Chapter 6

Nuclear Transmutation of Minor Actinide

Cheol Ho Pyeon

Abstract Integral experiments on critical irradiation of neptunium-237 (^{237}Np) and americium-241 (^{241}Am) foils are carried out in a hard spectrum core at KUCA with the use of the back-to-back fission chamber, and Monte Carlo calculations together with a reference nuclear data library are conducted for confirming the precision of numerical simulations. Subcritical irradiation of minor actinide (MA) by ADS is a very important step, before operating actual ADS facilities, in a critical assembly at zero power, such as KUCA, which is an exclusive facility for ADS that comprises a uranium-235 (^{235}U) fueled core and a 100 MeV proton accelerator. The first significant attempt is made to demonstrate the principle of nuclear transmutation of MA by ADS through the injection of high-energy neutrons into the KUCA core at a subcritical state. Here, the main targets of nuclear transmutation of MA by the ADS experiments are fission reactions of ^{237}Np and ^{241}Am , and capture reactions of ^{237}Np .

Keywords Nuclear transmutation · Minor actinides · BTB fission chamber · Neptunium-237 · Americium-241

6.1 Integral Experiments at Critical State

6.1.1 Critical Irradiation Experiments

6.1.1.1 Core Configuration

Critical irradiation experiments [1] were carried out in the A-core (reference core; Fig. 6.1) that has polyethylene (PE) moderator and reflector rods, and two fuel assemblies: normal “F” and special “f” (Fig. 6.2a, b, respectively). The normal fuel assembly “F” is composed of 60 unit cells, and upper and lower polyethylene blocks about 23” and 21” long, respectively, in an aluminum (Al) sheath ($2.1 \times 2.1 \times 60$ ”).

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Fig. 6.1 Top view of the KUCA core (reference core) for critical irradiation experiments on MA (Ref. [1])

A unit cell is composed of two highly enriched uranium (HEU) fuel plates $2 \times 2''$ square and $1/8''$ thick ($1/16'' \times 2$), polyethylene plates $2 \times 2''$ square and $1/8''$ thick, for the normal fuel plate “F” (HEU-PE). The special fuel assembly “f” (HEU-Pb) is composed of 40 unit cells with two HEU fuel plates $1/8''$ ($1/16'' \times 2$) thick, lead (Pb) plate $2 \times 2''$ square and $1/8''$ thick, 20 unit cells of HEU and polyethylene plates as well as in the normal fuel assembly.

6.1.1.2 Back-to-Back Fission Chamber

Critical irradiation experiments for measuring the fission reaction rate ratio were carried out with the back-to-back (BTB) fission chamber (Fig. 6.3) set inside a special void element, containing two foils: a test foil neptunium-237 (^{237}Np) or americium-241 (^{241}Am) and a reference one uranium-235 (^{235}U). The reference foil ^{235}U was a normalization factor that has the fission reactions $^{235}\text{U}(n, f)$, to obtain two fission reaction rate ratios: $^{237}\text{Np}/^{235}\text{U}$ and $^{241}\text{Am}/^{235}\text{U}$. The main characteristic of the BTB chamber is to obtain uniquely an original signal of fission event of one foil attached at the position of sample deposit, and a different signal of fission event of the other foil attached at the opposite side without any disturbance of the foil on the front side. The chamber has a structure of separating it from two electro-deposited foils that receive independently pulsed signals of fission events accumulated as electric pulses generated by fission fragments through the ionization process of filling gas

Fig. 6.2 Schematic drawing of fuel assemblies in the KUCA A-core (Fig. 6.1) (Ref. [1])

Fig. 6.3 BTB fission chamber (Ref. [1])

(97% argon and 3% nitrogen). For measuring capture reaction rate ratio, a test foil ^{237}Np was set in the BTB fission chamber, and a reference foil gold-197 (^{197}Au) was attached to the Al sheath containing the chamber. The reference foil ^{197}Au was a normalization factor that has the capture reactions $^{197}\text{Au}(n, \gamma) ^{198}\text{Au}$, to obtain capture reaction rate ratio: $^{237}\text{Np}/^{197}\text{Au}$.

