

23 RECORDS ADMINISTRATION  
AGNP

SECTION MEASUREMENT OF  
ION IN  $^{238}\text{Pu}$  BY  $^{237}\text{Np}(n,2n)$  REACTIONS

by

C. K. Paulson\*

and

E. J. Hennelly\*\*

Savannah River Laboratory  
E. I. du Pont de Nemours and Company  
Aiken, South Carolina 29801

SRL

A Paper Proposed for Publication in  
*Nuclear Science and Engineering*

This paper was prepared in connection with work under Contract No. AT(07-2)-1 with the U. S. Atomic Energy Commission. By acceptance of this paper, the publisher and/or recipient acknowledges the U. S. Government's right to retain a non-exclusive, royalty-free license in and to any copyright covering this paper, along with the right to reproduce and to authorize others to reproduce all or part of the copyrighted paper.

\* Now with Westinghouse Electric Company, Monroeville Nuclear Center, Monroeville, Pennsylvania.

\*\* Author to whom correspondence should be sent.

This document was prepared in conjunction with work accomplished under Contract No. AT(07-2)-1 with the U.S. Department of Energy.

### **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.

This report has been reproduced directly from the best available copy.

Available for sale to the public, in paper, from: U.S. Department of Commerce, National Technical Information Service, 5285 Port Royal Road, Springfield, VA 22161, phone: (800) 553-6847, fax: (703) 605-6900, email: [orders@ntis.fedworld.gov](mailto:orders@ntis.fedworld.gov) online ordering: <http://www.ntis.gov/ordering.htm>

Available electronically at <http://www.doe.gov/bridge>

Available for a processing fee to U.S. Department of Energy and its contractors, in paper, from: U.S. Department of Energy, Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831-0062, phone: (865) 576-8401, fax: (865) 576-5728, email: [reports@adonis.osti.gov](mailto:reports@adonis.osti.gov)

CROSS SECTION MEASUREMENT OF  
 $^{236}\text{Pu}$  FORMATION IN  $^{238}\text{Pu}$  BY  $^{237}\text{Np}(n,2n)$  REACTIONS

by

C. K. Paulson\*

and

E. J. Hennelly

Savannah River Laboratory  
E. I. du Pont de Nemours and Company  
Aiken, South Carolina 29801

ABSTRACT

High-energy cross sections for  $^{237}\text{Np}$  are needed to calculate unwanted  $^{236}\text{Pu}$  contaminant in  $^{238}\text{Pu}$  product made for heat source applications in space and medical projects. Recent determination of the  $^{237}\text{Np}(n,2n)$  cross section, averaged over a  $^{235}\text{U}$  fission spectrum, completes the information needed to calculate  $^{236}\text{Pu}/^{238}\text{Pu}$  ratios over a wide range of  $^{237}\text{Np}$  irradiation environments, from  $\text{D}_2\text{O}$  reflectors to PWR fuel. The value is 63 mb averaged over the spectrum  $>6.8$  MeV.

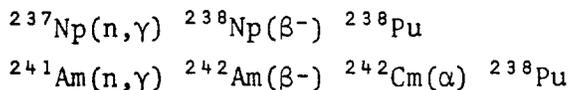
---

The information contained in this article was developed during the course of work under Contract AT (07-2)-1 with the U.S. Atomic Energy Commission.

\*Now with Westinghouse Electric Company, Monroeville Nuclear Center, Monroeville, Pennsylvania.

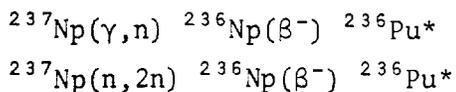
## INTRODUCTION

$^{238}\text{Pu}$  is produced by two methods: the irradiation of  $^{237}\text{Np}$  and the irradiation of  $^{241}\text{Am}$ . The nuclear reactions that produce  $^{238}\text{Pu}$  from these isotopes are:



$^{237}\text{Np}$  is a byproduct of uranium fuel irradiations.  $^{241}\text{Am}$  is a byproduct of plutonium irradiations.

