

$(n, 2n)$ Cross Sections for ^{238}U and ^{237}Np in the Region of 14 MeV*

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(Received 11 December 1972; revised manuscript received 22 May 1973)

The cross sections for formation of ^{237}U from ^{238}U and for the eventual formation of ^{236}Pu from ^{237}Np by the $(n, 2n)$ reaction were measured relative to the $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ cross section. Neutrons were produced by the irradiation of titanium tritide targets with 400-keV deuterons on the Livermore insulated core transformer (ICT) neutron generator. The determinations covered the neutron energy range from 13.7 to 14.9 MeV. From the partial cross section for ^{236}Pu formation, the cross section was calculated for the ^{237}Np total $(n, 2n)$ reaction, including the branch leading to $^{236}\text{Np}^m$.

I. INTRODUCTION

The long half-life and the availability of large quantities of ^{237}Np make possible the measurement of neptunium cross sections for a variety of nuclear reactions by the activation techniques generally used with common stable isotopes. We previously reported measurements of the neutron-capture cross section for ^{237}Np between 100 keV and 3 MeV, using calibrated Ge(Li) detectors to count the 980- and 1030-keV γ rays emitted by the capture product ^{238}Np .¹ Perkin and Coleman² measured the $(n, 2n)$ cross section at 14.5 MeV, relative to the reaction $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$, for ^{236}Pu production from ^{237}Np arising from the β^- decay of the 22-h $(n, 2n)$ product $^{236}\text{Np}^e$. We have used a technique similar to that of Perkin and Coleman to measure the neptunium cross sections between 13.7- and 14.9-MeV neutron energy for the same reaction. As a comparison and a check on the reliability of our flux measurements, we simultaneously measured the cross section for the reaction $^{238}\text{U}(n, 2n)^{237}\text{U}$ over the same energy range.

For purposes of cross-section calculations, we used half-lives of 2.14×10^6 yr and 2.85 yr for ^{237}Np and ^{236}Pu , respectively. The half-life used for ^{237}U was 6.75 day, and that for ^{24}Na was 15.0 h.

II. EXPERIMENTAL PROCEDURE

A. Preparation of Target Materials

Because of the small size of the neptunium samples and the necessity for careful determination of the α -particle emitters present in low abundance in the preirradiated neptunium, special precautions had to be taken to ensure the authenticity of the ^{236}Pu detected in the irradiated neptunium samples. We therefore explored the α content in detail, of the preirradiated, as well as the postirradiated neptunium.

As received from the supplier, neptunium can be expected to contain varying amounts of ^{238}Pu and ^{239}Pu . Even though our material had received

special purification before receipt, we found, upon pulse-height analysis of the neptunium, that roughly 0.9% of the total α activity was due to ^{238}Pu . No other α emitters could be detected in this relatively gross measurement. The neptunium was then subjected to the separation procedure described in the Appendix for the thorough separation of plutonium from neptunium. α pulse-height analyses were made on both the repurified neptunium fraction and the plutonium fraction removed from the neptunium. It was found that the final ^{238}Pu content of our repurified neptunium was so low that only an upper limit of 1.4×10^{-5} could be set for the α ratio $^{238}\text{Pu}/^{237}\text{Np}$ (this figure was later determined on the postirradiated samples, with some precision, to be 3.6×10^{-6}).

The pulse-height spectrum of the plutonium fraction removed from the unpurified ("as-received") neptunium, is given in Fig. 1. It is clear that both ^{239}Pu and ^{236}Pu were present in low abundance in the original neptunium, along with the ^{238}Pu . The relative α abundances found were $^{238}\text{Pu} : ^{239}\text{Pu} : ^{236}\text{Pu} = 1.00 : 1.44 \times 10^{-2} : 4.48 \times 10^{-5}$. Thus, the α ratio $^{236}\text{Pu}/^{237}\text{Np}$ of the original neptunium was 4×10^{-7} , and that of the repurified neptunium was 1.6×10^{-10} . It was particularly important to establish the latter since the preirradiation level of ^{236}Pu constituted a correction to the ^{236}Pu produced in the irradiation. All of the neptunium target samples that we irradiated were made from this highly purified material (containing less than 0.001 dis/min of ^{236}Pu per milligram of neptunium).

