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PRODUCT YIELDS FOR THE PHOTOFISSION OF ^{238}U, ^{237}Np AND ^{239}Pu

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Abstract : This paper shows the absolute yields for the mass distribution of photofission products of ^{238}U, ^{237}Np and ^{239}Pu. The thin targets of ^{238}U, ^{237}Np and ^{239}Pu were irradiated with the bremsstrahlung beam whose end-point energy was 20-, 30- and 60-MeV. Fission fragments recoiled out from the target were catched on the aluminum foil. The mass-yield distributions of fission fragments were obtained by measuring gamma-ray activities with a pure-Ge detector. Our mass distribution curves showed good agreement with the other experimental results.

(photofission, cross section, bremsstrahlung, mass distribution, ^{238}U, ^{237}Np, ^{239}Pu)

Introduction

Photofission mass distribution of ^{238}U, ^{237}Np and ^{239}Pu have already been reported by different authors. But these studies have discussed on the relative yield of the mass distribution of fission products with bremsstrahlung, since they have been interested in the reaction mechanism. We tried to obtain the absolute yield of the mass distribution of fission products, as a basic study on the transmutation of actinides in high level radioactive wastes by using photonuclear reactions. The high level radioactive wastes produced during reprocessing of spent nuclear fuels include the long-lived radionuclides of fission products such as ^{90}Sr and ^{137}Cs, and actinides such as U, Pu, Np, Am, and Cm.

In this report, we attempted to apply photonuclear reactions due to gamma rays having several tenth MeV enegy to the transmutation study, because of their advantage that intense high energy gamma rays can be more easily and cheaply obtained from the bremsstrahlung produced by electron linear accelerator than high energy proton beams. As a basic study for this purpose, we have started the experiment to get the absolute yield and the amounts of transmutation due to photofission of ^{238}U, ^{237}Np and ^{239}Pu.

Experimental

The experiments were performed by using two electron linear accelerators of Laboratory of Nuclear Science, Tohoku Uuiversity (Ee = 30 and 60 MeV) and Nuclear Engineering Research Laboratory, University of Tokyo (Ee=20 and 30 MeV)

Figure 1 shows a schematic view of the experimental arrangement. Targets of ^{238}U, ^{237}Np and ^{239}Pu nuclides were irradiated by the bremsstrahlung beam generated in the 1-mm-thick Pt converter. The electrons passed through the converter were bended downward with a cleaning magnet only in the LINAC of Tohoku University.

The ^{237}Np target is about 50 $\mu g/cm^2$ in thickness of 99.3% enriched ^{237}Np deposited on a nickel metal plate and its activity is 43.2 ± 0.9 nCi. The ^{239}Pu target (about 50 $\mu g/cm^2$ in thickness) is made of 99.3% enriched ^{239}Pu deposited on a nickel metal plate and its activity is 1.66 ± 0.03 μCi. The ^{238}U target is 0.025-mm-thick metal enriched up to 99.959% ^{238}U.

Each target was covered with a 0.1-mm-thick aluminum or polyethylene catcherfoil to collect fission products and a 0.01-mm-thick gold foil to measure the bremsstrahlung flux. The bremsstrahlung flux injected on the target was estimated from the yield of ^{196}Au which was pro-

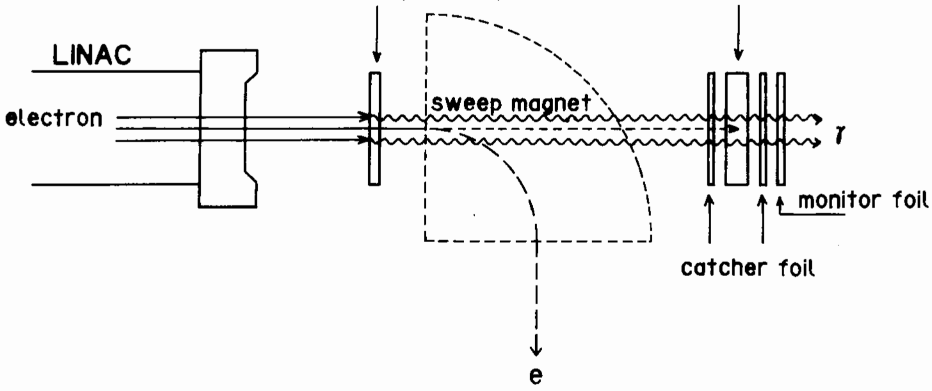

Fig. 1 Schematic view of the experimental arrangement

duced by $^{197}Au(\gamma, n)$ reaction by approximating the bremsstrahlung spectrum to be equal to the Schiff's formula[1]. The induced radioactivities of the catcher-foils attached on each target and ^{238}U target itself were measured with a pure-Ge detector and the nuclides and their activities were determined by analyzing with the NLAB system (NAIG Co. Ltd.).

