LAWRENCE LIVERMORE LABORATORY
University of California, Livermore, California, 94550

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THERMAL NEUTRON CAPTURE CROSS SECTION OF ^{235}Np

Jerry H. Landrum
Richard J. Nagle
Manfred Lindner

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Abstract

A neptunium sample containing roughly comparable levels of ^{235}Np , $^{236\text{m}}\text{Np}$, and ^{237}Np was irradiated in the thermal column of the Livermore pool-type reactor (cadmium ratio = 17) in a flux of $\sim 2 \times 10^{11}$ n/cm²/sec. A thermal capture cross section of 148 b was measured for

the reaction $^{235}\text{Np} (n, \gamma) ^{236\text{g}}\text{Np} \xrightarrow[22 \text{ hr}]{\beta^- \text{ } 50\% \text{ } K} ^{236}\text{Pu}$ by comparing the ^{236}Pu produced with the ^{238}Pu produced by the reaction $^{237}\text{Np} (n, \gamma) ^{238}\text{Np} \xrightarrow{2d\beta} ^{238}\text{Pu}$. The latter cross section was taken to be 170 b. It was assumed that 50% of the $^{236\text{g}}\text{Np}$ decays resulted in β^- emission to ^{236}Pu .

Introduction

The preparation of a submicrogram sample of chemically pure neptunium that contained the nuclides 235 , $^{236\text{m}}$, and 237 initially in the atom ratio 1.0:0.23:0.058, as determined by mass spectrometric analysis, has been reported.¹ Since the half-life of ^{235}Np is 396 days,² there appeared to be an opportunity to measure the thermal neutron partial capture cross section for the reaction:

It was not possible, by our method, to measure the cross section for formation of the long-lived isomer $^{236\text{m}}\text{Np}$, since, in any case, the sample already contained the isomer in 25% abundance.

Additional ^{237}Np was added to serve as a flux monitor through measurement of the reaction

An advantage of this method was that after irradiation, the sample was allowed to stand until both the 22-hr $^{236\text{g}}\text{Np}$ and the 50-hr ^{238}Np had decayed completely to the alpha-emitting nuclides ^{236}Pu and ^{238}Pu , respectively. Isolation and purification of a plutonium chemical fraction and subsequent alpha pulse height analysis yielded the ratio $^{236}\text{Pu}/^{238}\text{Pu}$. The cross section ratio was then calculated from the expression

$$\frac{\sigma \text{ to } ^{236}\text{Pu}}{\sigma \text{ to } ^{238}\text{Pu}} = \frac{^{236}\text{Pu}/^{235}\text{Np}}{^{238}\text{Pu}/^{237}\text{Np}} \quad (1)$$

A value of 170 b was assumed for the cross section for ^{238}Pu formation.³ To calculate the actual cross section for $^{236\text{g}}\text{Np}$ formation, it was assumed that 50% of the 22-hr $^{236\text{g}}\text{Np}$ decayed by beta emission to ^{236}Pu .

Preparation and Procedure

A known amount of ^{237}Np was added to two 20- μl samples, each containing about 1.25 μg of ^{235}Np as well as lesser quantities of ^{238}Np and ^{237}Np . To the first sample was added about 1 to 1.5 μg of ^{237}Np and to the second, about 4 to 4.5 μg . Both samples were then treated by the following chemical purification procedure to remove all detectable traces of ^{238}Pu or ^{236}Pu :

1. To the solution were added 2 ml of concentrated HCl and 4 drops of concentrated HCl. The solution was boiled to near-dryness.
2. Two ml of concentrated HCl were added and the solution passed through a Dowex 1X8, 3 mm i.d. $\times$ 5 cm (200-400 mesh) anion exchange resin column pretreated with concentrated HCl. The columns were washed with 1 ml of concentrated HCl.
3. The neptunium was eluted from the column with 2 ml of 5 N HCl.
4. The 5 N HCl eluate was diluted to 1 N HCl and the neptunium extracted into 0.4 M thenoyltrifluoroacetone (TTA) solution in benzene.
5. The TTA layer was treated with

9 M HCl to back-extract the neptunium fraction.

At this point, the two purified samples in 9 M HCl were further divided into roughly equal portions. One portion of each sample was transferred to a quartz tube; the solution was evaporated to dryness and the tube sealed. The two sealed samples were then irradiated.

The remaining portion of each sample was boiled to near-dryness and a few drops of concentrated HNO_3 added. This mixture was boiled to near-dryness and 1 ml of concentrated HCl was added. The HCl was also boiled to near-dryness, cooled, and a few drops of formic acid were added with gentle warming. Several milliliters of concentrated HCl were added and the solution passed through a Dowex 1X8 anion exchange column (200 to 400 mesh), both U^{+6} and Np^{+4} being retained on the resin. The Np^{+4} was then eluted with 0.5 ml of 5 N HCl. These two samples, thus separated from traces of uranium, were not irradiated, but were submitted for immediate mass spectrometric isotope ratio analysis before ^{235}U growth became significant.

Neutron Irradiation

Because both the thermal flux and the weight of the target material were very low, to produce a detectable level of the alpha-emitting product nuclides the samples were irradiated for a total of 240 hr over a period of about 1 month in the thermal column of the Livermore Pool Type Reactor (LPTR). In this facility, the cadmium ratio was 17 and the thermal

flux was approximately $2 \times 10^{11}/\text{cm}^2/\text{sec}$. Corrections were made for the decay of both ^{235}Np and ^{236}Pu during the irradiation.