The two primary measures of the quality of  $^{238}\text{Pu}$  produced are  $^{238}\text{Pu}$  isotopic assay and  $^{236}\text{Pu}/^{238}\text{Pu}$  ratio. High assay  $^{238}\text{Pu}$  is desirable because in most applications (heat sources for space and medical projects) weight minimization is important. High assay can be made by selection of proper irradiation conditions of either target nuclide.<sup>1,2</sup> A low  $^{236}\text{Pu}/^{238}\text{Pu}$  ratio is also desirable because  $^{236}\text{Pu}$  daughters produce high-energy gamma radiation. A low  $^{236}\text{Pu}/^{238}\text{Pu}$  ratio is especially important when  $^{238}\text{Pu}$  is used for medical applications because the radiation dose received by the patient should be as low as possible.  $^{236}\text{Pu}$  in the  $^{238}\text{Pu}$  produced by irradiating  $^{237}\text{Np}$  is formed by two reactions:



The gamma rays and neutrons initiating these reactions must have energies  $>6.8$  MeV.  $^{238}\text{Pu}$  formed by irradiating  $^{241}\text{Am}$  is essentially free from  $^{236}\text{Pu}$  contamination ( $<0.005$  ppb  $^{236}\text{Pu}$  in test samples irradiated at Savannah River).<sup>3</sup>

---

\*50% of the  $^{236}\text{Np}$  formed by  $(\gamma,n)$  and  $(n,2n)$  reaction decays to  $^{236}\text{Pu}$ , and 50% decays to  $^{236}\text{U}$ .

$^{237}\text{Np}(\gamma, n)$  cross sections as a function of energy were determined, and the results were reported earlier,<sup>4,5</sup> along with a preliminary  $^{237}\text{Np}(n, 2n)$  cross section measurement. Results of a remeasurement of the  $^{237}\text{Np}(n, 2n)$  cross section in a  $^{235}\text{U}$  fission spectrum is reported herein; the value is 63 mb averaged over the spectrum  $>6.8$  MeV.

### CROSS SECTION MEASUREMENT

Measurement of  $^{236}\text{Pu}$  formation in highly enriched  $^{235}\text{U}$ -aluminum fuel assemblies provides excellent data for determining the  $^{237}\text{Np}(n, 2n)$  cross section for the following reasons:

- $^{236}\text{Pu}$  formation relative to  $^{238}\text{Pu}$  production is high. Therefore, an accurate measured value of the  $^{236}\text{Pu}/^{238}\text{Pu}$  ratio can be obtained.
- $^{236}\text{Pu}$  formation from  $(\gamma, n)$  events represents less than 15% of the total.
- $^{236}\text{Pu}$  formation by high-energy fission neutrons from isotopes other than  $^{235}\text{U}$  is  $<1\%$ . Therefore, only the  $^{235}\text{U}$  fission spectrum must be considered.

Three samples of highly enriched fuel irradiated to about 30%  $^{235}\text{U}$  burnup were taken from a region of a uniform-lattice,  $\text{D}_2\text{O}$ -reactor charge where the flux gradient was approximately zero, and where other operating parameters (assembly power, flux distribution, fuel isotopic composition) were well known.  $^{236}\text{Pu}/^{238}\text{Pu}$  ratios were determined from alpha activity ratios measured by direct alpha pulse height measurements of purified samples<sup>3</sup> of plutonium that had been chemically separated from the dissolved fuel. Results of these measurements, corrected to reactor shutdown, are given in Table I.

