Studies on Plutonium, Neptunium, and Uranium Produced in the $^{241}\text{Pu} + ^{136}\text{Xe}$ Reaction

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(Received June 15, 1981)

$^{241}\text{Pu} + ^{136}\text{Xe}$ 반응에서 생성된 Pu, Np 및 U에 관한 연구

제 목
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(1981. 6. 15 작성)

Abstract

Plutonium, neptunium, and uranium isotopes which were produced in the interaction of $^{136}\text{Xe}$ and $^{241}\text{Pu}$ are separated and determined their cross sections. The cross sections are: $\sigma^{(241}\text{Pu}) = 66 \text{ mb}, \sigma^{(244}\text{Pu}) = 57 \text{ mb}, \sigma^{(246}\text{Pu}) = 57 \text{ mb}, \sigma^{(239}\text{Np}) = 15 \text{ mb}, \sigma^{(240}\text{U}) = 12 \text{ mb}, \sigma^{(244}\text{U}) = 6.4 \text{ mb}$ respectively.

요 약

Pu-241의 Xe-136반응에서 생성된 Pu, Np 및 U를 분리하여 그 핵반응 단변진을 측정하였다. 핵 반응단변진은 각각 $\sigma^{(241}\text{Pu}) = 66 \text{ mb}, \sigma^{(244}\text{Pu}) = 57 \text{ mb}, \sigma^{(246}\text{Pu}) = 57 \text{ mb}, \sigma^{(239}\text{Np}) = 15 \text{ mb}, \sigma^{(240}\text{U}) = 12 \text{ mb}, \sigma^{(244}\text{U}) = 6.4 \text{ mb}$였다.

As part of the continuing study of deep inelastic transfer reaction$^{(1-9)}$ in the actinide region, we have examined the interaction of $^{136}\text{Xe}$ with the heaviest naturally occurring isotope $^{244}\text{Pu}$.

Plutonium, neptunium, and uranium isotopes which were produced in bombardment$^{10}$ of $^{241}\text{Pu}$ with $^{136}\text{Xe}$ were separated using an anion exchange chromatographic technique$^{11}$ and isotopes of Pu, Np, and U were identified by analyzing the beta-decay curve of the activities of each element. The activities of each element were determined with a conventional proportional counting system.

Experimental Procedure

(a) Materials and Reagents

Resin, AG 1×80 anion exchange resin (400 mesh), available from Bio Rad Laboratories, Richmond, California. Before the resin was used, it was conditioned with acid and alkali and washed with water.

Apparatus, A section of pyrex tubing 0.3cm ID and 5cm length was used to
prepare the column. The tubing was pulled out to a tip at one end and glass wool was inserted to retain the resin. Additional apparatus are hot plate, heat lamp, platinum disc (one-inch diameter), microoipettes.

Column, Resin bed: 0.09cm²×4.0cm, column volume 0.36ml, flow rate 0.1 drop/40sec. adjusted by hydrostatic pressure, temperature 25°C, effluent volumes (column volumes=c.v.)

Non-absorbed fraction: 5 c.v.(1.8ml) of solution II
Pu fraction: 5 c.v.(1.1ml) of solution II
No fraction: 3 c.v.(1.1ml) of solution V
U fraction: 3 c.v.(1.1ml) of solution V
Solutions (I): 12 M HCl-0.05 M HNO₃
(II): 12 M HCl-0.15 M H₂SO₄ saturated with HCl gas
(V): 4 M HCl-0.1 M HF
(γ): 0.1 M HCl-1 M HF

(b) Sample Preparation
Recoil products of ²³⁹Pu⁺⁻¹³⁵Xe reaction were collected on the aluminum catch foils and dissolved with 10M sodium hydroxide, then removed rare earths and transition elements with lanthanide fluoride precipitation method. Meanwhile 100 lambda of Pu-238(8.34 μ Cl), Np-237(0.924 μ Cl) and U-233(4.44 μ Cl) was added to the dissolved solution to determine the chemical yield of Pu, Np, and U in the catcher foil.