Measured volumes of a solution containing 3.214 mg of purified neptunium were placed into each of five separate containers. To each was added a solution in HCl containing 146 α dis/min of ^{242}Pu tracer (as well as about 3 dis/min each of ^{238}Pu and ^{239}Pu and less than 0.001 dis/min of ^{236}Pu associated with the ^{242}Pu). The resulting mixture was evaporated to dryness, dissolved in a small volume of 10 N HCl, and transferred onto a filter paper that was 2.54 cm in diameter. The container was rinsed with HCl, and the rinse was added

to the filter paper. The solution thus absorbed on the filter paper was evaporated to dryness at very low heat. The dried paper was then sealed in a small envelope of polyvinylchloride film. Five identical specimens thus prepared served as neptunium targets for neutron irradiation, each containing less than 0.004 dis/min of ^{236}Pu .

The seven ^{238}U targets were 0.25-mm metal foils, each of them 2.5 cm in diameter. These were cleaned in dilute nitric acid, dried, weighed, and then sealed in thin polyvinylchloride film for handling and irradiation.

The neutron fluxes were measured from the ^{24}Na produced by $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ in weighed circular aluminum foils.

B. Neutron Irradiation

Neutrons were generated by the reaction $^3\text{H}(d, n)^4\text{He}$ with the 10-mA beam of 400-keV deuterons impinging on a thick target of titanium tritide in the Livermore ICT. The five neptunium, six aluminum, and seven uranium targets were arrayed at various angles from 10 to 120° on the surface of a machined 1.3-mm-thick aluminum sphere of 20.3 cm radius. The neutron source was located at the center of the sphere, and the samples were irradiated for approximately 14 h.

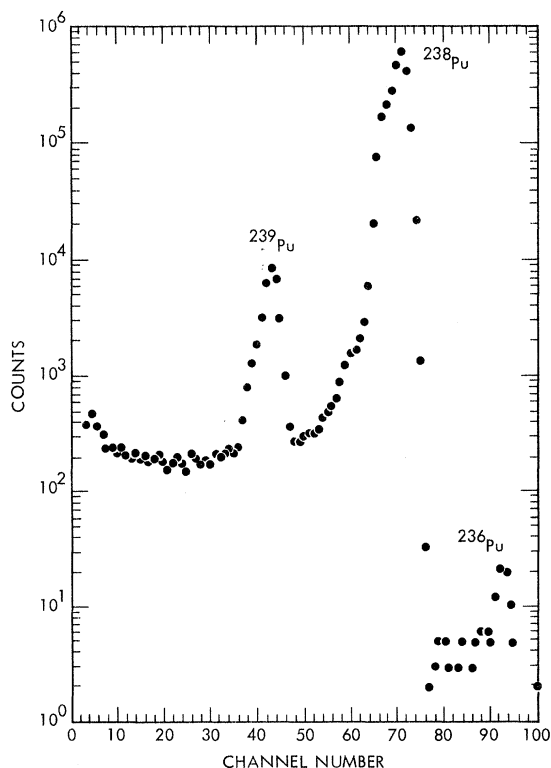


FIG. 1. α pulse-height spectrum of plutonium impurity in unpurified neptunium.

C. Postirradiation Activation Determination

The 15-h ^{24}Na produced in each of the aluminum foils was measured with a sodium iodide scintillation detector that had been previously calibrated against a 4π β - γ coincidence counter. A correction was made for the decay of the ^{24}Na during the irradiation.

