Determination of the $^{237}Np(n,2n)^{236}Np$ cross section at 15 MeV neutron energy

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The neutron cross section for the $^{237}Np(n,2n)^{236}Np^{g}$ reaction has been determined at a neutron energy of 15 MeV. Two independent methods were used: (1) α particles of ^{236}Pu following the β' decay of ^{236}Np were counted after chemical separation of Pu. (2) The 642.4 keV γ rays of $^{236}Np^{g}$ decay to ^{236}U were measured directly after the irradiation of the ^{237}Np sample. The neutron fluence was calculated from the $^{27}Al(n,\alpha)^{24}Na$ reaction and from the measurement of the fission product ^{97}Zr with a calibrated Ge(Li) spectrometer. The most precise value is 247 ± 22 mb. The error is mainly due to uncertainties of the nuclear data used for the cross-section calculation.

[NUCLEAR REACTIONS $^{237}Np(n,2n)$, $E=15$ MeV, measured σ.]

I. INTRODUCTION

The isotope ^{238}Pu, which is finding increasing practical applications, is mainly produced by the reaction $^{237}Np(n,\gamma)^{238}Np \xrightarrow{\beta'} {}^{238}Pu$. At neutron energies exceeding 6.8 MeV the following reaction becomes important: $^{237}Np(n,2n)^{236}Np \xrightarrow[50\%]{\beta'} {}^{236}Pu$. The cross section of this reaction is of great interest because of two reasons: (1) The penetrating γ rays emitted by the decay products of ^{236}Pu, especially ^{212}Bi and ^{208}Tl, are limiting the use of ^{238}Pu for biomedical and aerospace applications as an energy source.[1] (2) The ^{236}Pu content of plutonium fuel for fast breeder reactor systems is also important for the calculation of the neutron source data needed for shield design at fabrication plants.[2]

As there are discrepancies between the measured[3,4] and also between calculated[5,6] values, a program was started to measure the excitation function of the $^{237}Np(n,2n)$ reaction. First, the cross section at 15 MeV average neutron energy was redetermined. Because the 1.29×10^{6} y isomer of ^{236}Np does not contribute significantly to the buildup of ^{236}Pu, only the part of the reaction leading to the 22 h isomer of ^{236}Np is of interest.

Principally, three ways to determine the cross section have been taken into account, which are pointed out in the simplified reaction and decay scheme (Fig. 1): (a) the measurement of prompt γ rays accompanying the neutron emission, (b) the direct measurement of γ rays following the electron capture (EC) of ^{236}Np to ^{236}U, and (c) the measurement of α particles of the ^{236}Pu decay after chemical separation of plutonium.

Method (a) could not be performed successfully as the high background radiation disturbs the detection of prompt γ rays from the $(n,2n)$ reaction.[7] The two other methods are described and discussed in this paper.

II. EXPERIMENTAL

^{237}Np as received (Gesellschaft für Kernforschung, Karlsruhe) contained some plutonium isotopes and ^{241}Am which had to be removed before the neutron irradiation. A two-step anion exchange process was developed to purify the neptunium from these impurities as well as from the main content of ^{233}Pa which disturbs the neutron fluence measurement by γ counting of the fission product ^{97}Zr due to high γ background. The ^{236}Pu present in the purified Np sample was calculated from the α spectra to be less than 1% of the amount of ^{236}Pu after the irradiation. The details of the purification procedure will be published elsewhere.

The fast neutrons [15 MeV full width at half-maximum (FWHM): 1 MeV] were produced by bombarding a tritium target with deuterons, acceler-

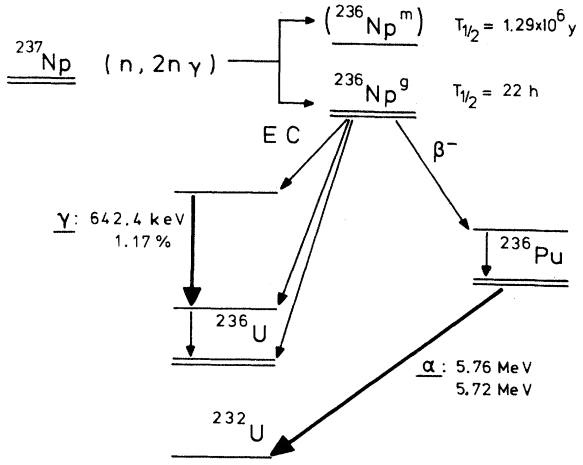

FIG. 1. Simplified reaction and decay scheme of the $^{237}Np(n,2n)$ reaction (measured transitions are indicated by heavy arrows).