TABLE I

 $^{236}\text{Pu}/^{238}\text{Pu}$  Ratios for Three Fuel Samples

Sample	$^{236}\text{Pu}/^{238}\text{Pu}$ Ratio (ppm)
1	4.84 $\pm$ 0.4
2	4.45 $\pm$ 0.4
3	4.55 $\pm$ 0.4
Average	4.61 $\pm$ 0.25

The  $^{237}\text{Np}(n,2n)$  cross section was calculated from these results by using methods described earlier.<sup>4</sup> High-energy gamma ray and neutron fluxes in the fuel (>6.8 MeV) were calculated using integral transport methods. Time-dependent isotopic concentrations were calculated at every 1% burnup increment of the  $^{235}\text{U}$  fuel. Calculated fuel isotopic concentrations agreed to within 5% of measured values. Particle fluxes and concentrations were input to CASPER, a special burnup code,<sup>4</sup> and the (n,2n) cross section was adjusted until the calculated value of the  $^{236}\text{Pu}/^{238}\text{Pu}$  ratio agreed with experiment.<sup>4</sup> The  $^{237}\text{Np}(n,2n)$  cross section calculated by this analysis can be represented as

$$\sigma_{37}^{n,2n} = \frac{\int_{6.8}^{25} \sigma_{37}^{n,2n}(E) \chi(E) dE}{\int_{6.8}^{25} \chi(E) dE} \quad (1)$$

where

$\sigma_{37}^{n,2n}(E)$  = energy-dependent  $^{237}\text{Np}(n,2n)$  cross section

$\chi(E)$  =  $^{235}\text{U}$  fission neutron spectrum

The calculated cross section is dependent on the assumed fission neutron spectrum. The form of the fission spectrum used was the Watt distribution<sup>6</sup>

$$\chi(E) = \exp \{-(E_f+E)/T\} \sinh [2(EE_f)^{1/2}/T]/(\pi E_f T)^{1/2} \quad (2)$$

where

$E_f$  = fragment kinetic energy per nucleon, MeV

$T$  = nuclear temperature of the fragment,  $2/3 (\bar{E}-E_f)$

$\bar{E}$  = average fission neutron energy, MeV

Although the accuracy of the (n,2n) cross section is dependent on the accuracy of this absolute fission spectrum, the accuracy of the product of the calculated (n,2n) cross section and integral of the fission spectrum above 6.8 MeV, to give a predicted reaction rate, is independent of the fission spectrum. Therefore, the  $^{237}\text{Np}(n,2n)$  cross section given in Table II is to be used only with the Watt distribution of the fission spectrum.  $^{237}\text{Np}(\gamma,n)$  cross sections reported previously<sup>4,5</sup> are also listed for convenience and comparison.

TABLE II

$^{237}\text{Np}(\gamma,n)$  and (n,2n) Cross Sections

Energy groups (MeV)	<u>6.8-8.0</u>	<u>8.0-10.0</u>	<u>6.8-25</u>
$^{237}\text{Np}$ cross sections (mb) <sup>a</sup>			
( $\gamma,n$ )	14	68	
(n,2n)			63 <sup>b</sup>

<sup>a</sup>Cross sections for 22-hr half-life isomer, half of which  $\beta$ -decays to  $^{236}\text{Pu}$ .

<sup>b</sup>Averaged over fraction of fission spectrum greater than 6.8 MeV. This fraction was calculated to be 0.0168 for  $^{235}\text{U}$  thermal neutron fission.

The value of 63 mb for the  $^{237}\text{Np}(n,2n)$  cross section is larger by 16 mb than the value reported previously.<sup>4,5</sup> However, the new value represents a significant improvement in accuracy because it

was determined from a well-defined irradiation environment and accurately measured initial and final contents of the fuel. The total error in the calculated value was determined to be  $\pm 10\%$ .

### TESTS OF CROSS SECTIONS

$^{236}\text{Pu}/^{238}\text{Pu}$  ratios were calculated for a variety of irradiation conditions and were reported previously.<sup>4</sup> The summary of results are given in Table III and corrected where necessary for the improved  $^{237}\text{Np}(n,2n)$  cross section. The  $^{236}\text{Pu}/^{238}\text{Pu}$  ratio calculated for YANKEE PWR fuel was a substantial test of these cross sections because all were derived from data obtained in well-moderated  $\text{D}_2\text{O}$  reactor charges containing  $^{237}\text{Np}$  targets. The various contributions to PWR  $^{236}\text{Pu}/^{238}\text{Pu}$  ratio are given in Table IV. Cross sections of nuclides used in burnup calculations are listed in Table V.

TABLE III

Calculated and Measured  $^{236}\text{Pu}/^{238}\text{Pu}$  Ratios

<u>Irradiation Environment</u>	<u><math>^{236}\text{Pu}/^{238}\text{Pu}</math> Ratio (ppm)</u>	
	<u>Calculated</u>	<u>Measured</u>
Savannah River Plant		
D <sub>2</sub> O Reflector - High Flux Charge		
- Heavy Al Target	0.78	0.76
- Light Al Target	0.30	0.30
In-Core - Mixed Lattice		
- Heavy Al Target	1.32	1.28
- Light Al Target, "islands" <sup>a</sup> two lattice positions from fuel	0.48	0.47
Power Reactor		
PWR Fuel (YANKEE)	9.51	9.2 $\pm$ 1

<sup>a</sup>"Islands" of neptunium targets were formed by removing assemblies adjacent to the targets.

TABLE IV

Calculated and Measured  $^{236}\text{Pu}/^{238}\text{Pu}$  Ratio  
for YANKEE Core I Fuel (ppm)

Calculated				Measured
$(\gamma, n)$ <8 MeV	$(\gamma, n)$ >8 MeV	$(n, 2n)$ >6.8 MeV	Total	Total
0.875	1.96	6.67	9.51	9.2 ± 1

TABLE V

List of Nuclides and Cross Sections

Nuclides	Cross Sections (barns)			
	$\sigma_{2200}$		Resonance Integral <sup>a</sup>	
	Capture	Fission	Capture	Fission
$^{235}\text{U}$	99.8	582.2	131	251
$^{236}\text{U}$	5.12	0	346	1.96
$^{237}\text{U}$	476.0	2.0	282	2.84
$^{238}\text{U}$	2.71	0	269	1.11
$^{236}\text{Np}$	0	0	0	0
$^{237}\text{Np}$	170	0.02	473	4.24
$^{238}\text{Np}$	200	2000	89.9	810
$^{236}\text{Pu}$	0	170	39.9	359
$^{238}\text{Pu}$	546.5	16.3	135	20.8

<sup>a</sup>Low energy cutoff of 0.625 eV; also assumes 1/E neutron spectrum.

## ACKNOWLEDGMENTS

The authors wish to thank Mr. C. J. Banick for his invaluable assistance and cooperation in measuring the  $^{236}\text{Pu}/^{238}\text{Pu}$  ratios and Mr. L. W. Patrick for arranging for and supervising the sample selections used for measurement.

## REFERENCES

1. E. J. Hennelly, "Production of Biomedical-Grade  $^{238}\text{Pu}$ ," *Trans. Am. Nucl. Soc.*, 11, (1968).
2. G. G. Eichholz, *Radioisotope Engineering*, Marcel Dekker, Inc., New York, Chapter 2, pp 86-88 (1972).
3. C. J. Banick and M. H. Goosey, *Improved methods for Determination of  $^{236}\text{Pu}$  in  $^{238}\text{Pu}$* , USAEC Report DP-1291, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, South Carolina (1972).
4. C. K. Paulson, *Calculation of  $^{236}\text{Pu}$  in  $^{238}\text{Pu}$  Produced by  $^{237}\text{Np}$  Irradiations*, USAEC Report DP-1309, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, South Carolina (1972).
5. C. K. Paulson, "Improved Determination of  $^{236}\text{Pu}$  Formation During  $^{238}\text{Pu}$  Production, *Trans. Am. Nucl. Soc.*, 14, 15 (1971).
6. J. Terrell, "Fission Neutron Spectra and Nuclear Temperatures," *Phys. Rev.*, 113, 527 (1959).
7. R. P. Matsen, "The Determination of Ratios of Effective Cross Sections from Measured Burnup Data for Yankee Rowe," *Nuclear Technology*, 15, 343 (1972).