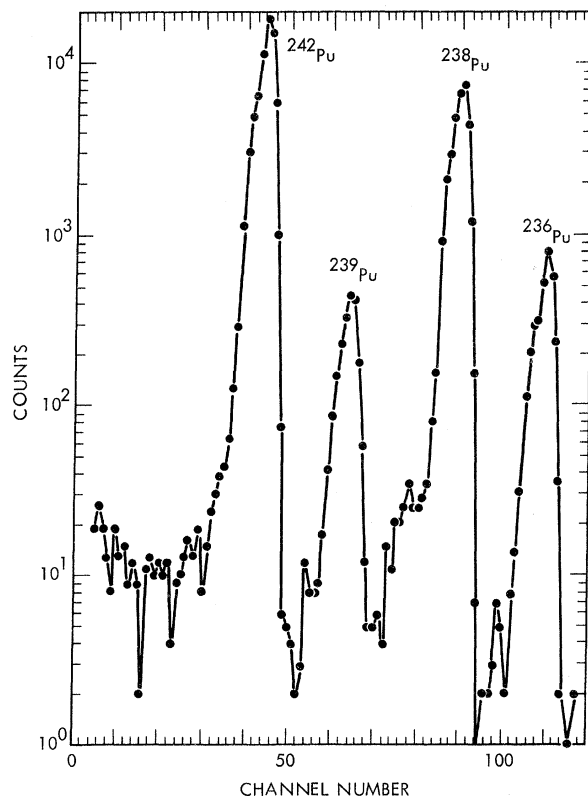
The uranium foils were dissolved, and 10% aliquots were taken for determination of the ^{237}U produced in each foil. This was achieved by measurement of the intensity of the 207.8-keV γ transition on a Ge(Li) detector coupled to a 4096-channel pulse-height analyzer. The Ge(Li) detector photopeak efficiency had been previously calibrated with a set of standard point sources obtained from the International Atomic Energy Agency in Vienna. Corrections were made for γ -ray attenuation in the solution and in the cap of the solution container. Corrections were also made for purely geometrical effects due to the finite dimensions of the source and detector. Because the $(n, 2n)$ cross section for ^{238}U is known to be comparable to its fission cross section at the energies investigated here,^{3,4} it could be confidently anticipated that ^{237}U would be a major activity component at the time of the pulse-height analysis. For this reason, and also because of the high resolution obtainable with our Ge(Li) system, no postirradiation chemical purification was performed on the uranium solutions; nor was the half-life of the photopeak checked by decay measurements. Instead, the complex γ -ray spectrum was analyzed by the least-squares peak intensity analysis and identification code developed by Gunnink, Levy, and Niday.^{5,6} Results of this computerized analysis confirmed the assumption that ^{237}U was a major component and that only a very minor, but accountable, contribution to the pertinent 207.8-keV photopeak was made by the 209.7-keV γ -ray photopeak due to ^{239}Np , which may have been produced by capture of room-scattered neutrons on the uranium foils. All other radionuclides with photopeaks that could possibly have interfered with the 207.8-keV γ ray were eliminated by the computer program on the basis of the absence of the associated γ rays that would have had to be present.

The irradiated neptunium samples were allowed to stand for several weeks to ensure the complete decay of the 22-h ^{236}Np to ^{236}Pu . The samples were then opened and a plutonium chemical fraction was isolated and purified from the neptunium (the radiochemical separation is given in the Appendix). The α -particle intensities of the ^{236}Pu produced during the irradiation and of the ^{242}Pu tracer added before the irradiation were determined on a silicon surface-barrier detector cou-

TABLE I. Conversion ratios for aluminum flux monitors.

Angle (deg)	$\langle E \rangle$ (MeV)	ΔE (MeV)	$10^{13} {}^{24}\text{Na}/{}^{27}\text{Al}$	% Standard deviation
14.5	14.94	0.62	7.70	0.14
44.6	14.72	0.50	7.66	0.16
64.3	14.46	0.33	7.66	0.18
90.5	14.09	0.10	7.83	0.14
104.9	13.90	0.0	7.58	0.15
113.9	13.77	0.10	7.44	0.15

pled to a 512-channel pulse-height analyzer. A typical α pulse-height spectrum of the plutonium fraction from one of the irradiated neptunium samples is shown in Fig. 2. It is clear that, in addition to ${}^{236}\text{Pu}$ and ${}^{242}\text{Pu}$, the spectrum contained α peaks due to ${}^{238}\text{Pu}$ and ${}^{239}\text{Pu}$. The ${}^{238}\text{Pu}$ was just the residual level not removed in the Np-Pu purification step, while the ${}^{239}\text{Pu}$ resulted from both that source and from the residual level in the ${}^{242}\text{Pu}$ tracer. Both of these "contaminant" peaks were ignored. The ${}^{236}\text{Pu}$ in the samples before irradiation was less than 0.1% of that produced in the irradiation and could be ignored.

