Irradiation of Natural Uranium by the Neutrons of the Japan Material Testing Reactor

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Synopsis

A 2.76 g of natural uranium dioxide was irradiated by the neturons of JMTR for about 60 days from the 7th cycle to the 9th cycle. U and Pu were isolated and the isotopic ratios were measured. The nature of the neturon capture reactions was discussed.

I. Introduction

In order to get enough transuranium elements, it is necessary to irradiate the target consisting of ^{238}U, ^{239}Pu, ^{241}Am, or ^{243}Am by more than 10^{21} nvt of neturons. In 1970, the Japan Atomic Energy Research Institute began irradiation service by the Japan Material Testing Reactor (JMTR) at Oarai Research Establishment and it became possible to irradate actinoid targets by more than $10^{14}n/cm^2$. sec of neutrons.

Actually, we have a plan to produce ^{252}Cf from ^{241}Am and ^{243}Am. But at the first stage, we must exersice in handling heavily irradiated target which constains a large amount of fission products and α-emitters. As the preliminary experiment, natural uranium was irradiated in the test running period of JMTR. By the analysis of the isotopic ratios of U and Pu, we also tried to know the nature of the neturon reactions of JMTR.

II. Experimental

Irradiation. Uranium dioxide pellets which contained 2.76 g of natural uranium dioxide were irradiated from the 7th to the 9th cycle of JMTR at the N–10 irradiation hole situated in Be reflector zone. The irradiated uranium was left to stand for about 140 days to reduce the radioactivity of fission products.

Chemical Separation. The irradiated U was separated by the use of the chemical processing equipment constructed by the authors in the 6th hot-cell of the Hot Laboratory of the Oarai Laboratory for Irradiation Experiment of the Research

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Institute for Iron, Steel and Other Metals. The outline of the chemical processing equipment is shown in Fig. 1.

The pellets were dissolved in a 6M HNO_3 solution and Pu was prepared to the tetravalent state by the addition of equal volume of diluted $NaNO_2$ solution. Then U(VI), Pu(IV), and Np(VI) were extracted by 30% (v/v) TBP-xylene solution (the first extraction). After the scrubbing the organic phase by a 3M HNO_3 solution, U and Pu were back-extracted by a 0.01M HNO_3 solution (the second extraction). The flow chart of the separation method is shown in Fig. 2.

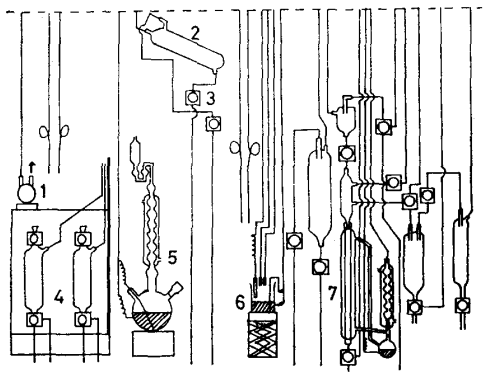

Fig. 1.

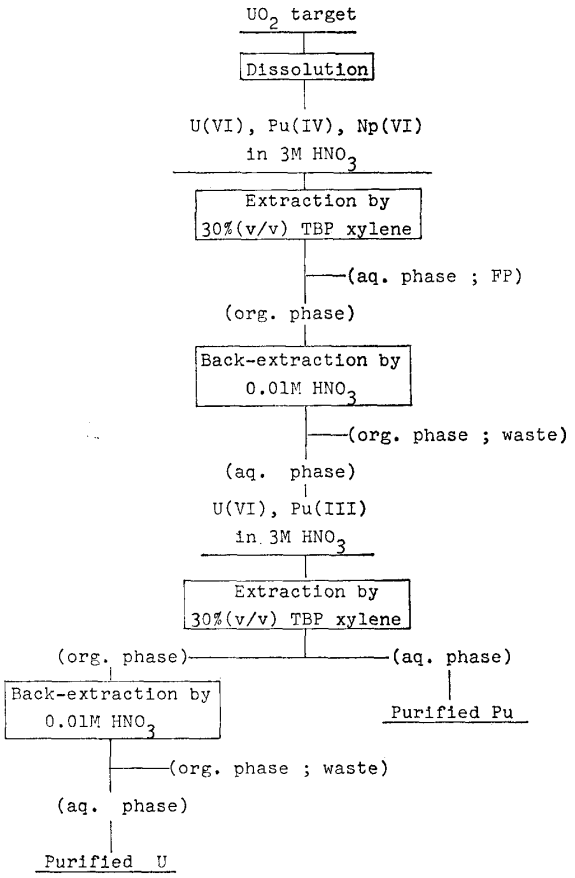

Fig. 2.

Fig. 1. Outline of the chemical processing equipment in the hot-cell.
1, A pump for the liquid-transport; 2, Dispenser; 3, Teflon-cock; 4, Extraction apparatus; 5, Dissolution flask; 6, Reaction vessel for oxidation-reduction reactions and pH adjustment; 7, Ion-exchange column with a jacket for the temperature control. All the dispensers, extraction vessels, flasks, beaker and ion-exchange column are made of Pyrex glass. Pipes for the liquid-transport are made of polyethylene tube irradiated by electrons.

Fig. 2. A flow chart for the chemical separation.

To obtain the purified U and Pu, the mutual separation of U and Pu was carried out, as is also shown in Fig. 2. The separation method was the similar method to the first extraction, whereas Pu was reduced to the trivalent state in order to be retained in the aqueous solution.

Alpha-ray and Gamma-ray Spectrometry. To examine the separation method which was carried out by means of remote operation in the hot-cell, the chemical yields of U, Pu, and some fission products were measured by α-ray and γ-ray spectrometry. A Si surface-barrier detector (ORTEC model BE–16–160–100) was applied to the determination of α-ray spectra and a Ge(Li) detector (ORTEC model 8101–1820) was applied to the determination of γ-ray spectra. A 4096ch. pulse-height-analyzer (TOSHIBA USC–1 model 10) was used for the accumulation of the spectra.

Mass Spectrometry. A mass spectrometer (ATLAS CH–4) was used for the measurement of the isotopic composition of U and Pu. An aliquot of the purified solution of U and Pu, which contained about 10 μg of U and 1 μg of Pu, respectively, was taken onto rhenium ribbon filaments and was evaporated to dryness. Ionization cases of the double filaments type were used throughout the scanning of mass spectra. For the detection of weak ion beam, a secondary electron multiplier tube was used. The operation condition of the mass spectrometer is summarized in Table 1.

Table 1. Operation condition of the mass spectrometer

Ion acceleration voltage		3.0 kV
Slit width	exit slit	0.1 mm
	collector slit	2.3 mm
Resolution		430 for Hg
Filament current	evaporation	2.6~3.2 A
	ionization	4.6~5.3 A

III. Results and discussion

Chemical Separation. We observed ^{144}Ce, ^{106}Ru, ^{134}Cs, ^{137}Cs, ^{95}Zr, and ^{95}Nb in the γ-ray spectrum of the irradiated U. The distribution of these nuclides was measured and the results are shown in Table 2, where the U-Pu fraction is the

Table 2. Distribution of the fission products in the U-Pu fraction

Fission product	Yield(%)	Total (mCi)[1)]
^{144}Ce	2.9	310
^{106}Ru	4.5	6.1
^{137}Cs	1.6	4.3
^{134}Cs	1.6	0.27
^{95}Zr - ^{95}Nb	5.2	~5

1) Normalized to the end of the irradiation.

aqueous phase which was obtained by the second extraction. As is shown in Table 2, all the fission products were effectively removed from the U-Pu fraction. We

also measured the chemical yield of Pu in the U-Pu fraction by α-ray spectrometry and found that the yield was greater than 90%. Form these observations, we concluded that the remote operation for the separation was successful.