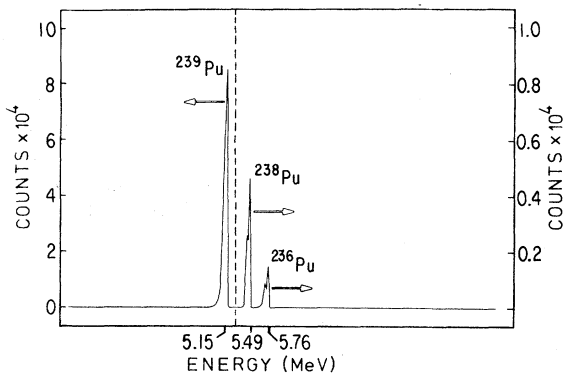

FIG. 2. α-particle energy spectrum of separated plutonium.

ated with the type K 3000 Van de Graaf generator, operated by the Gesellschaft für Strahlen- und Umweltforschung, Neuherberg. The ^{237}Np was irradiated as NpO_2 sealed in a polyethylene capsule surrounded by Cd to prevent (n,γ) and (n,f) reactions by thermalized neutrons. The sample was exposed to the neutron flux for 942 min. Four Al plates located before and behind the NpO_2 and the ^{237}Np(n,f) reaction were used as neutron fluence detectors. In addition, the relative flux of the neutrons was measured by two independent monitors and recorded. A calibrated Ge(Li)-detector system coupled to a 4096 channel pulse-height analyzer was used to measure the γ rays from ^{24}Na, the product of the ^{27}Al$(n,\alpha)^{24}$Na reaction.

After a cooling time of several days to allow the ^{236}Np^g to decay, plutonium was isolated after adding ^{239}Pu as a carrier by the same chemical procedure used for the purification of the Np sample. The ^{236}Pu content of the ^{239}Pu carrier did not exceed 0.03% of the ^{236}Pu produced by the $(n,2n)$ reaction. An aliquot of the Pu fraction was taken and evaporated on a platinum disk for α counting with a Si surface barrier detector (Fig.2).

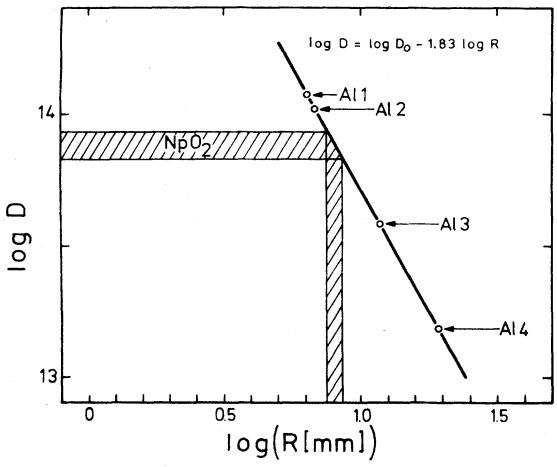

FIG. 3. Neutron fluence (D) versus distance (R) of Al plates and NpO_2 from the ^{3}H target.

III. RESULTS

A. Fluence determination

The neutron fluence (D) was calculated in the usual way with a slight modification. As the neutron flux is changing during the long irradiation time the activation of the Al plates has been calculated separately for small time intervals with a constant neutron flux and summarized. For this purpose the relative flux measurement was performed.