FIG. 2. α pulse-height spectrum of the plutonium fraction from purified irradiated neptunium.

III. RESULTS

The conversion ratios, ${}^{24}\text{Na}/{}^{27}\text{Al}$, measured at the listed lab neutron angles are given in Table I. Included also are the corresponding average thick-target neutron energies and the energy spreads taken from the work of Seagrave *et al.*⁷ Since the neutron energy distribution calculated by those authors for a given angle resembles a truncated Maxwellian distribution, the meaning of the spread ΔE is somewhat arbitrary. We have chosen to define it as the absolute value of the energy difference between the average neutron energy $\langle E \rangle$ and the limiting truncation energy. We reproduce in Fig. 3 a few typical calculated energy distributions, as given by Seagrave *et al.*,⁷ for lab angles 0, 90, and 150° for a deuteron energy of 400 keV.

The values given in Table I for $\langle E \rangle$ and ΔE were generally not available from Ref. 7 for the angles at which our measurements were made. Instead, these quantities were interpolated from their angular dependence, as plotted from the data taken from Ref. 7.

It can be easily shown for any flux distribution $\phi(E)$, that if a function σ vs E is approximately linear over the range ΔE , then the spectrum-averaged cross section, $\bar{\sigma}$, for a given average neutron energy, $\langle E \rangle$, is a good approximation to the microscopic cross section at the neutron energy that is nominally equal to the average neutron energy. Here, $\langle E \rangle = (1/\Phi) \int \phi(E)E dE$, $\bar{\sigma} = (1/\Phi) \times \int \phi(E)\sigma(E)dE$, and $\Phi = \int \phi(E)dE$. Since a linear relationship for $\bar{\sigma}$ vs $\langle E \rangle$ appears valid from our measurements, our reported average quantities can be interpreted as the microscopic cross sections and energies.

In this investigation we have reported our results on ${}^{238}\text{U}$ and ${}^{237}\text{Np}$ primarily as the ratio of

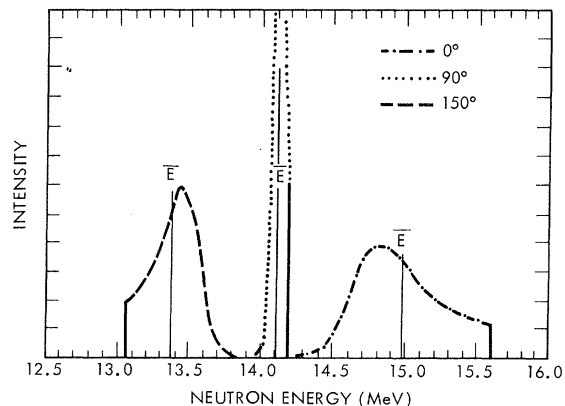
FIG. 3. Neutron energy distributions from Ref. 7 at several laboratory angles for the reaction ${}^3\text{H}(d, n){}^4\text{He}$ for 400-keV deuterons.

TABLE II. Cross sections and cross-section ratios for the reaction $^{238}\text{U}(n, 2n)^{237}\text{U}$.

Angle (deg)	$\langle E \rangle$ (MeV)	$10^{12} (^{237}\text{U}/^{238}\text{U})$	$10^{12} (^{24}\text{Na}/^{27}\text{Al})$	$\sigma(\text{U})/\sigma(\text{Al})$	% Standard deviation	$\sigma(\text{Al})$ (b)	$\sigma(\text{U})$ (b)	% Standard deviation
17.2	14.94	4.77	0.770	6.20	3.7	0.110	0.68	3.9
45.6	14.70	4.78	0.766	6.24	4.6	0.112	0.70	4.8
69.4	14.39	5.40	0.768	7.02	4.1	0.118	0.83	4.3
90.8	14.08	5.56	0.783	7.10	4.1	0.121	0.86	4.3
95.9	14.01	5.77	0.773	7.47	3.6	0.121	0.90	3.8
102.6	13.92	6.37	0.761	8.52	4.1	0.121	1.03	4.3
118.2	13.70	5.94	0.741	8.01	4.4	0.124	1.00	4.6

the particular cross section to that of the monitor reaction $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$. We chose to do this because the absolute ^{27}Al cross sections are always subject to new measurements and to changes in old values. Secondly, we calculated the absolute cross sections, based upon the $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ cross sections taken from the excitation function evaluated by Nethaway.⁸ The accurate measurements of Vonach *et al.*⁹ were the basis for Nethaway's evaluation.

Since the aluminum foils were not necessarily placed at precisely the same angles as were the uranium and neptunium foils, we plotted the data in Table I and then interpolated the required aluminum conversion ratios at the appropriate energy. The variation in the conversion ratios from angle to angle was small, so that the uncertainty introduced by interpolation was relatively unimportant.

Listed in Table II are the observed ^{238}U conversion ratios, the interpolated aluminum conversion ratios, the cross-section ratio, the assumed absolute cross section for the reaction $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ from the evaluation by Nethaway,⁸ and, finally, the resultant cross section for the reaction $^{238}\text{U}(n, 2n)^{237}\text{U}$.

Similarly, in Table III, we list the corresponding data for the formation cross section and cross-section ratios for ^{236}Pu .

Because the cross sections listed in the eighth column of Table III are only for the β^- decay branch of the 22-h isomer $^{236}\text{Np}^{\epsilon}$, we have as-

sumed equal branching for β^- and orbital capture¹⁰ decay, to arrive at the formation cross sections for the 22-h isomer. These cross sections are listed in the last column of Table III. The total (n, 2n) cross section (for both isomers) has been estimated and is listed in Table IV. The method of estimation is explained in the following section.

IV. DISCUSSION

Our data for uranium are plotted in Fig. 4 for cross sections based on Nethaway's⁸ evaluation of the cross-section measurements for the $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ reaction. Data of other investigators are shown for comparison. Our results appear to agree quite closely with those of Graves, Conner, Ford, and Warren as reported by Knight *et al.*³ and perhaps less well with the single data points of some of the other investigators.

The last column of Table III gives the calculated (n, 2n) cross sections for formation of the isomer $^{236}\text{Np}^{\epsilon}$, based upon an (EC/ β^-) branching ratio of 1.0. It is apparent that our value, interpolated at 14.5 MeV, is 0.31 b. It is at this energy that the only other measurement, that of Perkin and Coleman,² was made; they reported a cross section of 0.39 ± 0.06 b, a value 25% larger than our own. Because of the listed uncertainties, this is reasonable agreement. However, Perkin and Coleman further assumed that this represented essentially the total (n, 2n) cross section, reasoning that the formation

TABLE III. Cross sections and cross-section ratios for the reaction $^{237}\text{Np}(n, 2n)^{236}\text{Np}^{\epsilon}$ near 14 MeV.

Angle (deg)	$\langle E \rangle$ (MeV)	$10^{12} (^{236}\text{Pu}/^{237}\text{Np})$	$10^{12} (^{24}\text{Na}/^{27}\text{Al})$	$\frac{\sigma(\text{to } ^{236}\text{Pu})}{\sigma(\text{Al})}$	% Standard deviation	$\sigma(\text{Al})$ (b)	$\sigma(\text{to } ^{236}\text{Pu})$ (b)	% Standard deviation	$\sigma(\text{to } ^{236}\text{Np}^{\epsilon})$ (b)
11.3	14.95	0.94	0.771	1.22 ₄	3.5	0.110	0.134	3.7	0.27
42.5	14.74	0.97	0.766	1.27 ₂	3.4	0.112	0.142	3.5	0.28
69.0	14.39	1.03	0.768	1.34 ₈	3.4	0.118	0.158	3.5	0.32
88.0	14.12	1.12	0.781	1.43 ₂	3.8	0.121	0.172	4.0	0.34
113.2	13.77	1.18	0.744	1.58 ₆	3.4	0.124	0.196	3.6	0.40

TABLE IV. Inferred total $(n, 2n)$ cross sections for ^{237}Np near 14 MeV.

