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《**Original**》

Studies on Plutonium, Neptunium, and Uranium Produced in the $^{244}Pu+^{136}Xe$ Reaction

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$^{244}Pu+^{136}Xe$ 반응에서 생성된 Pu, Np 및 U에 관한 연구

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Abstract

Plutionum, neptunium, and uranium isotopes which were produced in the interaction of ^{136}Xe and^{244}Pu are separated and determined their cross sections. The cross sections are: $\sigma(^{245}Pu)=66$ mb, $\sigma(^{243}Pu)=57$ mb, $\sigma(^{246}Pu)=6.0$ mb, $\sigma(^{239}Np)=15$ mb, $\sigma(^{240}U)=12$ mb, $\sigma(^{245}U)=6.4$ mb respectively.

요 약

Pu-244와 Xe-136반응에서 생성된 Pu, Np 및 U을 분리하여 그 핵반응 단면적을 측정하였다. 핵반응단면적은 각각 $\sigma(^{245}Pu)=66$ mb, $\sigma(^{243}Pu)=57$ mb, $\sigma(^{246}Pu)=6.0$ mb, $\sigma(^{239}Np)=15$ mb, $\sigma(^{240}U)=12$ mb, $\sigma(^{237}U)=6.4$ mb이었다.

As part of the continuing study of deep inelastic transfer reaction[1-9] in the actinide region, we have examined the interaction of ^{136}Xe with the heaviest naturally occuring isotope.^{244}Pu.

Plutonium, neptunium, and uranium isotopes which were produced in bombardment[10] of ^{244}Pu with ^{136}Xe were separated using an anion exchange chromatographic technigue[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 n 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: 0.09cm^2×4.0cm, column volume 0.36ml, flow rate ca one 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: 6 c.v.(1.1ml) of solution Ⅲ

No fraction: 3 c.v.(1.1ml) of solution Ⅳ

U fraction: 3 c.v.(1.1ml) of solution Ⅴ

Solutions (Ⅰ): 12 M HCl-0.05 M HNO: (Ⅱ): 12 M HCl: (Ⅲ): 12M HCl-0.15 M HI saturated with HCl gas:(Ⅳ): 4 M HCl-0.1 M HF: (Ⅴ): 0.1 M HCl-1 M HF

(b) Sample Preparation

Recoil products of $^{224}Pu+^{136}Xe$ reaction were collected on the aluminum catcher foil and dissolved with 10M sodium hydroxide. then removed rare earths and transition elements with lanthanide fluoride preclpitation method[10]. Meanwhile 100 lambda of Pu-238(8.34 μ Ci). Np-237(0.924 μ Ci) and U-233(4.44 μ Ci) 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 untll 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 HNO_3) 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 ses. After the sample has passed into the resin bed, 0.5 c.v. of 12 M HCl is added bs 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 12M HCl-0.15M HI soultion 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% a rgon) were used to measure bata and alpha activities, respectively. To eliminate alpha activity which might remain in the sample, 5.0mg/cm aluminum 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 300mm^3, 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}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.

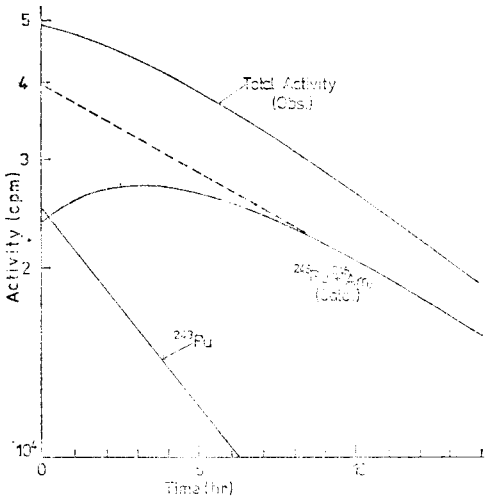

Fig. 1. Decay Curve of Pu Fraction (a)