The detail on this experiment is described elsewhere[2].

Results and Discussion

Mass yield distribution

Figures 2 and 3 show the photofission mass yield distributions of ^{238}U and ^{237}Np, respectively, for 20-, 30- and 60-MeV bremsstrahlung. The 41 nuclides were identified from the catcherfoil of ^{238}U, but fewer nuclides from the foils of ^{237}Np and ^{239}Pu, because of much less weights of ^{237}Np and ^{239}Pu. In order to get the absolute mass yields, the activities of catcherfoils and ^{238}U target were summed up, since some fission fragments produced remained in a thick ^{238}U target, while on the other hand, all fission fragments were approximated to be recoiled out from the ^{237}Np and ^{239}Pu targets.

Solid lines indicate the mass yield distributions of ^{238}U and ^{237}Np reported by E. Jacobs et al.[3] and M. Ya. Kondrat'ko et al[4], respectively. Their results are relative yields and are normalized to the present results. Since we did not use the chemical separation technique, the present results could not give the valley of the mass yield distribution corresponding to thesymmetric fission, and also indicate some fluctuations in the mass distribution. But as a whole, our results show good agreement with the other experimental results.

Transmutation yields

By integrating these yield curves, the transformation yields of the target nuclides, Δm, were obtained experimentally. The transmutation yields can also be calculated by using the following expression,

$$\Delta m = m \int_{E_{th}}^{E_0} \sigma(\gamma, n)\, \phi(E)\, dE = m \int_{E_{th}}^{E_0} \sigma(\gamma, f)\, \phi(E)\, dE \qquad (1)$$

m: weight of target nuclide
E : electron energy
Eth: threshold energy
$\phi(E)$: bremsstrahlung flux
$\sigma(\gamma, n)$: cross section of ^{238}U, ^{237}Np, ^{239}Pu (γ, n) reaction
$\sigma(\gamma, f)$: cross section of ^{238}U, ^{237}Np, ^{239}Pu (γ, f) reaction

The $\phi(E)$ is estimated from the ^{196}Au activity, A, as follows,

$$A = \int \phi(E)\, \sigma_{Au}(\gamma, n)\, dE \qquad (2)$$

where $\sigma_{Au}(\gamma, n)$ is the $^{197}Au(\gamma, n)$ reaction cross section given in Ref. (5). The (γ, n) and (γ, f) cross sections for ^{238}U and ^{237}Np are given in Ref. (6).

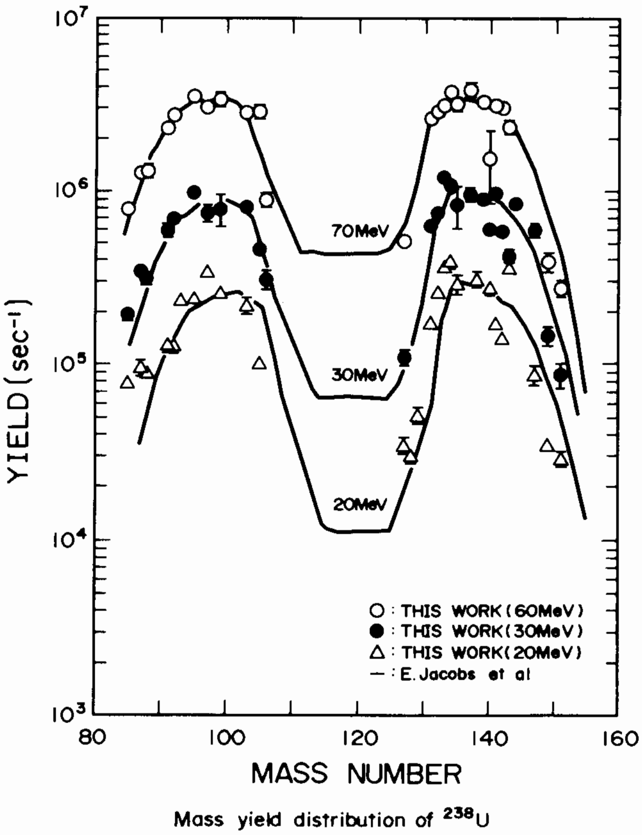

Fig. 2 Photofission mass yield distributions of ^{238}U for 20-, 30- and 60-MeV bremsstrahlung, compared with Jacobs' results[3]

