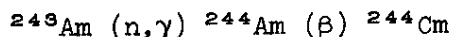
The  $^{244}\text{Cm}$  production depends strongly on fuel exposure. The concentration in Yankee fuel at 13,700 MWD/MTU is only one-tenth of that at 24,000 MWD/MTU.<sup>(7)</sup> Although the lower concentration is partially offset by higher fuel throughputs, the annual production rates for the lower exposure are only 20% of those for the higher exposure. An average exposure of 24,000 MWD/MTU in power reactors in the 1970's appears reasonable, as reactors are being designed now for exposures of 21,000 to 22,000 MWD/MTU (e.g., Connecticut Yankee, Dresden II).

Production of  $^{244}\text{Cm}$  by Plutonium Irradiations. The rate at which  $^{244}\text{Cm}$  can be produced by irradiating plutonium in power reactors depends on the quantity of plutonium that is irradiated, its isotopic composition, and the irradiation time. As shown below, the  $^{244}\text{Cm}$  yield from plutonium targets at power reactor fluxes [ $4 \times 10^{13}$  n/(cm<sup>2</sup>)(sec)] is only a small fraction of that produced directly in power reactor fuel. Reference 9 lists the following estimates for four years of irradiation:

Reactor	Wt of Pu Charged, kg	Power Level, MWt	$^{244}\text{Cm}$ Yield, g/kg Pu
Connecticut Yankee	90	1470	0.24
Malibu	75	1470	0.41
Nine-Mile Point	70	1540	0.76
Oyster Creek	124	1600	0.70
So. Calif. Edison	77	1210	0.24

The average yield for these five reactors is 0.47 g/kg of Pu irradiated; this number applies when  $^{239}\text{Pu}$  is the target and when 2.4% of the fuel volume is replaced by targets. If reactor-grade plutonium (~20%  $^{240}\text{Pu}$ ) is the target, the  $^{244}\text{Cm}$  yield can be increased 80%. On the basis of these data, less than 20 kwt of  $^{244}\text{Cm}$  can be obtained by 1980 by irradiating plutonium targets in power reactors. This is only 20% of the amount produced directly in power reactor fuel.

Production of  $^{244}\text{Cm}$  by Irradiating Americium.  $^{244}\text{Cm}$  production could be increased by irradiating plutonium at a neutron flux of about  $10^{14} \text{ n}/(\text{cm}^2)(\text{sec})$ . However, a more economical way is to separate  $^{243}\text{Am}$  from power reactor fuel, and to irradiate this isotope to form  $^{244}\text{Cm}$  by the reaction



No experimental data are available on the  $^{243}\text{Am}$  content of power reactor fuel. However, because of the excellent agreement (see Appendix) between measured and calculated concentrations of  $^{244}\text{Cm}$  in Yankee fuel, calculated yields of  $^{243}\text{Am}$  are believed to be sufficiently accurate for estimating purposes. As shown in Table II, the estimated  $^{243}\text{Am}$  concentration in fuel irradiated to 24,000 MWD/MTU is 40 g/MTU. We assumed that 90% of the  $^{243}\text{Am}$  can be converted to  $^{244}\text{Cm}$  each year (including allowance for chemical processing). This conversion rate requires a neutron flux greater than  $1 \times 10^{14} \text{ n}/(\text{cm}^2)(\text{sec})$ . The corresponding  $^{244}\text{Cm}$  production rates are plotted in Figure 4. Comparison with Figure 2 shows that the  $^{244}\text{Cm}$  production from  $^{243}\text{Am}$  is about six times the direct production in power reactor fuel.

