# MEASUREMENT OF FISSION AND CAPTURE CROSS SECTIONS FOR DESIGN OF TRANSMUTATION SYSTEMS

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## ABSTRACT

The fission cross section for $^{237}$Np from about 1 eV to about 5 keV relative to that for $^{235}$U was measured with back-to-back type double fission chambers using an electron linear accelerator (linac) driven lead slowing-down neutron spectrometer (lead spectrometer) . Although the shape of the present energy dependent cross section agrees with those of JENDL-3 (below 120 eV) and ENDF/B-VI, their absolute values are from 3 to 4 times smaller than the present data.

The thermal neutron capture and resonance neutron capture integral for $^{237}$Np were measured by the activation method in a standard thermal neutron field and in a standard l/E neutron field, respectively. The present result for the former, 154.7± 2.9 b, is about 17% smaller than those in JENDL–3 and ENDF/B–VI, but the preliminary data for the latter, 646 b reasonably agrees with those in both files.

## INTRODUCTION

In order to design any transmutation systems of long half–lived waste nuclides to much shorter ones, precise nuclear data are required. Although modern evaluated nuclear data files such as JENDL-3 and ENDF/B-VI contain most of the long half-lived waste nuclides, both higher actinoids and fission products, the precision and reliability of their nuclear data are often insufficient for designing their transmutation systems. Therefore, it is keenly requested to carry out more experimental works to obtain more precise and reliable nuclear data for these nuclides.

Among several higher actinoids or minor transuranium nuclides produced by power reactors, $^{237}$Np is thought to be one of the most burdensome ones, because of its large production rate in a reactor, very long half–life ($2.14\times10^{6}$y) and alpha activities in its decay chain. In order to transmute $^{237}$Np to nuclides with much shorter half-lives, it is proposed to use its fission and capture reactions, $^{237}$Np(n, f) and $^{237}$Np(n, $\gamma$)$^{238}$Np. We started to measure these reactions as precise as possible by making use of a few standard neutron fields which we had developed and characterized for long time.

## FISSION CROSS SECTION MEASUREMENT FOR Np-237

Several groups measured neutron cross sections in the intermediate and resonance regions with lead spectrometers with a conventional pulsed neutron source'[5]. More recently, Slovacek, et al. utilized an electron linac for a lead spectrometer and succeeded in measuring very small cross sections[6] such as the subthreshold fission of $^{238}$U and the cross sections for higher actinoids of which sample mass was extremely small[7].

We installed a lead spectrometer beside an electron linac of the Research Reactor Institute, Kyoto University and named it Kyoto University Lead Spectrometer (KULS). The size and the weight of KULS are 1.5m cubic and about 38 tons, respectively. The purity of the lead blocks composing KULS is 99.9%. We covered KULS by cadmium sheets 0.5mm thick and put it on a movable steel table. An air–cooled tantalum photoneutron targets was placed at the center of KULS. We made 11 experimental

holes in it. One of these holes was covered with a bismuth layer 10-15cm thick to suppress the high energy gamma rays from the neutron capture of lead[8]. The cross sectional view of KULS is shown in Figure 1.

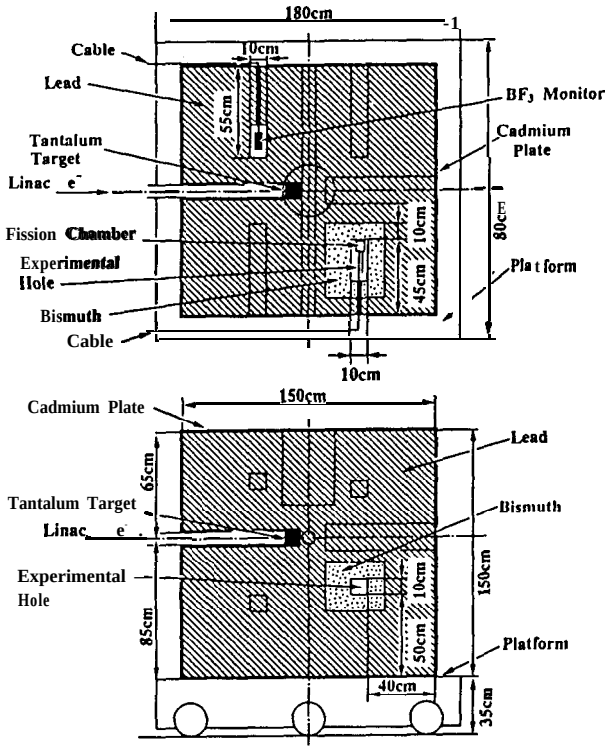



Figure 1 Cross sectional view of Kyoto University Lead Spectrometer, KULS.

The characteristics of KULS were obtained with a $BF_3$ counter and an argon-filled proportional counter with and without several resonance filters. The energy resolution at 27.5 eV shows 39±1%. The details of the characterization of KULS is given elsewhere[10].

The fission chambers in this work have two identical parallel type ionization chambers as shown in Figure 2. The chambers were made of aluminum and filled with a mixed gas of 97% argon and 3% nitrogen at the pressure of 1 atm. The distance between two electrodes was 8mm and the applied voltage was 400V.

Neptunium oxide $NpO_2$ of about 2mg was dissolved in hydrochloric acid of 0.2N and 0.1 ml of this solution was mixed into 2-propanol of 5 ml. Therewith we electrodeposited $NpO_2$ on a stainless steel plate 28mm in diameter and 0.2 mm thick. The diameter of the deposit was 2cm. Highly enriched (99.91%) uranium dioxide $UO_2$ was similarly electrodeposited on the stainless steel plate. After electrodeposition, each plate was sintered with a gas burner. The numbers of atoms in the sample deposit ($^{237}Np$) and in the reference one ($^{235}U$) were determined by the alpha ray spectrometry with a silicon surface barrier detector in vacuum, and are $(1.99\pm0.02)\pm10^{17}$ and $(4.12\pm0.09)\times10^{17}$, respectively. In the sample deposit, we found $^{238}Pu$ and $^{239}Pu$ to be 0.320±0.003ppm and 1.10±0.13ppm, respectively. The alpha rays from $^{234}U$ was counted for the reference deposit, and the contents of $^{234}U$ became 464±5ppm.

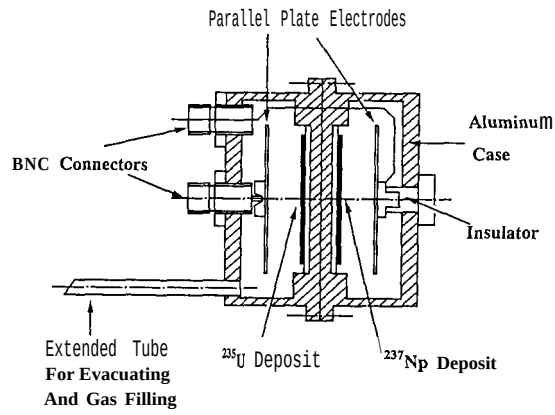



Figure 2 Cross sectional view of the back-to-back type fission chambers.

Two identical electronic circuits were prepared for the slowing-down time analysis. The start signal for timing was takn from the electron linac. Since we used quite thin sample and reference deposits, fission pulses were clearly discriminated from background ones.

Measurement of the fission ratio of $^{237}Np$ to $^{235}U$ was carried out in the bismuth-covered experimental hole. