Product Yields for the Photofission of 238U, 237Np and 239Pu

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V. 3 Product Yields for the Photofission of ^{238}U, ^{237}Np and ^{239}Pu

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1. Introduction

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 the present, the geological storage is considered to be the dominant approach for radioactive waste management, but it would be quite worthwhile for the reduction of nuclear wastes if these long-lived radionuclides could be transmutated efficiently into the short-lived or stable radionuclides. This nuclear waste transmutation technique has previously been examined by using the reactor neutrons and the accelerator produced protons.1)

There has ever been published only one experimental work which attempted to a contribution to the nuclear waste transmutation based on the proton spallation reaction.2)

In this report, we attempted to apply photonuclear reactions due to gamma rays having several tenth MeV energy to the transmutation study, because of their advantage that intense high energy gamma rays can be more easily and chéaply 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 of nuclear transmutation due to photofission of ^{238}U, ^{237}Np and ^{239}Pu.

Several studies have already been published on the relative yield of the mass distribution of fission products for the photofission of ^{237}Np and ^{238}U with bremsstrahlung.

Here, we obtained the absolute yield of the mass distribution of fission products and then the amounts of transmutation of ^{238}U, ^{237}Np and ^{239}Pu nuclides which are useful basic data for evaluation of the feasibility study of photo-transmutation of nuclear wastes.

2. Experimental

The irradiation were performed by using the LINAC of Laboratory of Nuclear Science, Tohoku University and Nuclear Engineering Research Laboratory, University of Tokyo.

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

The ^{237}Np target is about 50 μg/cm^2 in thickness of 99.3 % enriched ^{237}Np deposited on a nickel metal plate. The ^{239}Pu target 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 catcher foil to collect fission products and 0.01 mm thick gold foil to measure bremsstrahlung flux. The bremsstrahlung flux injected on the target was calculated from the yield of ^{196}Au which was produced by (γ,n) reaction from ^{197}Au. The induced radioactivities of the catcher foils of each targets and ^{238}U target were measured with a pure-Ge detector and the measured data were analyzed to determine nuclides and their activities by the NLAB system (NAIG Co. Ltd.).

The more detail conditions with this experiment are described elsewhere.[3,4)]

3. Results and Discussion

Figures 2 and 3 are the photofission mass yield distributions of ^{238}U and ^{237}Np, respectively, for 20-, 30- and 60-MeV bremsstrahlung. Solid lines indicate mass yield distributions of ^{238}U and ^{237}Np reported by E. Jacobs[5)] and M. Ya. Kondrat'ko[6)], respectively. Since we did not use the chemical separation technique, our results could not give the valley of the mass yield distribution corresponding to the symmetric fission, and also indicate some fluctuations in the mass distribution. But as a whole, our result show good agreement with the other experimental results.

By integrating these yield curves, we obtained the transmutation yields of the target nuclides experimentally. On the other hand, transmutation yields are computed by using the following expression.

$$\Delta m = m \int_{E_{th}}^{E_0} \sigma_{(\gamma,n)} \Phi(E)dE = m \int_{E_{th}}^{E_0} \sigma_{(\gamma,f)} \Phi(E)dE,$$

m: weight of target nuclide
E_0: electron energy
Φ(E): flux of bremsstrahlung
σ(γ,n): cross section of (γ,n) reaction
σ(γ,f): cross section of (γ,f) reaction.

Table 1 shows the comparison of the experimental and computed values of transmutation yields. The experimental yields show pretty good agreement with the computed yields.

Taking account for that the photofission occurs at photon energy about 8 MeV, we evaluated the transmutation rate, that is, the transmutation yield divided by the bremsstrahlung flux integrated above 8 MeV in its energy. The transmutation rates of the last column in Table 1 shows the tendency that the rates slightly decrease with the bremsstrahlung energy, but the bremsstrahlung flux per one electron rapidly increases with the electron energy, then the transmutation yield per one electron increase with the electron energy.

We are now performing the similar experiments with 30- and 60-MeV bremsstrahlung, using the chemical separation tehcnique.

References

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Table 1. Comparison of the experimental and computed values of transmutation yields.

Target	Electron energy E_0(MeV)	Weight m(g) $\times10^{-5}$	Reaction	Transmutation yield		Photon flux $\Phi\cdot S$(>8MeV)* $\times10^{11}$	$\Delta m/(m\Phi S)$ (γ^{-1}) $\times10^{-26}$
				Experimental $\times10^{-18}$	Computed $\times10^{-18}$		
^{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): Area of target.