D was calculated for every single Al plate. By drawing the neutron fluence as a function of the distances of the plates from the ^{3}H target on a log/log scale, a straight line with a slope of -1.83 is obtained (Fig. 3). From this curve the average neutron fluence through the Np was evaluated using the equation

$$\log D(R) = \log D_0 - 1.83 \log R,$$

where D_0 represents the fluence at 1 mm distance. The obtained neutron fluence is

$$D = (6.77 \pm 0.24) \times 10^{13} \text{ neutrons cm}^{-2}.$$

The irradiated sample was measured during the

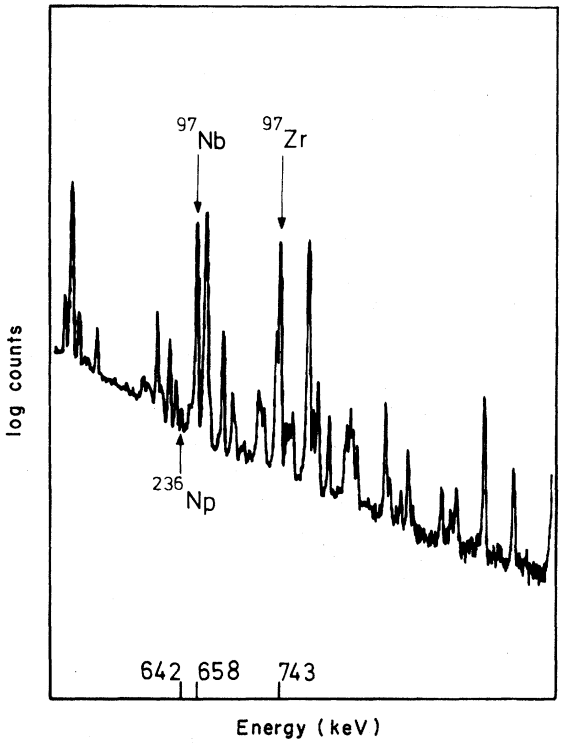

FIG. 4. The γ energy spectrum of the irradiated neptunium sample.

storage time—necessary to allow the decay of the $^{236}Np^{g}$—with a calibrated γ spectrometer, and the spectra were recorded. From the very complex γ spectra (Fig. 4) $^{236}Np^{g}$ and ^{97}Zr as well as ^{97}Nb could be identified by γ energy and half-life determination. The fission yield of ^{97}Zr has been taken from this decay curve, obtained by a least square fit using the observed activity of ^{97}Zr at several times. Corrections have been made for the dead time of the spectrometer, spectrometer efficiency, self-absorption in the sample, abundance of the measured γ transition, and the decay during the counting time (about 6 h) before drawing the curves. The above-mentioned changing of the neutron flux has also been taken into account for the fluence calculation. Nuclear data were taken from Ref. 8. The fluence determination by measuring a fission product directly has the great advantage of being independent of any geometrical arrangement and chemical yield determinations. Its disadvantage is the uncertainty of the nuclear data for the ^{237}Np fission with fast neutrons.[9] From this calculation the obtained neutron fluence is

$$D = (6.40 \pm 0.73) \times 10^{13} \text{ neutrons cm}^{-2}.$$

The agreement of both values is quite good. Uncertainties arise mainly by the efficiency determination of the Ge(Li) spectrometer due to high counting rates and dead time of the electronic equipment, and the peak analysis of the complicated fission spectra.

B. Calculation of the cross section

The number N of ^{236}Pu atoms after the β decay of ^{236}Np is

$$N_{^{236}Pu} = 0.49 N_{^{237}Np} \sigma_{(n,2n)} D, \tag{1}$$

and $\sigma_{(n,2n)}$ is given by

$$\sigma_{(n,2n)} = \frac{1}{0.49D} \frac{N_{^{236}Pu}}{N_{^{237}Np}}. \tag{2}$$

The factor 0.49 was derived from Gindler's value[10] of the branching ratio $EC/\beta^- = 1.05$ for the ^{236}Np decay. The ratio $N_{^{236}Pu}/N_{^{237}Np}$ cannot be measured directly. For this reason the ratio $N_{^{239}Pu}/N_{^{237}Np}$ has been determined from the α spectrum of the ^{237}Np sample after adding the ^{239}Pu carrier. From the α spectrum of the Pu fraction after the chemical separation, the ratio $N_{^{239}Pu}/N_{^{236}Pu}$ is obtained. The ratio $N_{^{236}Pu}/N_{^{237}Np}$ can be replaced by

$$\left[\frac{N_{^{236}Pu}}{N_{^{239}Pu}}\right]_{Pu} \left[\frac{N_{^{239}Pu}}{N_{^{237}Np}}\right]_{Np}.$$