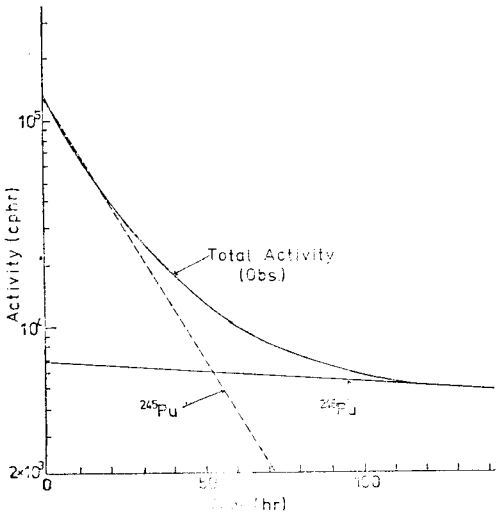

Fig. 2. Decay Curve of Pu Fraction (b)

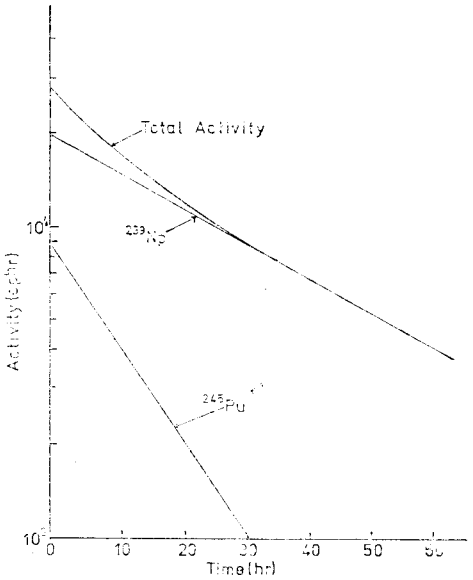

Fig. 3. Decay Curve of Np Fraction

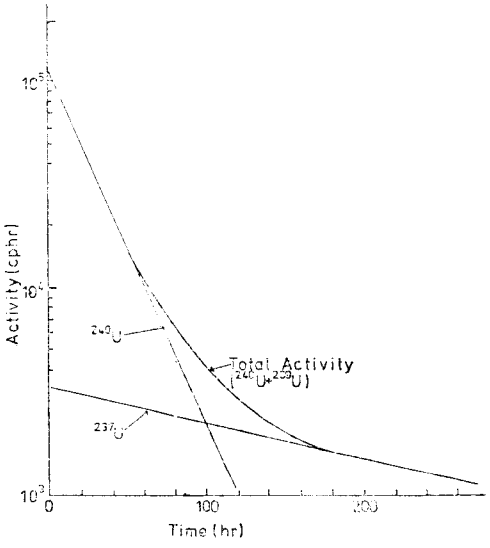

Fig. 4. Decay Curve of U Fraction

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 cuve, we are able to identify ^{245}Pu-^{245}Am ($t_{1/2}$=10.5 hr), ^{243}Pu ($t_{1/2}$=5.0 hr) and ^{246}Pu ($t_{1/2}$=10.85 d) isotopes. Figure 3 shows the decay curve of the Np fraction. The decay curve shows ^{239}Np (2.35 d) activity and is contaminated by the strong ^{245}Pu (10.5 hr) activity in the Np. The contamination from Pu in the Np fractionis calculated to be about 5% of the total Puactivity. Figure shows the decay curves of the U fraction. From the decay curves, ^{240}U (14.1hr) and ^{237}U (6.75 d) Isotopes can be identified.

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

Chemical Yield of Pu, Np, and U Separation			
Isotope	Pu-238	Np-237	U-233
Standard Activity	2085 cpm	231 cpm	1109 cpm
Sample Activity	695 cpm*	31.2 cpm	—
Yield	33%	13.5%	13.5%**

* The correction is made by the activity of Pu-240 which was transfered 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.