**Target was strongly activated and small photopeak of ^{237}U was underestimated.

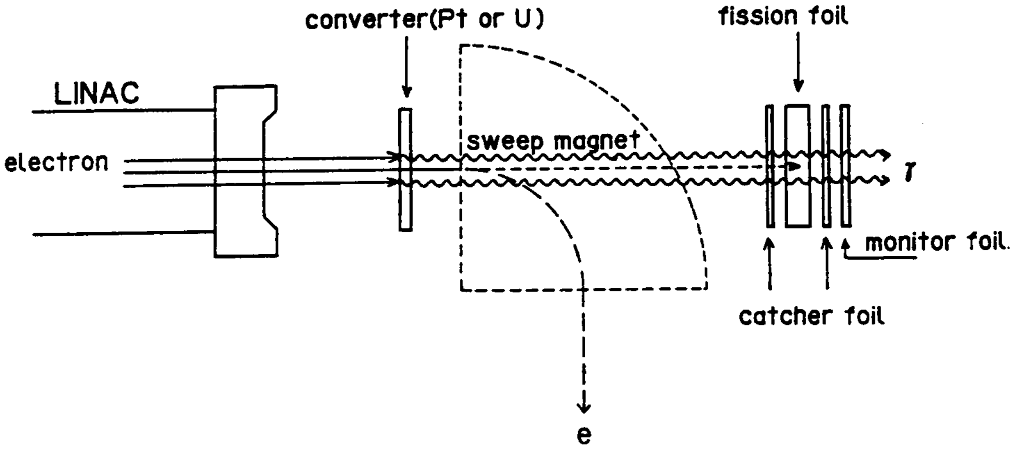

Fig. 1. Schematic veiw of the experimental arrangement.

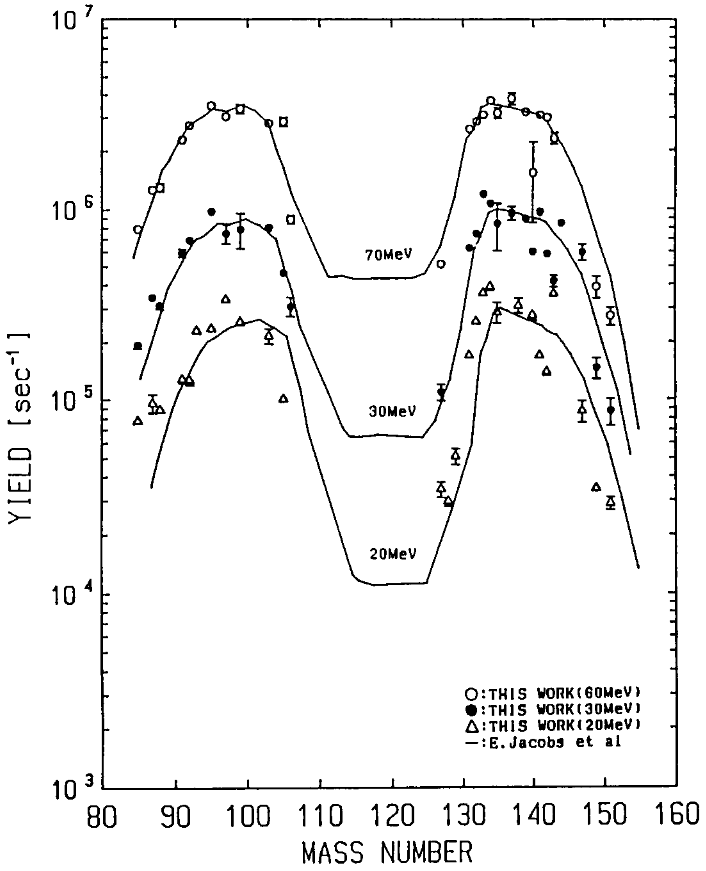

Fig. 2. Photofission mass yield distributions of ^{238}U for 20-, 30- and 60-MeV bremsstrahlung, compared with Jacobs' results.[5)]

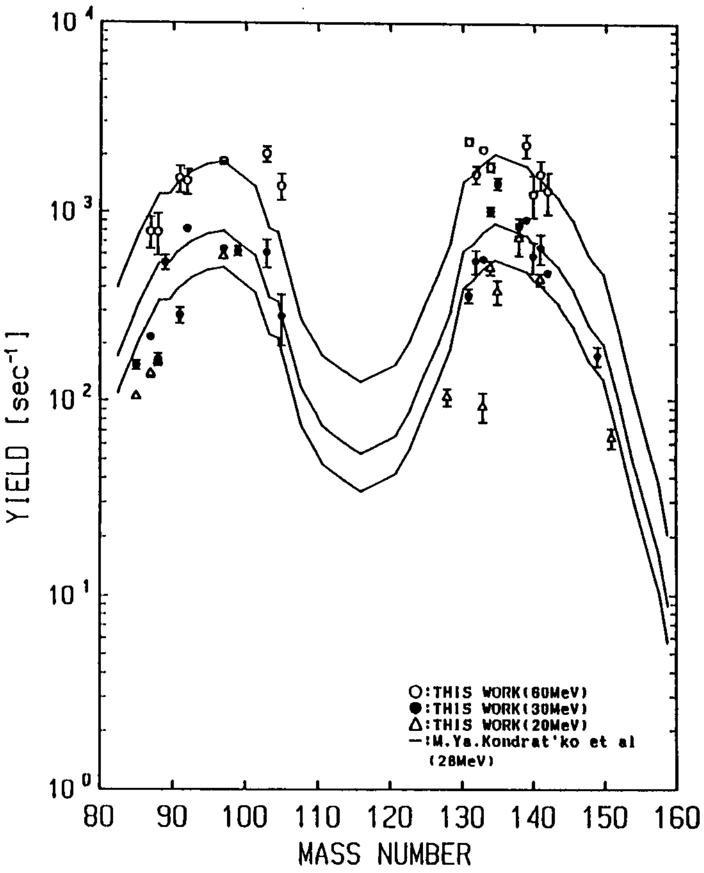

Fig. 3. Photofission mass yield distributions of ^{237}Np for 20-, 30- and 60-MeV bremsstrahlung, compared with Kondrat'kos' results.[6)]