The BTB fission chamber was set at the special void element position in HEU-Pb zone (15, O; Fig. 6.4a), whereas in HEU-PE zone the position was (15, K; Fig. 6.4b). The hard and intermediate spectra are shown in Fig. 6.5. As shown in Fig. 6.4a, b, one and five normal fuel assemblies were added to the cores, respectively, when comparing with reference core shown in Fig. 6.1. Further fuel assemblies were added to reach criticality for two irradiation experiments at the critical state shown in Table 6.1. A series of critical irradiation experiments on ^{237}Np and ^{241}Am foils was conducted under the following core conditions: irradiation time 1 h; reactor power 3.5 W; neutron flux $(4.73 \pm 0.24) \times 10^7 \text{ s}^{-1} \text{ cm}^{-2}$.

6.1.1.3 ^{237}Np and ^{241}Am Foils

Two ^{237}Np and ^{241}Am thin test foils (both 99.99% purity, 20 mm diameter, and 4 nm thick), electro-deposited on Al backing (28 mm diameter and 0.2 mm thick), had an isotopic mass, respectively, equal to 89 and 15 μg as shown in Table 6.2. Thin ^{235}U reference foils (20 mm diameter and 0.6 nm thick, 5 or 10 μg weight), used to determine the fission reaction rate ratios, had a 99.91% enrichment. Test and reference ^{237}Np and ^{197}Au foils (8 mm diameter and 0.05 mm thick) were used to determine the capture reaction rate ratio. After the irradiation at critical state, ^{237}Np and ^{197}Au capture reaction rates were deduced by the saturated radioactivity obtained from the γ -ray spectra of irradiated test and reference foils, respectively, with the use of a high-purity germanium (HPGe) detector. The γ -ray energy generated by the capture reactions was as follows: 984.45, 1025.87, and 1028.53 keV in ^{237}Np ; 411.80 keV in ^{197}Au , as shown in Table 6.3.

6.1.2 Experimental Analyses

6.1.2.1 Experimental Analyses

During the irradiation experiments, the pulsed-height signals (voltage) induced by fission reactions were stored by the use of a multi-channel analyzer to discriminate the signals originated from fission products generated by fission reactions. In the HEU-Pb zone, pulsed-height distributions were clearly observed at two peaks of ^{237}Np (2.0 V and 5.0 V) and ^{235}U (1.6 V and 3.4 V) foils attributed to light and heavy fragments in fission products, as shown in Fig. 6.6, demonstrating the counts by their fission reactions. For ^{241}Am , one peak (0.1 V) was observed in the pulsed-height distribution shown in Fig. 6.7, and the counts by fission reactions were obtained by

(a) Location of BTB fission chamber (15, O) in HEU-Pb zone

(b) Location of BTB fission chamber (15, K) in HEU-PE zone

(c) Partial fuel rod "10" shown in Fig. 6.4(b)

Fig. 6.4 Core configuration of critical irradiation experiments on MA (Ref. [1])

Fig. 6.5 Calculated neutron spectra (MCNP6.1 with ENDF/B-VII.1) of HEU-PE (15, K) and HEU-Pb (15, O) fuel zones shown in Figs. 6.2a, b, respectively (Ref. [1])

Table 6.1 Positions [mm] of control and safety rods at critical state in HEU-Pb and HEU-PE cores in Figs. 6.4a, b, respectively (Ref. [1])

Measured fission reaction rate ratio	C1	C2	C3	S4	S5	S6
<i>HEU-Pb zone</i>						
$^{237}\text{Np}/^{235}\text{U}$ (10 μg)	723.27	1200.00	1200.00	1200.00	1200.00	1200.00
$^{241}\text{Am}/^{235}\text{U}$ (5 μg)	720.35	1200.00	1200.00	1200.00	1200.00	1200.00
<i>HEU-PE zone</i>						
$^{237}\text{Np}/^{235}\text{U}$ (5 μg)	698.23	1200.00	1200.00	1200.00	1200.00	1200.00
$^{241}\text{Am}/^{235}\text{U}$ (10 μg)	694.28	1200.00	1200.00	1200.00	1200.00	1200.00

1200.00 [mm]: Position of upper limit

Table 6.2 Number of atoms of ^{237}Np , ^{241}Am , and ^{235}U foils (Ref. [1])