$\langle E \rangle$ (MeV)	Total $\sigma(n, 2n)$ (b)	% Standard deviation
14.95	0.37	7.2
14.74	0.39	7.1
14.39	0.44	7.1
14.12	0.47	7.4
13.77	0.54	7.2

cross section for the long-lived isomer $^{236}\text{Np}^m$ (Ref. 16) was probably negligible. From other experiments,¹⁷ we found that, when ^{237}Np was exposed to an intense thermonuclear neutron flux in a number of nuclear devices, both ^{236}Pu and $^{236}\text{Np}^m$ could be detected in the neutron reaction products. The ^{236}Pu was determined by the α pulse analysis of a plutonium fraction, and the $^{236}\text{Np}^m$ by the mass-

spectrometric analysis of a neptunium fraction. From the analyses of such samples from a number of devices, we have found that the value for the ratio $^{236}\text{Np}^m/^{236}\text{Pu}$ was (0.76 ± 0.03) . Thus, the value for the ratio $^{236}\text{Np}(\text{total})/^{236}\text{Pu}$ becomes 2.76. If this ratio is assumed to hold for the irradiation in the region of 14 MeV in the present work, then we need only multiply each observed ^{236}Pu cross section by 2.76. This was done in Table IV, and the resultant values are plotted in Fig. 5.

An attempt has been made to compare these measurements with the calculated values of Pearlstein¹⁸ and of Smith and Grimesey.¹⁹ This comparison is shown in Fig. 5. The lack of agreement between the measured points and calculated curves is striking. Pearlstein's calculations depended upon proper choice of values for the net nonelastic cross section. The values were then multiplied by calculated factors to obtain the part that was due to the $(n, 2n)$ cross section. The net nonelastic cross section, in turn, was taken to be the difference between the measured nonelastic and fission cross sections (as well as a small correction for direct interaction). Since this leads to a difference between two fairly large numbers, it is possible that errors as large as a few hundred millibarns might be involved. As a matter of fact, Smith and Grimesey show that various experiments disagreed by as much as several hundred millibarns in the values reported for the fission cross section of ^{237}Np . Furthermore, Pearlstein used a nonelastic cross section taken from a smooth curve, from which measured values seem to differ by hundreds of millibarns. Therefore, the serious disagreement between our measured values and Pearlstein's calculations should not be surprising.

Smith and Grimesey also calculated the $(n, 2n)$ cross section for ^{237}Np cross-section data available up to 1969,¹⁹ and their calculated curve is reproduced in Fig. 5 as well as possible from that evaluation. It is, like the Pearlstein curve, lower than our adjusted experimental cross sections.

We have investigated the possibility that the serious disagreement might be caused by the previous assignment of an (EC/β^-) ratio of about 1.0 for the decay of $^{236}\text{Np}^f$, since such a branching ratio automatically doubles the cross section that we have found for the formation of ^{236}Pu . Gindler and Sjöblom²⁰ determined an (EC/β^-) ratio of 1.05 ± 0.17 . The β^- branch was determined by ^{236}Pu α growth, while the electron capture branch was determined by a mass-spectrometric measurement of the growth of ^{236}U . The latter clearly would circumvent the uncertainties associated with interpretation of γ -ray, x-ray, and electron-spectrum data. Unfortunately, the pregrowth level of ^{236}U in their purified Np sample was such that the