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

To determine chemical yields of Pu, Np, and U. alpha activities of standard Pu-238 (8.34 μ Ci), Np-237 (0.924 μCi), and U-233 (4.44 μ 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 Calculatons

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

$$\sigma=\frac{m}{I\cdot N(1-e^{-\lambda t_b})\ e^{-\lambda t_c}\cdot c\cdot \gamma_c\cdot \gamma_b}$$

where

m=Counting rate (counts/min) after cooling

I=Intensity of Xe ion per min

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

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

Isotopes	$t_{1/2}$	$^{+}t_c$	m	c	r_b	σ (mb)	ref.
^{245}Pu	10.5 hr	1.5 hr	4.0×10 cpm	33%	2.24[a]	66	Fig. 1, 2
^{243}Pu	5.0 hr	1.5 hr	2.5×10 cpm	33%	1	57	Fig. 1, 2
^{246}Pu	10.85 d	45.8 hr	7.0×10 cphr	33%	2[b]	6.0	Fig. 2
^{239}Np	2.35 d	12.7 hr	2.0×10 cphr	13.5%	1	15	Fig. 3
^{240}U	14.1 hr	2.17 hr	1.2×10 cphr	13.5%	2[c]	12	Fig. 4
^{237}U	6.75 d	2.17 hr	3.4×10 cphr	13.5%	1	6.4	Fig. 4

Y_e=16.6%

a ^{245}Pu-^{245}Am mixture

b ^{246}Pu-^{246}Am mixture

c ^{240}U-^{240}Np mixture

t_b=Bombardment time of ^{244}Pu target
t_c=Chemical separation and cooling time
λ=Decay constant of each isotope
c=Counting efficiency
γ_c=Chemical yield of each element
γ_b=Ratio of total activity to qiven isotope activity

The result of cross section of ^{244}Pu+Xe reaction products are shown in Table 2.

(d) Estimate of the Transfer Ratio of ^{244}Pu 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 affer the end of bombardment. Two alpha peaks were observed at the 349 and 285 channels and the peaks were identified as ^{239}Pu and ^{240}Pu. These peaks were hand integrated and corrected for the alpha tail.

Peak Channel	Energy Mev	Peak Area	dpm
349	5.4992	3406	4170
285	5.1683	129	158

Targe weight[10] of ^{244}Pu are 215 μg. Alpha dpm of ^{240}Pu per μg target=1.54×10^3 dpm. The transfer ratio of ^{244}Pu from the source to the catcher foil is given as follows:

$$\text{Transfer ratio}=\frac{158\,(\text{dpm})}{215\,(\mu\text{g})\times1.54\times10^3\,(\text{dpm}/\mu\text{g})}$$
$$=0.048(\%)$$

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 analvsis of products from this neutron rich target than from the bulk of heavy ion studies using ^{238}U 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 subiect to all of the uncertainity inherent in beta decay analysis. For example, it is not clear whether the presence of activities aving half-lives not corresponding to known isotopes may invalidate data for these activities which apparently correspond to known isotopes. Additionlly, considerable uncertainities arise in the evaluation of absolute disintegration rates from observed count rates due to efficiency and yield correction. However, subject to thes eerrors. data from U isotopes do support the idea of relatively narrow mass distribution lying near "Stability". Again, it is likely thisis an effect arising from the bombardment of the neutron excessive ^{244}Pu target.

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Appendix[10]

The target 215 μg of ^{244}Pu electrosprayed

by P. A. Baisden onto a 12.5 μm Be foil. Mass analysis performed at Oak Ridge National Laboratory showed it to contain 98.6% ^{244}Pu, ~1% ^{242}Pu, and small amounts of $^{238-241}$Pu. The diameter was 6.3mm yielding an average target thickness of 672 μg/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 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 caugh in a cone of 51 μm 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 slammer 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 ^{244}Pu target was irradiated with a beam of approximately 1 μamp ^{136}Xe^{29+} for 4.42hr. This averages to 2.28×10^{11}Xe/sec. After degradation of the incident 1156 MeV Xe beam in the 1.8mg/cm^2 Havar window and the 12.5 μm Be backing the average incident beam energy was calculaed to be about 965 MeV. The energy loss in the target was calculated to be about 20 MeV.

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