(c) Column Operation
Resin as a slurry in water is added to the column until a resin bed about 4cm in length is formed: it is then pretreated with 5 c.v. (2 ml) of solution 1 (12 M HCl+1dr HNO₃) and the sample solution (actinide+ fission products in 12 M HCl) is added. Flow rate is controlled by hydrostatic pressure to about 1 drop per 30 sec. After the sample has passed into the resin bed, 0.5 c.v. of 12 M HCl is added as wash, taking care not to disturb the resin at the top of the bed. When the wash solution has passed into the bed an additional 4.5 c.v.(1.5 ml) of eluent are added and elution continued. The effluent is discarded; it contains the non-absorbed elements. Then plutonium is eluted by passing 5 c.v.(2.1 ml) of 12 M HCl-0.15M H₂SO₄ saturation saturated with gas through the resin. The effluent is collected on a one-inch diameter platinum plate while heating with lamp. In this process a drop of the effluent is dried before next drop comes off.

Neptunium is removed with 3 c.v.(1.1 ml) of 4 M HCl-0.1 M HF. Uranium is eluted with 0.5 M HCl-1 M HF. The effluents are collected the same as for Pu. Column operation time for the separation of Pu, Np, U, and non adsorbed elements into individual fraction is about 60 min.

(d) Counting Procedure
(i) Beta proportional counter (external sample, gas flow type 10% methane-90% argon used at voltage setting of 1900 v for beta counting) and alpha proportional counter (internal sample gas flow type 7% methane-93% argon) were used to measure beta and alpha activities, respectively. To eliminate alpha activity which might remain in the sample, 5.0mg/cm aluminium absorbers were inserted between the sample and counter in case of beta counting.

(ii) Alpha Pulse Analysis
Alpha pulse analysis of Pu sample was carried out using gold-plated surface barrier detector obtained from Ortec, Inc. The detector had an average area of 300mm², an energy resolution (FWHM) of 20 kev, a sensitive thickness of ~100 microns.
and operated at a voltage of 100 volts. The detector was calibrated using \(^{238}\text{Pu}\) standard. To obtain a good resolution of alpha pulse analysis, the Pu sample was dissolved in 12 M HCl and the solution was placed on an anion exchange column, then eluted with 2 M HCl and collected on a platinum disc.

Fig. 1. Decay Curve of Pu Fraction (a)

Fig. 2. Decay Curve of Pu Fraction (b)

Fig. 3. Decay Curve of Np Fraction

Fig. 4. Decay Curve of U Fraction
Results

(a) Decay Curves of Pu, Np and U

Figures 1, and 2 show the decay curves of the Pu fraction. From the observed decay curve, we are able to identify $^{241}\text{Pu}$-$^{241}\text{Am}(t_{1/2}=10.5 \text{ hr}), \quad ^{240}\text{Pu}(t_{1/2}=5.0 \text{ hr})$ and $^{244}\text{Pu}(t_{1/2}=10.85 \text{ d})$ isotopes. Figure 3 shows the decay curve of the Np fraction. The decay curve shows $^{239}\text{Np}(2.35 \text{ d})$ activity and is contaminated by the strong $^{244}\text{Pu}(10 \text{ 5 hr})$ activity in the Np. The contamination from Pu in the Np fraction is calculated to be about 5% of the total Pu activity. Figure shows the decay curves of the U fraction. From the decay curves, $^{234}\text{U}(14.1 \text{ hr})$ and $^{235}\text{U}(6.75 \text{ d})$ isotopes can be identified.

(b) Determination of Chemical Yields of Pu, Np, and U during Chromatographic Ion Exchange Separation.

To determine chemical yields of Pu, Np, and U, alpha activities of standard Pu-238 (8.34 $\mu$ Ci), Np-237 (9.24 $\mu$Ci), and U-233 (4.44 $\mu$ Ci) solutions which are introduced before the chemical separtion procedure, one compared with the alpha activities of Pu, Np, and U chemical yield of U, Pu, and Np separations.