Foil	Number of atoms
^{237}Np (HEU-Pb)	$(2.27 \pm 0.02) \times 10^{17}$
^{237}Np (HEU-PE)	$(1.48 \pm 0.02) \times 10^{17}$
^{241}Am (HEU-Pb)	$(3.62 \pm 0.01) \times 10^{15}$
^{241}Am (HEU-PE)	$(3.94 \pm 0.01) \times 10^{15}$
^{235}U (5 μg)	$(1.31 \pm 0.02) \times 10^{16}$
^{235}U (10 μg)	$(2.67 \pm 0.02) \times 10^{16}$

Table 6.3 Main characteristics of ^{237}Np and ^{197}Au capture reactions (Ref. [1])

Reaction	Half-life	γ -ray energy (keV)	Emission rate (%)
^{237}Np (n, γ) ^{238}Np	2.177 d	984.45	47.8
		1025.87	9.6
		1028.53	20.3
^{197}Au (n, γ) ^{198}Au	2.69517 d	411.80	95.5

Fig. 6.6 Measured pulsed heights of ^{237}Np and ^{235}U fission reaction rates in HEU-Pb zone (15, O; Fig. 6.4a) at critical state (Ref. [1])

Fig. 6.7 Measured pulsed heights of ^{241}Am and ^{235}U fission reaction rates in HEU-Pb zone (15, O; Fig. 6.4a) at critical state (Ref. [1])

making lower-level discrimination about 1.0 V and integrating the count per channel over 1.0 V. Also, the difference in pulsed heights between 5 and 10 μg of ^{235}U for ^{241}Am fission reaction rates was clearly observed by comparing the HEU-Pb and HEU-PE zones shown in Figs. 6.7 and 6.8, respectively.

In addition to the fission reaction rates, capture reaction rates were obtained by measuring the γ -ray of ^{237}Np foils in HEU-Pb and HEU-PE zones with the HPGe detector after the irradiation experiments, as shown in Figs. 6.9 and 6.10, respectively. For the ^{237}Np capture reaction rates, three peaks (Table 6.3) were found to be clearly in the γ -ray measurements of two irradiation experiments in both HEU-Pb and HEU-PE zones, and ^{237}Np capture reaction rates were experimentally deduced through the saturated radioactivity.

Fig. 6.8 Measured pulsed heights of ^{241}Am and ^{235}U fission reaction rates in HEU-PE zone (15, K; Fig. 6.4b) at critical state (Ref. [1])

Fig. 6.9 Measured γ -ray spectrum of ^{237}Np capture reaction rates in HEU-Pb zone (15, O; Fig. 6.4a) at critical state (Ref. [1])

Fig. 6.10 Measured γ -ray spectrum of ^{237}Np capture reaction rates in HEU-PE zone (15, K; Fig. 6.4b) at critical state (Ref. [1])

6.1.2.2 Numerical Analyses

Numerical analyses were carried out with the use of MCNP6.1 [2] together with ENDF/B-VII.1 [3]; analyses of ¹⁹⁷Au capture reaction rates were done with JENDL/D-99 [4]. The comparison between measured and calculated results, as shown in Table 6.4, shows a good agreement demonstrating a relative difference within 5% in the C/E (calculation/experiment) value for ²³⁷Np fission reaction rates ratio in the HEU-Pb core. For the ²⁴¹Am fission reaction rates ratio, the C/E discrepancy was about 10% at HEU-Pb and HEU-PE zones, demonstrating a difference between the neutron spectra of the two zones shown in Fig. 6.5. Here, the ²³⁷Np capture reaction rate ratio revealed notably good agreement between the experiments and the calculations, as shown in Table 6.5, with a relative difference of about 5% in C/E values.

From the results in Tables 6.4 and 6.5, MCNP6.1 simulations with ENDF/B-VII.1 demonstrated a fairly agreement with the experimental data of ²³⁷Np and ²⁴¹Am fission and capture reaction rates.

Table 6.4 Comparison of the results between experiments and calculations (MCNP6.1 with ENDF/B-VII.1) of fission reaction rate ratios at critical state (Ref. [1])

Fission reaction rate ratio	Calculation	Experiment	C/E
HEU-Pb zone			
²³⁷ Np/ ²³⁵ U (10 μg)	0.067 ± 0.001	0.071 ± 0.004	0.95 ± 0.06
²⁴¹ Am/ ²³⁵ U (5 μg)	0.079 ± 0.003	0.069 ± 0.001	1.14 ± 0.05
HEU-PE zone			
²³⁷ Np/ ²³⁵ U (5 μg)	–	–	–
²⁴¹ Am/ ²³⁵ U (10 μg)	0.036 ± 0.001	0.034 ± 0.001	1.08 ± 0.02

C/E: Calculation/Experiment

Table 6.5 Comparison of the results between experiments and calculations (MCNP6.1 with ENDF/B-VII.1) of capture reaction rate ratios at critical state (Ref. [1])

Capture reaction rate ratio	Calculation	Experiment	C/E
HEU-Pb zone			
²³⁷ Np/ ¹⁹⁷ Au	2.09 ± 0.10	2.15 ± 0.33	0.97 ± 0.16
HEU-PE zone			
²³⁷ Np/ ¹⁹⁷ Au	1.88 ± 0.08	2.02 ± 0.08	0.93 ± 0.05

C/E: Calculation/Experiment

6.1.3 Discussion

6.1.3.1 Fission Reaction Rate Ratio

The region-wise contribution of energy was examined by MCNP6.1 and ENDF/B-VII.1, for ^{237}Np and ^{241}Am fission reaction rates ratios to ^{235}U , with the use of reaction rates normalized by summarizing entire reaction rates over the whole energy. For the sake of comparison in Fig. 6.11, the energy distribution of ^{237}Np and ^{235}U fission reaction rates are shown for the critical irradiation in the HEU-Pb zone shown in Fig. 6.4a, demonstrating that ^{237}Np fission rate has high sensitivity to fission reactions around a few MeV region as well as in the neutron spectrum of the HEU-Pb zone shown in Fig. 6.5. Also, ^{235}U fission reactions were found to be dominant over the thermal and epi-thermal neutron regions for critical irradiation in the HEU-Pb zone, as shown in Fig. 6.11.

Fission reaction rates of ^{241}Am were acquired mainly in a few MeV region in HEU-Pb and HEU-PE zones shown in Figs. 6.12 and 6.13, respectively, although

Fig. 6.11 Region-wise contribution of energy of ^{237}Np and ^{235}U fission reaction rates by MCNP calculations in HEU-Pb zone at critical state (Ref. [1])

Fig. 6.12 Region-wise contribution of energy of ^{241}Am and ^{235}U fission reaction rates by MCNP calculations in HEU-Pb zone at critical state (Ref. [1])

Fig. 6.13 Region-wise contribution of energy of ^{241}Am and ^{235}U fission reaction rates by MCNP calculations in HEU-PE zone at critical state (Ref. [1])

striking peaks of ^{241}Am fission reactions were observed in a wide range of the thermal and epi-thermal neutron regions. Moreover, by comparing the results in Figs. 6.12 and 6.13, a difference in the reaction rates energy distribution between HEU-Pb and HEU-PE zones was clearly observed over the entire energy regions, demonstrating the important effect of the neutron spectrum variation on ^{241}Am fission reaction rates, as shown in Table 6.4.

Through numerical analyses of the region-wise contribution of energy to ^{237}Np and ^{241}Am fission reactions, the effect of neutron spectrum variation (Fig. 6.5) between the HEU-Pb and HEU-PE zones was soundly confirmed on the irradiation of ^{237}Np and ^{241}Am foils at a critical state.

6.1.3.2 Capture Reaction Rate Ratio

Capture reaction rates of ^{237}Np were successfully obtained by the measurement of γ -ray spectra in HEU-Pb and HEU-PE zones after critical irradiation, and used for the evaluation of capture reaction rate ratio by comparison of ^{197}Au capture reaction rates. From the calculated capture reaction rate results in Fig. 6.14, two main peaks can be noticed around the thermal neutron region, and ^{237}Np capture reaction rates were found to be highly sensitive to the thermal neutron spectrum field even in the HEU-Pb zone.