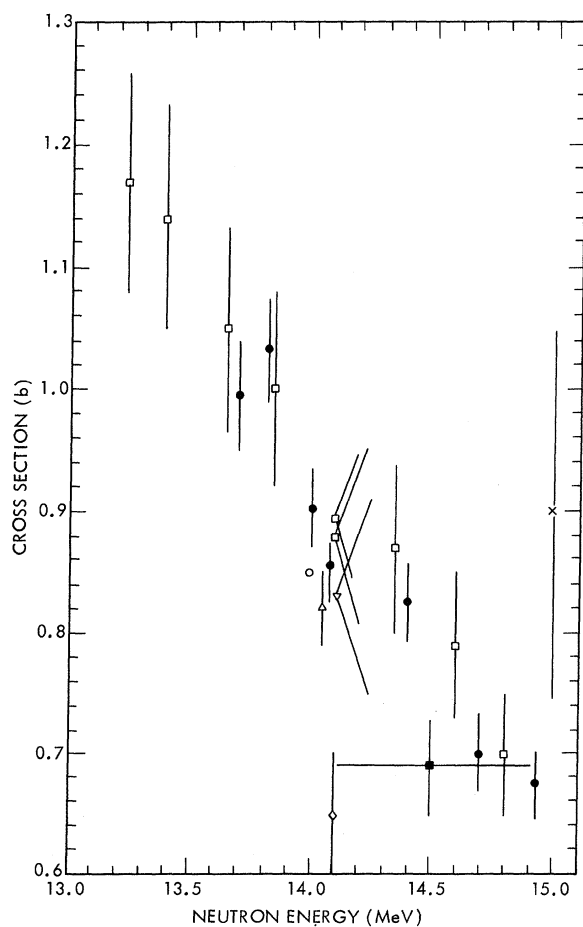


FIG. 4. Cross section for $^{238}\text{U}(n, 2n)^{237}\text{U}$. Symbols for the data points are as follows: \bullet denotes this work; \blacksquare denotes Ref. 2; \square denotes Ref. 3; \triangle denotes Ref. 11; \times denotes Ref. 12; \circ denotes Ref. 13; ∇ denotes Ref. 14; \diamond denotes Ref. 15.

^{236}U observed to grow from the $^{236}\text{Np}^f$ decay represented only about a 10% increase. This difficulty arose because a factor of 10^7 purification from uranium was barely sufficient for their observation of ^{236}U growth. From their error limits, however, even this small growth increase was apparently significant.

Gray²¹ made a thorough study of the conversion- and Auger-electron spectra as well as the x rays associated with the decay of $^{236}\text{Np}^f$. Again, by a measurement of the ^{236}Pu α growth, and the intensity of uranium K x rays (he found no transitions of energy exceeding 44 keV associated with $^{236}\text{Np}^f$ decay), he found a K/β ratio of 0.75.

It thus seems that the adjustment in cross section that we made to allow for the K-capture branch in the $^{236}\text{Np}^f$ decay is reasonably sound, and we have concluded that the calculations of Pearlstein¹⁸ differ because of the uncertainties in the measured cross sections that he used in his calculations.

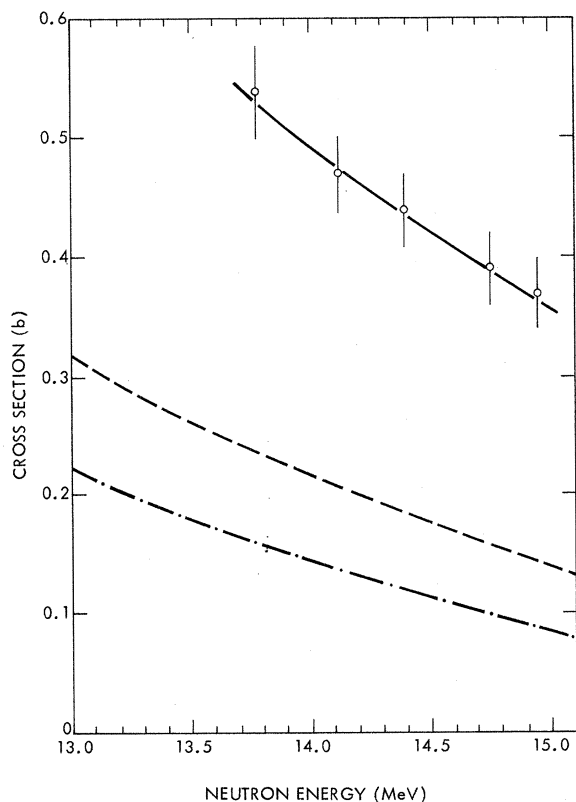


FIG. 5. Cross section for $^{237}\text{Np}(n, 2n)^{236}\text{Np}$. Symbols for the data points are as follows: \circ denotes the experimental data of this work; --- denotes Ref. 18, semi-empirical calculations; - · - denotes Ref. 19, semi-empirical calculations.