Isotopic Composition of U and Pu. A α-ray spectrum of the purified U, which was the sample for mass analysis, is shown in Fig. 3. Even in the purified U, α-rays of Pu isotopes were observed. For the mass analysis of U, ^{238}Pu have influence on

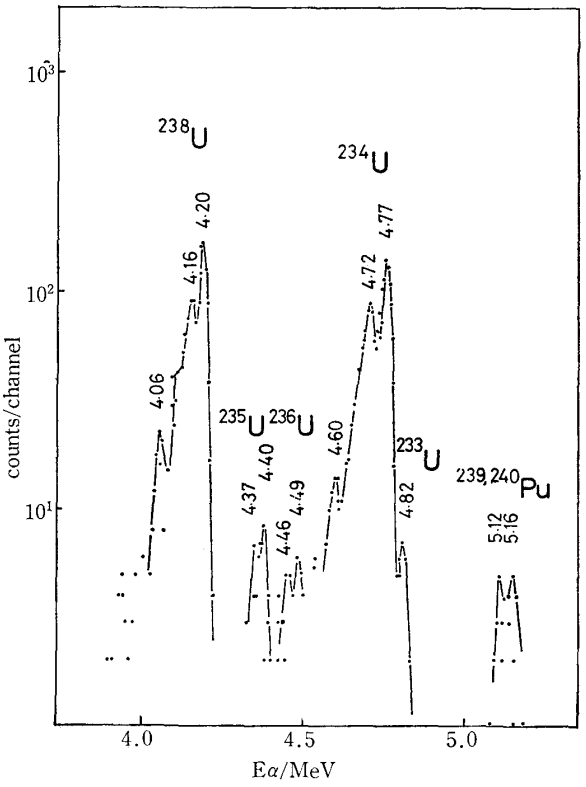

Fig. 3. Alpha-ray spectrum of the purified U.

the determination of the mass peak of ^{238}U. But it is always obvious that the content of ^{238}Pu in the Pu isotopes produced from ^{238}U is very small. In addition, the half-life of ^{238}Pu is much shorter than ^{238}U, ^{239}Pu, and ^{240}Pu. Therefore, we can neglect the influence of ^{238}Pu on the mass peak of ^{238}U, even if the trace contamination by Pu was observed in the α-ray spectrum of the mass sample of U.

Similarly, the complete removal of U from the purified Pu is very difficult. An example of the mass spectrum of the purified Pu is shown in Fig. 4. In Fig. 4, two spectral patterns of the mass peaks of 238, 239, and 240 are shown and the second spectral patterns was obtained about 3 minutes later from the time of the determination of the first pattern. Obviously, the mass peaks of 239 and 240 are attributable to ^{239}Pu and ^{240}Pu, respectively. As is shown in Fig. 4, the mass peak of 238 which would be attributable to ^{238}Pu or ^{238}U decreased markedly within the duration of the spectrum determination whereas the mass peaks of 239 and 240 increased slightly. This fact indicates that the mass peak of 238 is attribtable