As shown in Table 6.5, the selection of ^{197}Au as reference foil was experimentally meaningful for the evaluation of the $^{237}\text{Np}/^{197}\text{Au}$ capture reaction rate ratio, even if no significant differences were observed between the results in the HEU-Pb and HEU-PE zones.

Fig. 6.14 Region-wise contribution of energy of ^{237}Np capture reaction rates by MCNP calculations in HEU-Pb zone at critical state (Ref. [1])

6.2 ADS Irradiation at Subcritical State

6.2.1 Experimental Settings

6.2.1.1 Core Configuration

Subcritical irradiation experiments [2] of MA by ADS were carried out in the solid-moderated and -reflected (A-core) at KUCA by the injection of high-energy protons onto lead-bismuth (Pb–Bi) target. As shown in Fig. 6.15, the ADS core consists of normal and partial fuel assemblies, and polyethylene moderators and reflectors. Normal fuel assembly “F” is composed of 60 unit cells, as shown in Fig. 6.2a. Numeral “12” (Fig. 6.16b) is a partial fuel assembly for reaching a critical mass, and represents the number of unit cells in a normal fuel assembly.

6.2.1.2 Proton Beams

High-energy neutrons generated by the interaction between high-energy protons and Pb–Bi target that set at the location of (15, H) shown in Fig. 6.15 were injected into the KUCA A-core. The main characteristics of proton beams were as follows: 100 MeV energy, 0.5 nA intensity (1 nA at most), 30 Hz pulsed frequency, and 100 ns beam width. The Pb–Bi target was 50 mm in diameter and 18 mm thick; the thickness was determined by taking into account the range and the full stoppage of 100 MeV protons inside the Pb–Bi target [6]. During the injection of 100 MeV protons (no vacuum; (15, A-H) in Fig. 6.15) onto the Pb–Bi target, the size of the proton beam spot was about 40 mm in diameter (circle line), as demonstrated by the scan of the Gafchromic film [7], at the front side (end of proton beams) of Pb–Bi target (15, H; Fig. 6.15), that is highly sensitive to the charged particles shown in Fig. 6.17. Furthermore, the neutron yield was $(1.33 \pm 0.04) \times 10^8 \text{ s}^{-1}$, as measured through $^{115}\text{In} (n, n') ^{115\text{m}}\text{In}$ reaction rates with the use of the saturated activity that

Fig. 6.15 Top view of the KUCA A-core for MA irradiation experiments by ADS (Ref. [5])

Fig. 6.16 Schematic drawing of “12” partial fuel rod (Fig. 6.15) (Ref. [5])

was deduced by the foil activation method [8] (over 0.3 MeV threshold energy; In foil; $10 \times 10 \times 1$ mm) obtained at the location of the Pb–Bi target.

6.2.1.3 Neutron Characteristics

For carrying out the ADS experiments, subcriticality state was made by inserting control rod C2 into the lower limit position (0.00 mm) and withdrawing other rods (C1, C3, S4, S5, and S6) fully (1200.00 mm) from the core. Subcriticality was then deduced experimentally by combining control rod C2 reactivity worth by the rod

Fig. 6.17 Scanning of Gafchromic film after proton irradiation at location of (15, H; Pb–Bi target) in Fig. 6.15 (Ref. [5])

drop method and control rod C2 calibration curve by the positive period method: about 225 ± 10 pcm as a reference value, as shown in Table 6.6. Additionally, the supplemental result obtained by the α -fitting method [9] was 215 ± 9 pcm. During the injection of high-energy neutrons into the core, the reactor power and the neutron flux were experimentally obtained by the foil activation method [8] with the use of two gold foils (bare and cadmium-covered) irradiated at the location between (15, M) and (15, O) in Fig. 6.15 as follows: 1.35 ± 0.07 W and $(1.82 \pm 0.09) \times 10^7$ s⁻¹cm⁻², respectively, for four hours of irradiation.