The indices Pu and Np at the brackets indicate the Pu spectrum and the Np spectrum, respectively. With this derivation Eq. (2) becomes

$$\sigma_{(n,2n)} = \frac{1}{0.49D} \left[\frac{N_{^{236}Pu}}{N_{^{239}Pu}}\right]_{Pu} \left[\frac{N_{^{239}Pu}}{N_{^{237}Np}}\right]_{Np}. \tag{3}$$

By this kind of calculation no absolute determination of either the ^{236}Pu content or the ^{239}Pu carrier is necessary which could introduce another uncertainty. Only the relative α counts are needed to calculate the cross section. N is related to the α counts S by Eq. (4):

$$N = \frac{A}{\lambda} = \frac{AT_{1/2}}{\ln 2} = \frac{ST_{1/2}}{\ln 2 (g)(t_c)}, \tag{4}$$

where

A = activity,

$\lambda = \ln 2 / T_{1/2}$ disintegration constant,

$T_{1/2}$ = half-life,

g = efficiency (mainly geometry factor),

t_c = counting time;

g and t_c are the same for couples ^{237}Np-^{239}Pu and ^{239}Pu-^{236}Pu, respectively.

The combination of Eqs. (3) and (4) leads to Eq. 5:

$$\sigma_{(n,2n)} = \frac{1}{0.49D} \left[\frac{S_{^{236}Pu}}{S_{^{239}Pu}}\right]_{Pu} \left[\frac{S_{^{239}Pu}}{S_{^{237}Np}}\right]_{Np} \frac{[T_{1/2}]_{^{236}Pu}}{[T_{1/2}]_{^{237}Np}}. \tag{5}$$

Due to the half-lives of the nuclides only a correction for the decay of ^{236}Pu during cooling (110 day) was made. Taking D from the Al detectors $\sigma_{(n,2n)}$ is

$$\sigma = 247 \pm 22 \text{ mb}.$$

With D from fission measurements the value is

$$\sigma = 261 \pm 38 \text{ mb}.$$

The same calculation can be made with the ^{236}Np activity determined by γ measurement. The γ transition with the energy of 642.4 keV has an abundance of only 1.17%, introducing a great uncertainty into the calculation of σ. For this reason the values can only be taken to support the values found by α measurements. The values are 285 mb and 300 mb with the neutron fluence calculated from the Al detectors and ^{97}Zr fission yield, respectively.

IV. DISCUSSION

As mentioned earlier the cross section reported in this paper does not include the part of the $(n, 2n)$ reaction leading to the long-lived isomer of ^{236}Np. Landrum, Nagle, and Lindner[4] found by intense thermonuclear neutron flux experiments a value for the ratio of the long-lived ^{236}Np to ^{236}Pu of

0.76 ± 0.03. Adapting this value the 247 mb have to be multiplied by the factor $[1 + 0.76(0.49)]$ to get an over-all $(n, 2n)$ cross section of 339 mb.

The error of our result is mainly determined by the uncertainty included in the EC/β^- branching ratio of 1.05 ± 0.17 reported by Gindler and Sjoblom.[10] The statistical errors of α counting and ^{24}Na measurements did not exceed 1.4%. So the over-all uncertainty $F = (\sum f_i^2)^{1/2}$ is 9.1% for the reported 247 mb. This value is preferred to the other calculations because errors introduced by the uncertainty of the nuclear data for the fission of ^{237}Np [$\sigma_f = 2.5 \pm 0.07$ b; ^{97}Zr yield $= 5.43 \pm 0.49\%$, $E_n = 14.5$ MeV(Ref. 9)], and the very small abundance of the ^{236}Np γ transition, which can be inaccurate by as much as 30%(Ref. 8), are much higher than those which are associated with the $\sigma(n,\alpha)$ value reported for ^{27}Al by Vonach *et al.*[11] and possible uncertainties in the position of the neptunium sample during irradiation. The value of 247 mb is in good agreement with the cross section leading to $^{236}Np^g$ reported in Ref. 4 as 0.27 b for 14.95 MeV neutrons, if one considers the possibility of systematical errors of these cross section measurements. We believe our method is good enough to warrant its use in the measurement of $\sigma_{(n,2n)}$ at other neutron energies.

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