V. ERRORS AND UNCERTAINTIES

The uncertainties may be classified as those introduced by our own measurements and those introduced from associated measurements made by other investigators. It was precisely to avoid dependence on the measurements of others that we consider our basic measurement to be the cross section relative to that of the aluminum monitor, rather than the absolute value of the cross section itself.

The principal sources of random error in our work were those generated by the statistical uncertainties of the radionuclide measurements, and the results of these were shown in Tables II and III as the percent standard deviation of the ratios $\sigma(\text{U})/\sigma(\text{Al})$ and $\sigma(^{236}\text{Pu})/\sigma(\text{Al})$, respectively. The magnitude of these random errors is less than 5%. Uncertainties arising from weighing and from angular alignment of targets during irradiation were considered small in comparison with the statistical uncertainties of counting.

Although the use of "infinitely thick" titanium tritide targets gives rise to a considerable energy spread in each sample, the average neutron energy is known with considerably greater precision than the energy spread implies. Likewise, the finite sample width and neutron source width give rise to an angular spread—and thus to a spread in neutron energy. However, the average neutron energy from these effects has been shown²² to be essentially indistinguishable from that for a point neutron source and a point target. Therefore, no corrections were applied for energy-spread effects from these causes.

Uncertainties are also inherent in the aluminum monitor cross sections. A reasonable estimate of the random uncertainty should be the statistical uncertainty associated with each datum point of 1.2% reported by Vonach *et al.*⁹ We have thus adopted this value.

Two further sources of random error associated with the ^{236}Np cross-section determination are the values (1.05 ± 0.17) for the (EC/β^-) branching ratio for the $^{236}\text{Np}^f$ isomer,²⁰ and (0.76 ± 0.03) for the $(^{236}\text{Np}^m/^{236}\text{Pu})$ isotope ratio.¹⁷ These uncertainties taken together, lead to an over-all uncertainty of $\pm 6.2\%$ in the ratio $^{236}\text{Np}(\text{total})/^{236}\text{Pu}$.

There are probably systematic errors both in our measurements and in those of the monitor reaction that are very difficult to assess. It is generally assumed that most radiochemical cross-section measurements could well be inaccurate by as much as 5 or 10%. However, the errors reported in this work are random, and do not include systematic uncertainties.

APPENDIX: CHEMICAL SEPARATION OF NEPTUNIUM FROM PLUTONIUM

The source of ^{237}Np for these experiments was the oxidized coating from an 0.2-mm neptunium foil already purified from the bulk of the ^{238}Pu (which is generally associated with bulk neptunium) by the Rocky Flats Division of the Dow Chemical Company.

After dissolution of the neptunium oxide coating in an appropriate acid, the gross separation of neptunium and plutonium was accomplished by valence reduction of plutonium to Pu(III) , followed by the extraction of neptunium into a benzene solution of thenoyltrifluoroacetone. Traces of plutonium were then removed from the neptunium fraction by a series of three ion exchange separations in which

the plutonium was preferentially eluted upon chemical reduction. Essentially the same separation was employed to remove traces of neptunium from the plutonium fraction, except that in that case, it was the plutonium fraction that was recycled several times.

α -particle spectra of either neptunium or plutonium were determined only on samples of either neptunium or plutonium that were electropolated onto platinum disks. The electropolated specimens were so thin that there was no visible solid material on the disk. The α pulse analysis of the plutonium fraction thus removed from the impure neptunium is shown in Fig. 1. Since the same chemical separation was applied to the purified, but irradiated, neptunium, a typical α pulse-height analysis for that case is shown in Fig. 2.

*This work was performed under the auspices of the U.S. Atomic Energy Commission.

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