After the irradiation, the samples were allowed to stand for several weeks until decay of the parent isotopes ^{236}Np and ^{238}Np was "complete." The quartz vials were opened and both samples were

subjected to the following chemistry for the isolation and purification of the plutonium fraction:

1. The contents of the vial were removed by repeated treatment with warm 6 N HCl. To the combined HCl leachings was added 1 drop of HI, and the entire contents was then boiled to near-dryness.

2. One ml of concentrated HCl-0.5 N HI was added and the solution was passed through a column of Dowex 1X8 (100-200 mesh) anion exchange resin; the neptunium remained on the column. The column was washed with concentrated HCl-0.5 N HI; the wash was collected with the eluate. The neptunium fraction was treated as in step 8, below.

3. The eluate-plus-wash was boiled to near-dryness and 0.5 ml of concentrated HNO_3 added. The resultant solution was boiled to near-dryness and taken up in concentrated HCl.

4. The solution was passed through a Dowex 1X8 anion exchange column and the column washed with concentrated HCl. Eluate and wash were discarded.

5. The plutonium was eluted with 3 ml of concentrated HCl-0.5 N HI and the eluate was boiled to near-dryness.

6. One-half ml of concentrated HNO_3 was added and the resultant solution boiled to near-dryness. The excess HNO_3 was destroyed by repeated boiling with concentrated HCl.

7. The solution was made just acid to methyl red and the plutonium plated on a 1-in.-diameter platinum cathode disk at 1.5 A for 30 min.

8. The column containing the neptunium fraction (from step 2) was washed with concentrated HCl and the neptunium eluted with 5 N HCl.

9. The 5 N HCl was boiled to near-dryness and a U-Np separation like that described above (see Preparation and Procedure) was carried out.

(The final uranium-free samples of irradiated neptunium were also mass-analyzed for comparison of the isotope ratio with the unirradiated sample.)

The alpha-particle pulse-height spectra for both plutonium samples were measured on a silicon surface barrier detector. The spectrum for one of the samples is shown in Fig. 1. The atom ratio $^{236}\text{Pu}/^{238}\text{Pu}$ was calculated from the alpha pulse-heights (αPH) using the expression

$$\frac{^{236}\text{Pu}}{^{238}\text{Pu}} = \frac{\alpha\text{PH}(236)}{\alpha\text{PH}(238)} \times \frac{T_{1/2}(236)}{T_{1/2}(238)} \quad (2)$$

Fig. 1. Pulse-height spectrum of plutonium fraction from $^{235,6,7}\text{Np}$ irradiated with thermal neutrons.

The half-lives used were 2.851 yr and 86.41 yr for ^{236}Pu and ^{238}Pu , respectively.⁴

Table 1 shows the alpha pulse-height ratios for the two samples of plutonium, corrected for decay to the end of the neutron irradiation.

The initial addition of ^{237}Np to the ^{235}Np solution was necessary to obtain a pulse-height spectrum in which the ^{238}Pu peak was comparable to that of the ^{236}Pu . Since the ^{235}Np capture cross section

could only be guessed, the anticipated ^{236}Pu peak was in doubt. Something closer to unity would have been preferable for the ratio.

Table 1. $^{236}\text{Pu}/^{238}\text{Pu}$ alpha pulse-height ratios.

Plutonium sample	Ratio	Std. dev., %
1B	10.93	2.46
2B	3.99	1.70

Mass Spectrometry

The replicate halves of both the irradiated and unirradiated neptunium samples were analyzed on a double-focusing, 60-deg sector-type mass spectrometer of 12 in. radius. At least six determinations of the mass ratio $^{237}\text{Np}/^{235}\text{Np}$ were made on each sample. The overall averages are given in Table 2.

The reference time for all calculations and decay corrections was taken as the end of the neutron irradiation.

Table 2 shows that there was no difference in the mass ratios for irradiated

and unirradiated samples. Further, it confirms the essential significance of the mass-spectrometric method for neptunium isotope ratio determinations.

Table 2. Average $^{237}\text{Np}/^{235}\text{Np}$ mass ratios

Neptunium sample	Irradiated	Mass ratio	Std. dev., %
1A	no	1.189	0.51
1B	yes	1.195	0.55
2A	no	3.361	0.50
2B	yes	3.339	0.53

Cross Section for the Reaction $^{235}\text{Np} (n, \gamma) ^{236}\text{Np}$ 22 hr

Two independent values for the cross section ratio for production of ^{236}Pu and ^{238}Pu , result from Tables 1 and 2, and are given in Table 3.

The average cross section ratio is thus $0.434 \pm 1.5\%$. If it is assumed that just

Table 3. Cross section ratio ($^{235}\text{Np} \rightarrow ^{236}\text{Pu}/^{237}\text{Np} \rightarrow ^{238}\text{Pu}$) for thermal neutrons.

Sample	$\sigma(6)/\sigma(8)$	Std. dev., %
1	0.428	2.5
2	0.438	1.3

50% of the ^{236}Np decays by beta emission to $^{236}\text{Pu}^4$ and that the ^{237}Np capture cross section is 170 b, then the thermal-

neutron capture cross section leading to the 22-hr ^{236}Np isomer is
 $\sigma(235) = 2 \times 170 \times 0.434 = 148 \text{ b } (\pm 1.5\%)$.

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