TABLE II

Radioisotopes in Power Reactor Fuels

Data derived from experimental data on Yankee Fuel

Fuel Exposure, MWD/MTU	Concentration at Discharge <sup>(a)</sup> , g/MTU				
	Pu	$^{239}\text{Np}$	$^{244}\text{Cm}$	$^{147}\text{Pm}$	$^{243}\text{Am}$
8,500	5,100	(153)	-	(80)	-
13,700	7,190	242	0.55	(133)	(6)
24,300	10,500	455	7.9	(180)	(40)
24,300 with Fuel Recycle	[15,000]	[635]	[82.0]	(180)	[230]

(a) Measured concentrations except where indicated as follows:

( ) interpolated  
[ ] extrapolated

## EFFECTS OF FUEL RECYCLE

It is likely that plutonium will eventually be recycled in power reactors, especially if uranium prices increase. If such recycling becomes sizable in the 1970's, and if uranium is also recycled,  $^{238}\text{Pu}$  and  $^{244}\text{Cm}$  production can be increased.

Actinide concentrations in spent fuel from plutonium recycle were calculated and are given in Table II. These estimates showed that when fuel initially containing 1 wt % Pu is irradiated to 24,000 MWD/MTU, the  $^{237}\text{Np}$ ,  $^{243}\text{Am}$ , and  $^{244}\text{Cm}$  concentrations will be 1.4, 5.8, and 10 times as great as those resulting from irradiation of virgin uranium to this same exposure. No more than about 25% of the power reactor capacity could employ recycle in the early 1970's even if all of the plutonium and uranium then available were used for that purpose. The total production of  $^{238}\text{Pu}$  and  $^{244}\text{Cm}$  would increase by 10% and 130%, respectively, as shown in Figure 5, if:

- Plutonium and uranium fuels are recycled in 25% of the capacity, and
- Neptunium and americium targets are irradiated.

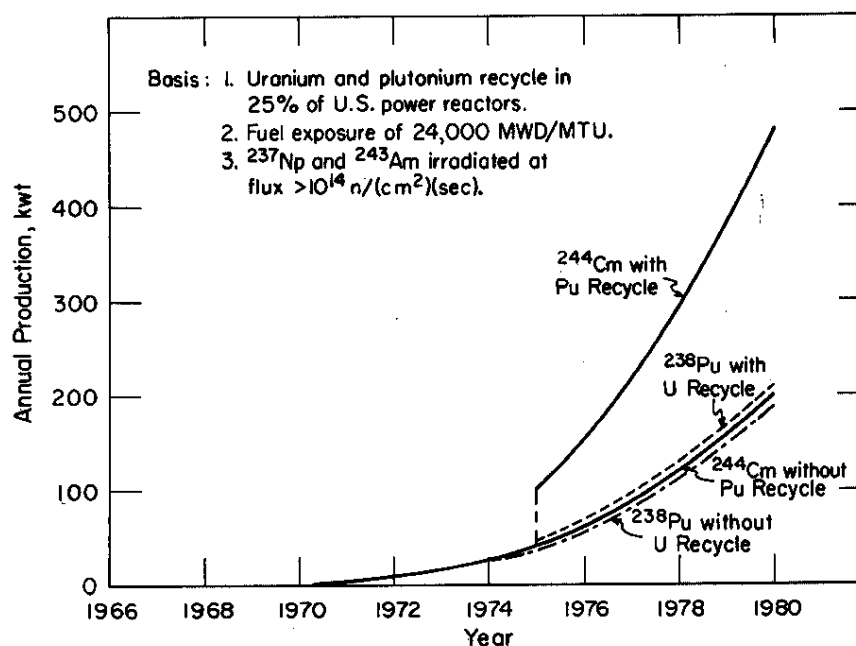


FIG. 5 EFFECT OF PLUTONIUM AND URANIUM RECYCLE  
ON  $^{238}\text{Pu}$  AND  $^{244}\text{Cm}$  PRODUCTION

## CHEMICAL PROCESSING

If spent power reactor fuels are to become a source of  $^{238}\text{Pu}$  and  $^{244}\text{Cm}$ , suitable facilities must be provided for the separation and purification of these isotopes and their precursors,  $^{237}\text{Np}$  and  $^{243}\text{Am}$ . These facilities are in addition to those now projected for recovery of the primary components,  $^{239}\text{Pu}$  and uranium. By 1980, such facilities must annually recover hundreds of kilograms of  $^{237}\text{Np}$ ,  $^{238}\text{Pu}$ ,  $^{243}\text{Am}$ , and  $^{244}\text{Cm}$  from power reactor fuels and targets. Also, large quantities of materials must be refabricated into fuel or target components and recycled through the reactors. Some of these materials will have to be fabricated by remote methods.