(c) Cross Section Calculations

The cross section of each isotope was calculated from the following equation:

$$\sigma = \frac{m}{N(1-e^{-\frac{m}{c}})} e^{-\frac{m}{c}} \gamma_e \gamma_b$$

where

$m =$ Counting rate (counts/min) after cooling

$I =$ Intensity of Xe ion per min

$N =$ Initial counting rate (counts/min) of sample

Table 1: Chemical Yield of Pu, Np, and U Separation

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Pu-238 (cpm)</th>
<th>Np-237 (cpm)</th>
<th>U-233 (cpm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Standard Activity</td>
<td>2085</td>
<td>231</td>
<td>1169</td>
</tr>
<tr>
<td>Sample Activity</td>
<td>695*</td>
<td>31.2</td>
<td>—</td>
</tr>
<tr>
<td>Yield</td>
<td>33%</td>
<td>13.5%</td>
<td>13.5%**</td>
</tr>
</tbody>
</table>

* The correction is made by the activity of Pu-240 which was transferred from the target to the catcher foil. The value corresponds to 10% of sample activity. Alpha pulse analysis of Pu-240 will be described in section (d).

** Because the porous teflon plug to retain resin was not available, We used glass wool instead of the teflon plug, and in U fraction separation the glass wool was dissolved in 0.5 M HCl-1 M HF solution. Because of the thick U sample, alpha counting of the U sample was impossible. So the yield was estimated as same as Np fraction.

Table 2: Cross Section of Pu, Np, and Uranium Isotopes

<table>
<thead>
<tr>
<th>Isotopes</th>
<th>$t_{1/2}$</th>
<th>$t_e$</th>
<th>$m$</th>
<th>c</th>
<th>r_e</th>
<th>$\sigma$ (mb)</th>
<th>ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}\text{Pu}$</td>
<td>10.5 hr</td>
<td>1.5 hr</td>
<td>$4.0 \times 10$ cpmp</td>
<td>33%</td>
<td>2.24*</td>
<td>66</td>
<td>Fig. 1, 2</td>
</tr>
<tr>
<td>$^{240}\text{Pu}$</td>
<td>5.0 hr</td>
<td>1.5 hr</td>
<td>$2.5 \times 10$ cpmp</td>
<td>33%</td>
<td>1</td>
<td>57</td>
<td>Fig. 1, 2</td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>10.85 d</td>
<td>45.8 hr</td>
<td>$7.0 \times 10$ cpmp</td>
<td>33%</td>
<td>2*</td>
<td>6.0</td>
<td>Fig. 2</td>
</tr>
<tr>
<td>$^{239}\text{Np}$</td>
<td>2.35 d</td>
<td>12.7 hr</td>
<td>$2.0 \times 10$ cpmp</td>
<td>13.5%</td>
<td>1</td>
<td>15</td>
<td>Fig. 3</td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>14.1 hr</td>
<td>2.17 hr</td>
<td>$1.2 \times 10$ cpmp</td>
<td>13.5%</td>
<td>2*</td>
<td>12</td>
<td>Fig. 4</td>
</tr>
<tr>
<td>$^{237}\text{U}$</td>
<td>6.75 d</td>
<td>2.17 hr</td>
<td>$3.4 \times 10$ cpmp</td>
<td>13.5%</td>
<td>1</td>
<td>6.4</td>
<td>Fig. 4</td>
</tr>
</tbody>
</table>

$\gamma_e = 16.6\%$

* $^{241}\text{Pu}$-$^{241}\text{Am}$ mixture

$\gamma_b = 16.6\%$

* $^{241}\text{Pu}$-$^{241}\text{Am}$ mixture

$\gamma_b = 16.6\%$

* $^{241}\text{Pu}$-$^{241}\text{Am}$ mixture
t_b = Bombardment time of 243Pu target
t_c = Chemical separation and cooling time
t_d = Decay constant of each isotope
c = Counting efficiency
\gamma = Chemical yield of each element
\gamma_v = Ratio of total activity to given isotope activity

The result of cross section of 243Pu+Xe reaction products are shown in Table 2.

(d) Estimate of the Transfer Ratio of 244Pu from the Source to the Catcher Foil.

Alpha pulse analysis of the refined Pu sample was done with the surface barrier detector. The count was started 15 days after the end of bombardment. Two alpha peaks were observed at the 349 and 285 channels and the peaks were identified as 243Pu and 244Pu. These peaks were hand integrated and corrected for the alpha tail.