The neutron spectrum was numerically attained by the PHITS code [11], for the locations of the Pb–Bi target and the BTB fission chamber during the injection of high-energy neutrons, as shown in Fig. 6.18a, b, respectively. At the location of Pb–Bi target, high-energy neutrons showed a sharp peak around 2 MeV region and a unique distribution ranging between 10 and 100 MeV, as shown in Fig. 6.18a. In spite of small effect of high-energy neutrons over 10 MeV in ADS, as shown in Fig. 6.18b, no significant difference between the neutron spectra in ADS and critical cores was found at the location of the BTB fission chamber (15, O; Fig. 6.15).

Table 6.6 Measured core condition during injection of high-energy neutrons (Ref. [5])

Subcriticality [pcm] (Reference)	Subcriticality [pcm] (by α -fitting method)	Neutron flux (s ⁻¹ cm ⁻²)	Reactor power (W)
225 ± 10	215 ± 9	$(1.82 \pm 0.09) \times 10^7$	1.35 ± 0.07

$\beta_{eff} = 810 \pm 10$ [pcm] and $\Lambda = (3.30 \pm 0.01) \times 10^{-5}$ [s] by MCNP6.1 (Ref. [3]) with JENDL-4.0 (Ref. [10])

(a) Pb-Bi target at location of (15, H)

(b) BTB chamber at location of (15, O; Fig. 6.15) in ADS and critical cores

Fig. 6.18 Calculated neutron spectra (PHITS) at locations of (15, H) and (15, O) in Fig. 6.15 (Ref. [5])

6.2.1.4 ^{237}Np and ^{241}Am Foils

To measure fission and capture reactions of ^{237}Np and ^{241}Am (both 99.99% purity, 89 μg and 15 μg weight, respectively), the BTB fission chamber was used in subcritical irradiation experiments and set at the location of (15, O; Fig. 6.15) of a special void element. The main function of the BTB chamber is to obtain simultaneously two signals from specially installed test (^{237}Np or ^{241}Am) and reference (^{235}U ; 99.91% enrichment, 10 μg weight) foils. For fission reaction rates, the two main signals from the test and reference foils come from electric ones of fission fragments caused by

fission events. Capture reaction rates of ^{237}Np and ^{197}Au were deduced by the saturated radioactivity [8] obtained from two signals of the γ -ray emission of the ^{237}Np test foil in the BTB chamber and the ^{197}Au reference one that set at the location of (15, O) of the Al sheath (special void element), with the use of a germanium detector. From the two signals of test and reference foils, the fission reaction rate ratio was experimentally acquired by $^{237}\text{Np}/^{235}\text{U}$ or $^{241}\text{Am}/^{235}\text{U}$, and the capture one by $^{237}\text{Np}/^{197}\text{Au}$, and finally, validated by comparison with previous experimental results [1] at a critical state.

6.2.2 Demonstration of Nuclear Transmutation

Fission reaction rates were experimentally obtained by the pulsed-height distributions (voltage) of ^{237}Np and ^{241}Am fission events, as shown in Figs. 6.19a, b, respectively, demonstrating original electric signals of test and reference foils. As shown in Fig. 6.19a, the fission events of two ^{237}Np and ^{235}U foils were clearly observed over entire pulsed heights. Moreover, for ^{241}Am and ^{235}U shown in Fig. 6.19b, discrimination between the signals of fission fragments and α -ray induced by ^{241}Am was made at 1.2 V, determining the small fission events by ^{241}Am over 1.2 V and the same fission events by ^{235}U over the entire pulsed heights, as confirmed in the critical irradiation [1]. Meanwhile, the difference between pulsed-height distributions of ^{235}U shown in Figs. 6.19a, b was mainly attributable to reproducibility of the gain in the BTB fission chamber when changing the ^{235}U sample installed. The fission reaction rate ratio determined by the experimental results was found to be a notable 0.048 ± 0.001 and 0.035 ± 0.003 for $^{237}\text{Np}/^{235}\text{U}$ and $^{241}\text{Am}/^{235}\text{U}$, respectively, the same as at critical irradiation shown in Table 6.7, although no data of $^{237}\text{Np}/^{235}\text{U}$ were obtained at critical irradiation due to invalid data acquisition of two electric signals (^{237}Np and ^{235}U) actuated in the BTB fission chamber.