$^{237}\text{Np}$  is now recovered from spent fuel at Savannah River and Hanford. The commercial plant being built by Nuclear Fuel Services is not equipped to recover  $^{237}\text{Np}$ , nor is it equipped to process targets after irradiation. Thus, a substantial investment in processing plants is required for the power reactor industry to produce  $^{238}\text{Pu}$ .

A similar situation exists with regard to  $^{243}\text{Am}$  and  $^{244}\text{Cm}$ . No facilities exist or are projected to recover these isotopes except in experimental amounts. However, even though the most probable chemical processes use very corrosive solutions, technology will be available by the end of this decade for designing separations facilities.

## ECONOMIC FACTORS

A detailed discussion of probable production costs for isotopes is beyond the scope of this report; however, a few general statements can be made. First, an important factor that will determine large-scale use will be the policies that must be developed on the pricing of the various reactor products. Second, large-scale processing in the future should reduce costs significantly, and in certain cases, the major expense will be the cost of chemical processing. For example, directly produced  $^{147}\text{Pm}$ ,  $^{243}\text{Am}$ , and  $^{244}\text{Cm}$  can be separated together with the rare earths and trivalent actinides in the aqueous chemical process. The costs per gram for separating these products would therefore be similar since concentrations are comparable. Equally low costs for recovering  $^{237}\text{Np}$  might be expected for similar reasons.  $^{237}\text{Np}$  and  $^{243}\text{Am}$  are thus potentially low-cost feed materials for further irradiation at higher fluxes to produce large quantities of  $^{238}\text{Pu}$  and  $^{244}\text{Cm}$ .

The availability of large quantities of  $^{244}\text{Cm}$  and  $^{238}\text{Pu}$  at low unit costs in the late 1970's and beyond could have a significant and beneficial effect on the potential market for these products. This

availability, however, will be possible only if the chemical processes that are selected can be adapted to yield the desired products economically.

## APPENDIX

### ANALYSIS OF YANKEE PRODUCTION DATA

The principal experimental data<sup>(6,7)</sup> used in deriving the results in this report are summarized in Figures A-1 and A-2.

In Figure A-1, the symbols Y-1, Y-2, Y-3, and Y-5 identify samples that were analyzed in detail at Battelle.<sup>(7)</sup> Batch 1 and Batch 2 refer to the Yankee Core I and Core II material that is being shipped to Nuclear Fuel Services. The upper curve was drawn through a large number of data points obtained by WAPD<sup>(6)</sup> for the asymptotic or average neutron spectrum in the core. The lower curve is typical of the perturbed regions of the core, e.g., near the control rods, and was also drawn through many data points.<sup>(6)</sup> The lattice diagram shows the central core location of the samples measured by Battelle.