<table>
<thead>
<tr>
<th>Peak Channel Energy Mev</th>
<th>Peak Area dpm</th>
</tr>
</thead>
<tbody>
<tr>
<td>349</td>
<td>5.4992</td>
</tr>
<tr>
<td>285</td>
<td>5.1683</td>
</tr>
</tbody>
</table>

\[ Targe\ weight^{100} \text{ of } 244Pu \text{ are } 215 \ \mu g. \text{ Alpha dpm of } 244Pu \text{ per } \mu g \text{ target} = 1.54 \times 10^7 \text{ dpm.} \]

The transfer ratio of 244Pu from the source to the catcher foil is given as follows:

\[ \text{Transfer ratio} = \frac{158 \text{ (dpm)}}{215 \text{ (}\mu g\text{)}} \times 1.54 \times 10^7 \text{ (dpm/}\mu g\text{)} \]

\[ = 0.048 (\%) \]

Discussion

Overall, six isotopes of U, Np, and Pu were analyzed by beta-decay curve of the activities of each element. It should be noted that no correction has been made for the unknown efficiency of collection by the recoil catcher arrangement since neither the angular distribution nor the energy spectrum of the products of these deep inelastic transfer reaction is known.

This work suggests then that either the isotopic yield may be narrower than previously measured or may be shifted somewhat with respect to the valley of stability. This effect, while not wholly surprising is more evident in analysis of products from this neutron rich target than from the bulk of heavy ion studies using 235U as a target.

Some support for this conclusion may come from the cross section for U isotopes which derive from relatively small mass and charge transfer. These are both subject to all of the uncertainty inherent in beta decay analysis. For example, it is not clear whether the presence of activities aying half-lives not corresponding to known isotopes may invalidate data for these activities which apparently correspond to known isotopes. Additionally, considerable uncertainties arise in the evaluation of absolute disintegration rates from observed count rates due to efficiency and yield correction. However, subject to these errors, data from U isotopes do support the idea of relatively narrow mass distribution lying near “Stability”. Again, it is likely this is an effect arising from the bombardment of the neutron excessive 244Pu target.

Acknowledgement

The work has been done at Lawrence Berkeley Laboratory, University of California, when the author visited there as a guest researcher in 1978-1980. The author wishes to express his gratitude to Dr. G. T. Seaborg and Dr. D. C. Hoffman for their kind assistance and advice.

Appendix^{100}

The target 215 \mu g of 244Pu electrosprayed
by P. A. Baisden onto a 12.5 \( \mu \)m Be foil. Mass analysis performed at Oak Ridge National Laboratory showed it to contain 98.6\% \( {\text{241}}_{\text{Pu}} \), 1\% \( {\text{242}}_{\text{Pu}} \), and small amounts of \( {\text{239}}_{\text{Pu}} - {\text{241}}_{\text{Pu}} \). The diameter was 6.3mm yielding an average target thickness of 672 \( \mu \)g/cm\(^2\).

The irradiation made use of a new facility at the Super HILAC, the zero degree beam line designed and implemented by one of the authors, and fabricated at LBL. The beam emerges at zero degrees relative to the axis of the accelerating tanks of the Super HILAC; because there is no bending involved, the intensity of the beam incident upon the target is maximized. Two independent cooling systems are used. The target and the beam window are cooled by jets of nitrogen gas at low pressure and high velocity, and the collimators and beam stop are cooled by circulating water.

In this experiment, recoils were caught in a cone of 51 \( \mu \)m Al which subtends a solid angle of 89\% of the forward hemisphere while allowing the unscattered beam to emerge without striking the Al. This arrangement was necessary due to the large scale production of lanthanides from the interactions of Xe and Al. The target assembly is also designed for rapid access to and dismounting of catcher foils. The entire actinide target assembly may be isolated from the accelerator by means of a slapper valve activated either by absolute pressure or by the rate of increase of pressure in the beam line. Further protection arises from a large volume scattering chamber located between the accelerator and target assembly. This target system was produced specifically for the irradiation of high actinides or other highly radioactive targets.

The \( {\text{244}}_{\text{Pu}} \) target was irradiated with a beam of approximately 1 \( \mu \)amp \( {\text{133}}_{\text{Xe}}^{3+} \) for 4.42hr. This averages to 2.28\times10\(^{14}\)Xe/sec. After degradation of the incident 1156 MeV Xe beam in the 1.8mg/cm\(^2\) Havar window and the 12.5 \( \mu \)m Be backing the average incident beam energy was calculated to be about 965 MeV. The energy loss in the target was calculated to be about 20 MeV.

References