The capture reaction rate ratio was deduced by the saturated radioactivity on the basis of the γ -ray emission of ^{237}Np and ^{197}Au capture reactions (Fig. 6.20 and Table 6.8) was found to be a remarkable 1.88 ± 0.28 , almost the same as at critical irradiation shown in Table 6.9.

From the results of fission and capture reaction rate ratios shown in Tables 6.7 and 6.9, respectively, fission and capture reaction events by ADS were successfully confirmed, and as a conclusion, nuclear transmutation of ^{237}Np and ^{241}Am by ADS was experimentally achieved and demonstrated in the KUCA core.

6.3 Conclusion

The integral experiments on irradiation of ^{237}Np and ^{241}Am in cores at critical conditions were carried out in the KUCA A-core with a neutron hard spectrum. The fission reaction rate ratios ($^{237}\text{Np}/^{235}\text{U}$ and $^{241}\text{Am}/^{235}\text{U}$) and the capture reaction rate ratio

Fig. 6.19 Measured pulsed heights of ^{237}Np , ^{241}Am , and ^{235}U fission reaction rates at subcritical state (Ref. [5])

Table 6.7 Comparison of the results of fission reaction rate ratio at subcritical and critical states (Ref. [5])

Reaction rate ratio	Subcritical state	Critical state (Ref. [1])
$^{237}\text{Np}/^{235}\text{U}$	0.048 ± 0.001	—
$^{241}\text{Am}/^{235}\text{U}$	0.035 ± 0.003	0.034 ± 0.001

($^{237}\text{Np}/^{197}\text{Au}$) were measured by a BTB (back-to-back) fission chamber. To investigate the behavior of ^{237}Np and ^{241}Am fission and capture reaction rates in the KUCA A-core, results were deeply analyzed by both the experimental signals from the BTB fission chamber and the numerical results of MCNP6.1 calculations with ENDF/B-VII.1. The calculated decomposition in energy of the fission and capture reaction rates allowed us to isolate the most important energy regions in the responses. From the results of experimental and numerical analyses, integral experiments on the irradiation of MA in cores at critical conditions were successfully carried out

Fig. 6.20 Measured γ -ray spectrum of ^{237}Np capture reaction rates at subcritical state (Ref. [5])

Table 6.8 Main characteristics of ^{237}Np and ^{197}Au capture reactions (Ref. [5])

Reaction	Half-life	γ -ray energy (keV)	Emission rate (%)
$^{237}\text{Np} (n, \gamma) ^{238}\text{Np}$	2.177 d	984.45	27.8
		1025.87	9.6
		1028.53	20.3
$^{197}\text{Au} (n, \gamma) ^{198}\text{Au}$	2.69517 d	411.80	95.5

Table 6.9 Comparison of the results of capture reaction rate ratio at subcritical and critical states (Ref. [5])

Reaction rate ratio	Subcritical state	Critical state (Ref. [1])
$^{237}\text{Np}/^{197}\text{Au}$	1.88 ± 0.28	1.88 ± 0.08

in the KUCA-A hard spectrum cores. In future studies, nuclear transmutation of MA by ADS is foreseen for implementation at KUCA, with the combined use of a hard spectrum core and 100 MeV protons, on the basis of measured and calculated methodologies obtained in the current at critical core conditions irradiation experiments on MA.

100 MeV proton beams were injected onto the Pb–Bi target, and subcritical irradiation experiments of MA (^{237}Np and ^{241}Am) by ADS were carried out with the use of high-energy neutrons generated by the interaction of 100 MeV protons and the Pb–Bi target. In the subcritical irradiation experiments, fission reaction rates of ^{237}Np and ^{241}Am were acquired by the electric signals of fission fragments obtained from the BTB fission chamber, and capture reaction rates of ^{237}Np were obtained by the measurement of the γ -ray spectrum after the irradiation. Here, for the first time, nuclear transmutation of ^{237}Np and ^{241}Am was soundly implemented by ADS that comprises a subcritical core and a 100 MeV proton accelerator with Pb–Bi target, and demonstrated at KUCA through the experimental results of the reaction rate ratio

obtained by combining the test (fission: ^{237}Np and ^{241}Am ; capture: ^{237}Np) and the reference (fission: ^{235}U ; capture: ^{197}Au) foils.

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