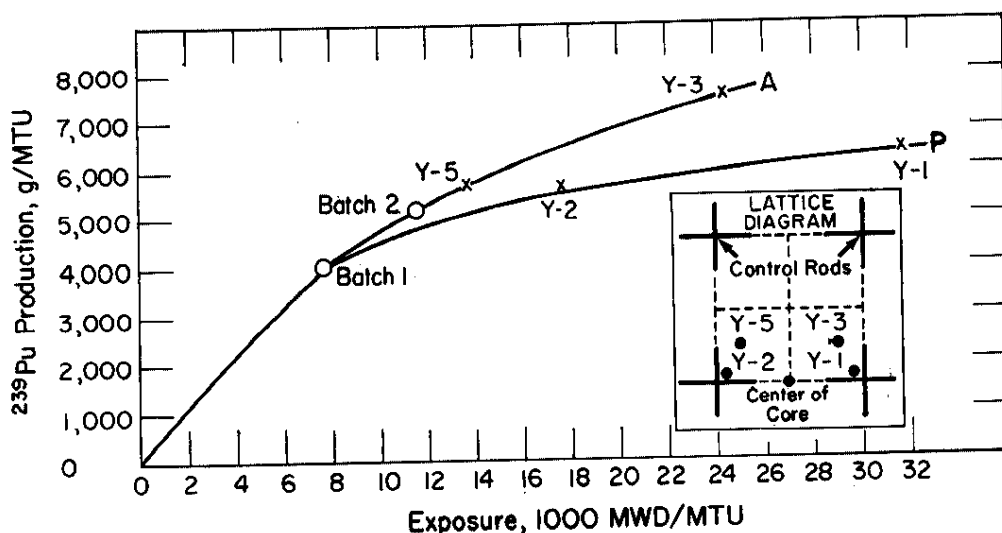


FIG. A-1  $^{239}\text{Pu}$  PRODUCTION IN YANKEE POWER REACTOR

Samples Y-3 and Y-5 were from the asymptotic region and fall nicely on that curve (labeled A). The other two samples were from a perturbed region adjacent to the cruciform control rods, and the data for these samples also fall nicely on the perturbed spectrum curve (labeled P). To establish the average performance of the core, only the samples Y-3 and Y-5 have relevance with regard to the production of higher isotopes such as  $^{243}\text{Am}$  and  $^{244}\text{Cm}$  in a pressurized water core. Both the asymptotic

and the perturbed spectra were calculated; the calculated spectra confirm the application of the Savannah River Burnup Code to predicting power reactor actinide production. The calculated and experimental data are summarized in Figure A-2.