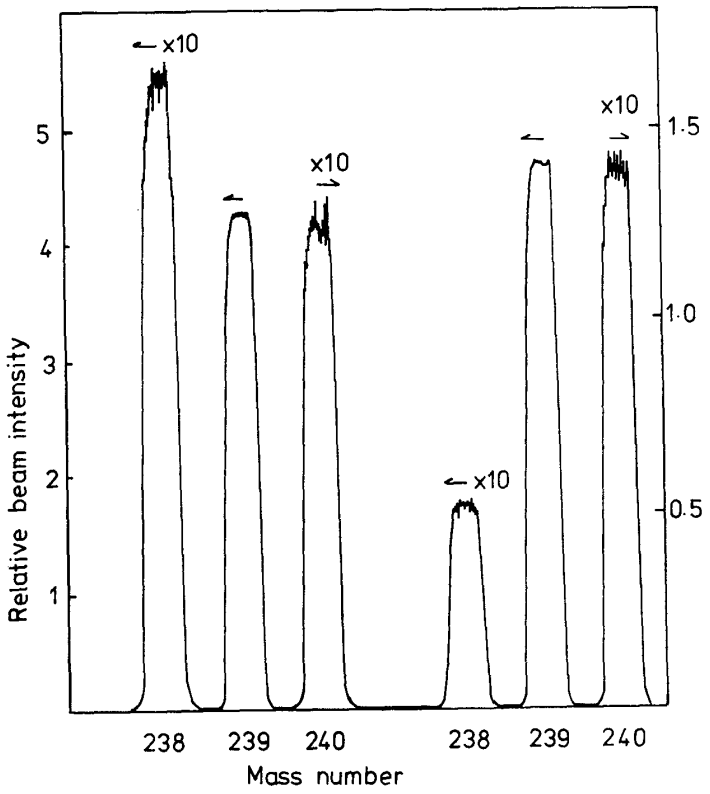

Fig. 4. Mass peaks of 238, 239, and 240 observed in the purified Pu.

to ^{238}U and is not attributable to ^{238}Pu. The difference between the characteristics of the ion beams of Pu and U will be explained by the difference of the vapour pressure of Pu and U. As is typically shown in Fig. 3, we observed the α-ray of ^{233}U which is undoubtedly the product of the fast neutron reaction on ^{234}U, i.e., $^{234}U(n, 2n)^{233}U$. ^{238}Pu is also produced by the fast neutron reaction on ^{238}U or ^{239}Pu. But the cross sections of thermal neutron reactions are generally much greater than the cross sections of (n, 2n) reactions. Then, we did not consider about the fast neutron reactions.

Consequently, we measured the istopic ratios between ^{235}U, ^{236}U, and ^{238}U and between ^{239}Pu, ^{240}Pu, and ^{241}Pu. The examples of the mass spectra are shown in Figs. 5, 6, and 7. And the isotopic ratios are shown in Table 3.

Table 3. The isotopic ratio of U and Pu in the irradiated target

	Measured (×100)	Calculated (×100)
$^{235}U/^{238}U$	0.657±0.010	0.657
$^{236}U/^{235}U$	1.86 ±0.23	1.650
$^{240}Pu/^{239}Pu$	2.846±0.045	2.873
$^{241}Pu/^{240}Pu$	3.29 ±0.20	3.270

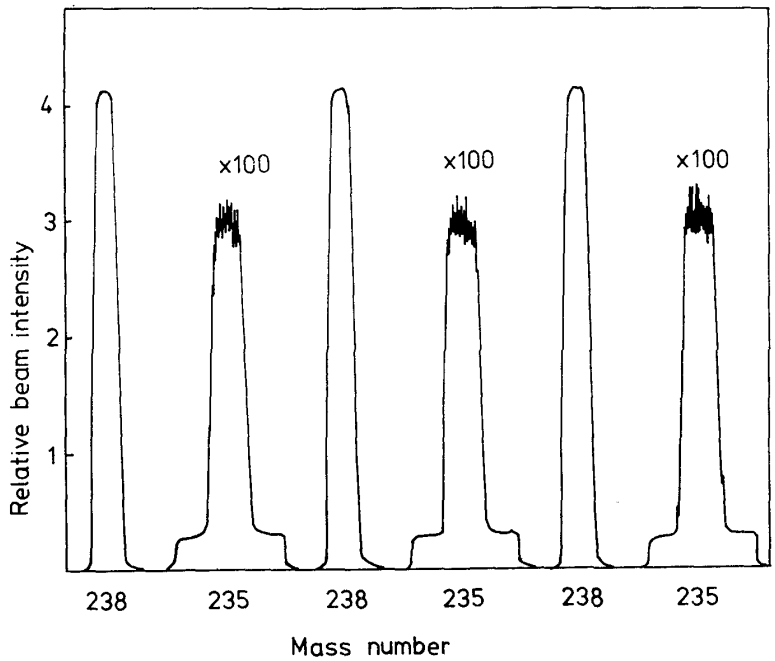

Fig. 5. Mass peaks of ^{238}U and ^{235}U.

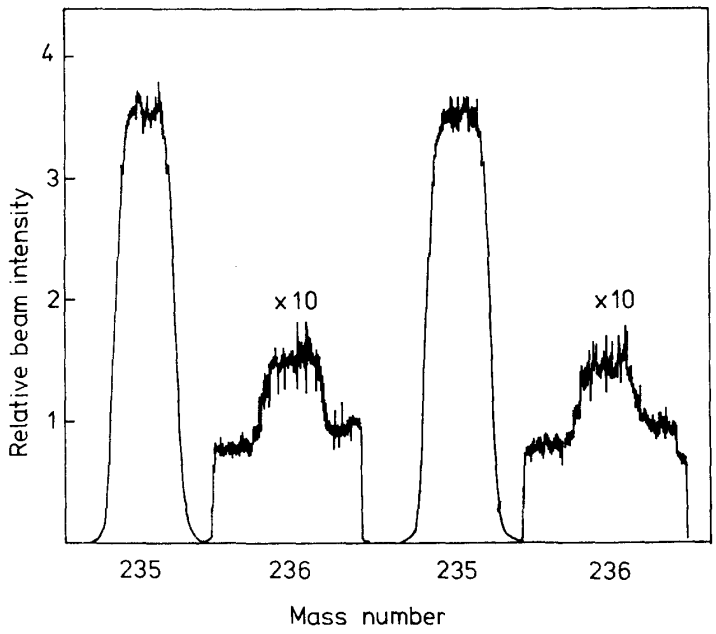

Fig. 6. Mass peaks of ^{236}U and ^{235}U.

Build-up of Pu and the Heavier Nuclides. For the calculation of the build-up of transuranium elements, M. Hara *et al.*, wrote a computer program to calculate the yields of the chain reactions. The complete description of the program is given in Ref. 1. The program have the ability to construct the build-up chains by the program itself. And then, we can easily take into account for all the production chains of every nuclide.

(1) M. Hara, T. Mitsugashira, A. Satô and S. Suzuki, Sci. Rep. RITU, **A28** (1979), 41.

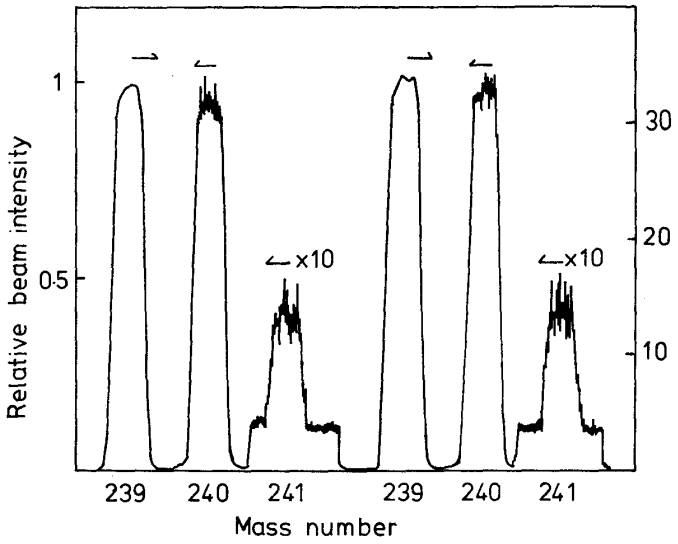

Fig. 7. Mass spectrum of the purified Pu.

To evaluate the rate constants of the nuclear transformation reactions, we used the data tabulated in Table 4.[2-5] The effective cross sections were calculated by the method of C.H. Westcott.[6,7] We also took into account for the fluctuation of neutron flux during the irradiation and assumed that the flux is proportional to the reactor power. The yields and the isotopic ratios of actinides were calculated adjusting two adjustable parameters, i.e., the epithermal ratio which is defined with respect to the fluence[6,7] and the thermal neutron flux. The results of the calculation, which were obtained by means of the best set of the adjustable parameters, are shown in Table 3.

As is shown in Table 3, the calculated isotopic ratios were agreed well with the measured values. As the result, we could conclude that the epithermal ratio was 0.06 and that the thermal neutron flux was 3.17×10^{13}n/cm^2·sec at the N–10 irradiation hole when the reactor power was 30 MWt. The effective cross sections are summarized in Table 4. As we did not consider about the absorption of neutron by the target, the actual flux and the epithermal ratio will be 1.7–2.0 times greater than the fitted values. And the maximum power of JMTR is 50MWt. Therefore, we can say that the neutron flux of JMTR is high enough to apply JMTR to the production of transcurium elements from ^{241}Am or ^{243}Am.

As is described above, the nature of the neutrons of JMTR is sutiable for the production of transcurium elements. On the planning of the production of transcurium elements, we often ignore the contribution by the minor production

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(3) R.W. Stoughton and J. Halperin, Nucl. Sci. Engng., **6** (1959) 10.
(4) H. Ihle, H. Michael, A. Neubert, A.J.F. Blair, P. Damle, and M.V. Bodnarescu, J. Inorg. Nucl. Chem., **34** (1972) 2427.
(5) O.J. Eder and M. Lammer, IAEA-SM-170/12 (1972).
(6) C.H. Westcott, AECL–1101 (1960).
(7) C.H. Westcott, AECL–3255 (1969).

Table 4. Thermal cross sections at 70°C[1], resonance integrals, and effective cross sections

Nuclides	Half-life	Decay	Capture cross section (barns)			Fission cross section (barns)		
			Thermal	Σ[2]	σ_{eff}	Thermal	Σ[2]	σ_{eff}
^{234}U	2.47×10^{5}y	α	99	600	142			
^{235}U	7.1×10^{8}y	α	98.2	140	108	584	270	603
^{236}U	2.39×10^{7}y	α	5.4	415	35			
^{237}U	6.75 d	β^-	370		370	2		2
^{238}U	4.51×10^{9}y	α	2.73	269	22.1			
^{239}U	23.54 m	β^-	22		22	14		14
^{240}U	14.1 h	β^-						
^{237}Np	2.14×10^{6}y	α	169	870	232	0.019	6	0.5
^{238}Np	2.1 d	β^-	43	10	44	2200	510	2237
^{239}Np	2.35 d	β^-	25 (g) 35 (m)		25 (g) 35 (m)			
^{240}Np	63 m	β^-						
^{240m}Np	7.3 m	β^-						
^{238}Pu	86.4 y	α	588	164	600	16.3	16.7	17.5
^{239}Pu	24390 y	α	270	1390	370	740	2270	903
^{240}Pu	6580 y	α	288	8220	878			
^{241}Pu	13.2 y	β^- 99.8% α 0.0023%	365	160	377	1010	580	1052
^{242}Pu	3.79×10^{5}y	α	20		20			

1) Temperature of the moderator H_2O. 2) Resonance integral.

chains even when the high flux reactor is available. We found out the following fact by the calculation of the production of ^{240}Pu from ^{238}U.

The main production chain for ^{240}Pu is

$$^{238}U(n,\gamma)\,^{239}U \xrightarrow[\beta^-]{} {}^{239}Np \xrightarrow[\beta^-]{} {}^{239}Pu(n,\gamma)\,^{240}Pu \qquad (1).$$

But it is possible to get ^{240}Pu as follows.

$$^{238}U(n,\gamma)\,^{239}U \xrightarrow[\beta^-]{} {}^{239}Np(n,\gamma)\,^{240m,240}Np \xrightarrow[\beta^-]{} {}^{240}Pu \qquad (2)$$

$$^{248}U(n,\gamma)\,^{239}U(n,\gamma)\,^{240}U \xrightarrow[\beta^-]{} {}^{240m,240}Np \xrightarrow[\beta^-]{} {}^{240}Pu \qquad (3)$$

When the thermal neutron flux is 3.17×10^{13}n/cm^2·sec and the epithermal ratio is 0.06, the branching ratio of (n, γ) reaction with respect to the β^--decay is only 10^{-6} and the branching ratio of (n, γ) reaction of ^{239}Np is only 10^{-4}, as is easily under-

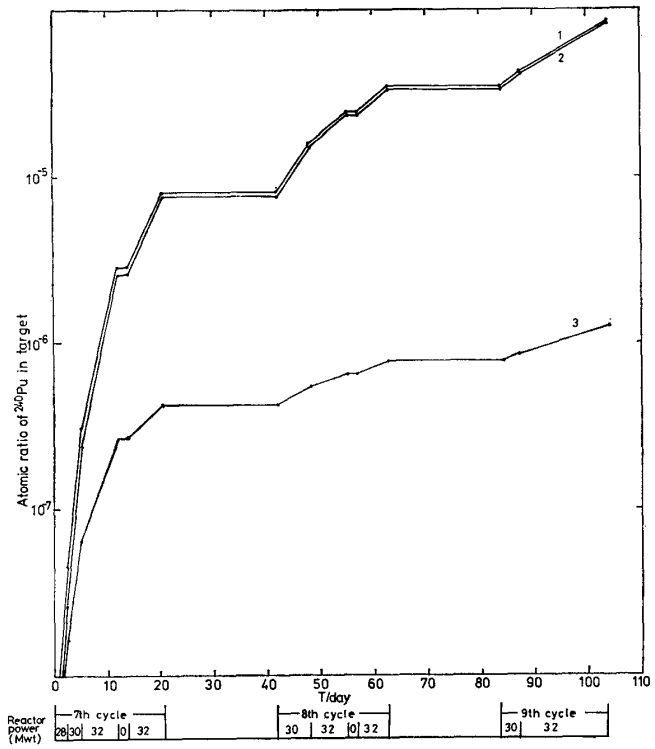

Fig. 8. Build-up of ^{240}Pu from ^{238}U.
1, Total ^{240}Pu; 2, ^{240}Pu produced via ^{239}Pu, i.e., by the reaction (1); 3, ^{240}Pu produced via ^{240m,240}Np, i.e., by the reactions (2) and (3).

stood from the data in Table 4. Therefore, for the production of ^{240}Pu and the heavier nuclides, the contribution by the reaction (1) is much higher than that of the reactions (2) and (3). But it is necessary to accumulate appreciable ^{239}Pu in order to produce ^{240}Pu by the reaction (1). The build-up of ^{240}Pu in the irradiation condition of the present work is shown in Fig. 8. As is shown in Fig. 8, the contribution by the reactions (2) and (3), i.e., via ^{240m,240}Np, can not be negligible. According to our calculation, the ratio of ^{240}Pu produced by the reactions (2) and (3) to the total amount of ^{240}Pu is 4.7% at the end of the 7th cycle, 2.3% at the end of the 8th cycle, and 1.5% at the end of the 9th cycle. Thus the ratio is decreased with the increase of the irradiation time or with the build-up of ^{239}Pu. But it should be mentioned that the rate constant of (n, γ) reaction of the nuclide of short half-life increases with the increase of the neturon flux. Therefore, the contribution of the (n, γ) reaction of the nuclide of short half-life can not be ignored when the irradiation is carried out by JMTR or by the reactors of the higher neutron flux.

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