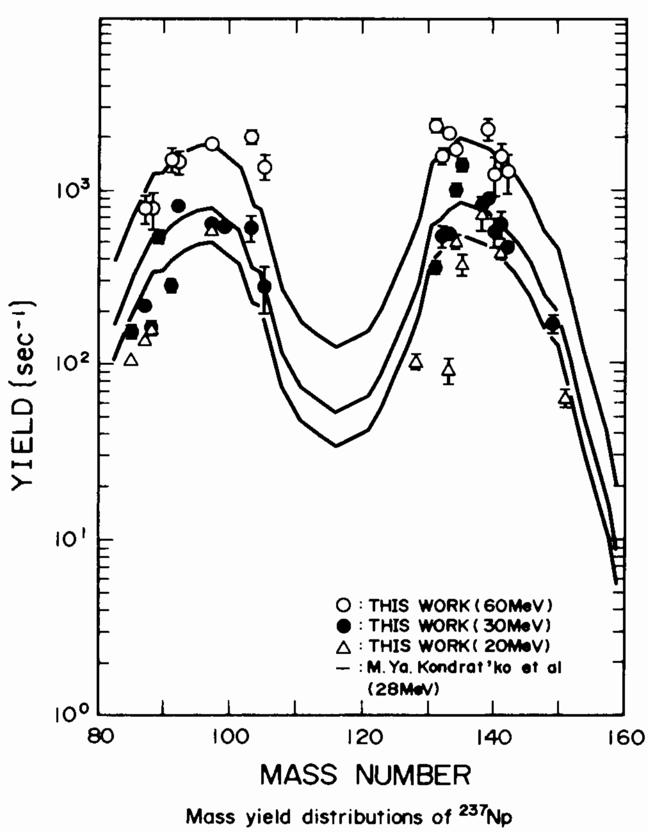

Fig. 3 Photofission mass yield distributions of ^{237}Np for 20-, 30- and 60-MeV bremsstrahlung, compared with Kondrat'kos' result[4]

Table 1 shows the comparison of the experimental and calculated values of transmutation yields. The experi-me ntal yields of ^{238}U (γ , f) reaction show good agreement with the results calculated from Eq. (1) within 10% difference. But for ^{237}Np (γ , f) reaction, the difference between experimental and calculated yields is a little larger. This may come from the errors of the extrapolated values of ^{237}Np (γ , f) cross section data given in Ref. (6), since the data is limited in the photon energy below 18 MeV. The experimental yield of ^{238}U (γ , n) reaction is about 30% larger than the calculated yield.

Considering that the photofission occurs above about 8MeV photon energy, the transmutation rate was obtained by dividing the transmutation yield, $\Delta m/m$, by the bremsstrahlung flux integrated above 8 MeV. The transmutation rates given in the last column of Table 1 show the tendency that the rates slightly decrease with the bremsstrahlung energy, but the bremsstrahlung flux generated from one electron steeply increases with the electron energy, then the transmutation yield per one electron increases with the electron energy.

The transmutation yield thus obtained will be useful as the basic data for the design of photonuclear transmutation of radioactive wastes.

References

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Table 1 Comparison of measured and calculated transmutation yields for ^{238}U and ^{237}Np

Target	Electron Energy Ee (MeV)	Target Weight m(g) $\times 10^{-5}$	Nuclear Reaction	Transmutation Yield Δm (g・s^{-1}) Experimental $\times 10^{-18}$	Calculated $\times 10^{-18}$	Photon Flux (>8MeV)* $\phi \cdot S$ (s^{-1}) $\times 10^{+11}$	Transmutation Rate $\Delta m/(m\phi S)$ $\times 10^{-26}$
^{238}U	20	8070	(γ , f)	1720	1870	3.37	6.32
			(γ , n)	2390	3140		8.78
^{238}U	30	7170	(γ , f)	5060	4110	11.0	6.42
			(γ , n)	(706)**	6090		—
^{238}U	30	7330	(γ , f)	5830	5260	13.8	5.76
			(γ , n)	10600	7470		10.5
		7090	(γ , f)	5950	5500	15.1	5.56
			(γ , n)	10700	7870		10.0
^{238}U	60	7330	(γ , f)	22100	20100	72.0	4.19
			(γ , n)	36900	25500		6.99
^{237}Np	20	6.13	(γ , f)	3.29	2.00	2.32	23.1
^{237}Np	30	6.13	(γ , f)	11.8	7.87	13.5	14.2
^{237}Np	30	3.01	(γ , f)	5.04	4.05	14.5	11.5
^{237}Np	60	3.01	(γ , f)	11.7	12.2	77.9	4.99

* $S = (1.27/2)^2 \pi = 1.27$ (cm^2) : Target size

** Photon peak of ^{237}U was underestimated due to the background from other strong peaks in the highly activated target.