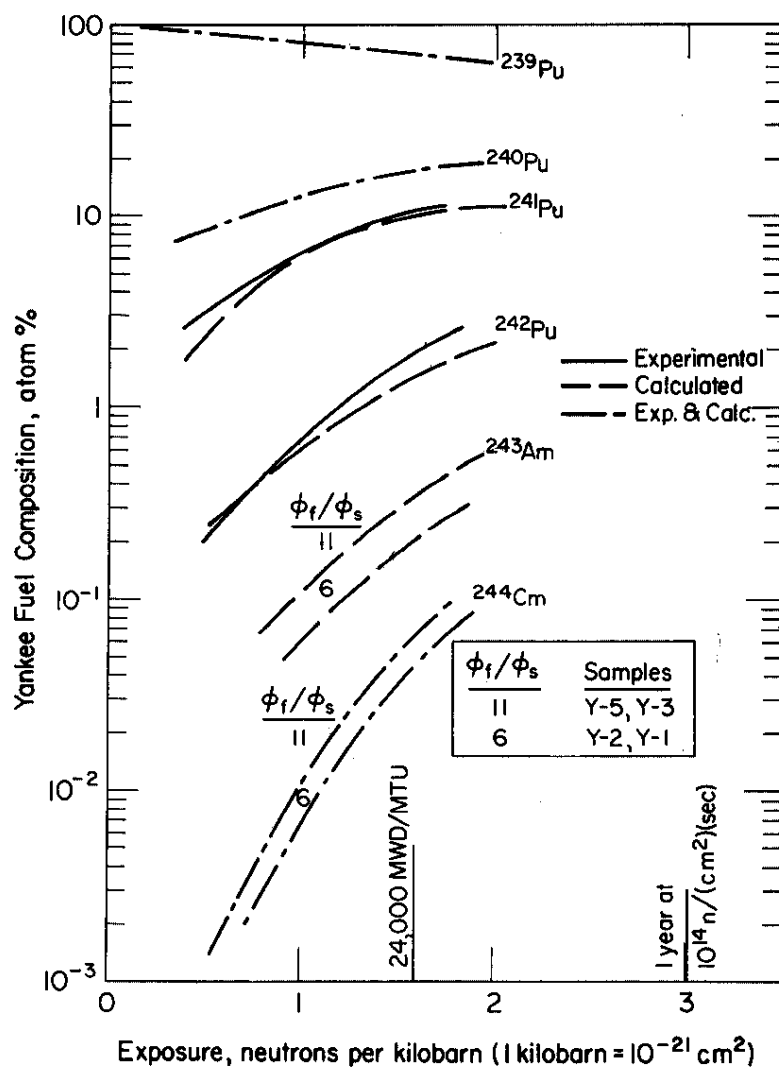


FIG. A-2 COMPARISON OF CALCULATED AND EXPERIMENTAL DATA ON YANKEE FUEL COMPOSITION

The curves for the calculated and experimental data overlap for  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ . The experimental data curves for all the plutonium isotopes were drawn through about 50 experimental points determined by WAPD. These data were used by WAPD to modify their own calculational techniques. The Battelle data agree exactly with the WAPD data. The curves for  $^{244}\text{Cm}$  for both spectra also overlap the experimental data given by Battelle. The quantity  $\phi_f/\phi_s$  is the ratio of fast-to-slow flux used to duplicate the spectrum. The ratios were obtained from WAPD reports<sup>(6)</sup> for the Yankee Reactor. The hardness of the neutron spectrum accounts for the unexpectedly large amount of  $^{244}\text{Cm}$  production in Yankee fuel; the thermal or slow neutron flux is only  $\sim 1.5 \times 10^{13}$  n/(cm<sup>2</sup>)(sec). Figure A-2 also shows the exposures in neutrons per kilobarn that are equivalent to 24,000 MWD/MTU and to 1 year at  $10^{14}$  n/(cm<sup>2</sup>)(sec); these are two other useful measures of fuel exposure. These exposures apply only to the cases calculated, and their mutual equivalence is not unique, but changes as a function of fuel enrichment, plutonium content, and neutron spectra. Because the burnup code was used so successfully to calculate  $^{244}\text{Cm}$  for two spectra, the calculated, interpolated yields of  $^{243}\text{Am}$  are believed to be equally accurate, even though no comparable experimental data are available. The burnup code was also used to extrapolate actinide production for hypothetical fuel recycle conditions in the late 1970's. No decrease in relative accuracy should result from these extrapolations.



## REFERENCES

1. H. Taylor. "NASA Sees Rising Need for Isotopes." Missiles and Rockets 16, No. 17, 12 (1965).
2. "The Jersey Central Story: Nuclear Power Under 4 Mills." Nucleonics Week 5, No. 8, 1-5 (1964).
3. "Power Reactor Market Advances; New Forecasts Favorable." Nucleonics 23, No. 9, 17-18 (1965).
4. G. F. Tape, F. K. Pittman, and M. F. Searl. "Future Energy Needs and the Role of Nuclear Power." Proceedings of Third International Conference on Peaceful Uses of Atomic Energy, Geneva, 1, 69-76 (1964).
5. Federal Power Commission. National Power Survey. Washington: U. S. Government Printing Office (1964).
6. Yankee Core Evaluation Program. Quarterly Progress Report for the Period Ending December 31, 1964. USAEC Report WCAP-6070, Westinghouse Atomic Power Division, Pittsburgh, Pa. (1965).
7. F. P. Roberts and H. H. Van Tuyl. Promethium-146, Fission Product, and Transuranium Isotope Content of Power Reactor Fuels, Comprehensive Chemical Analyses. USAEC Report BNWL-45, Battelle-Northwest, Richland, Wash. (1965).
8. "The Impressive Scope of the Fission Products Conversion and Encapsulation Plant." Nucleonics Week 6, No. 30, 5 (1965).
9. A. F. Rupp, J. A. Cox, and F. T. Binford. Radioisotope Production in Power Reactors. USAEC Report ORNL-3792, Oak Ridge National Laboratory, Oak Ridge, Tenn. (1965).
10. D. R. Vondy, J. A. Lane, and A. T. Gresky. "Production of  $\text{Np}^{237}$  and  $\text{Pu}^{238}$  in Thermal Power Reactors." Ind. Eng. Chem. Process Design and Development 3, 293-6 (1964).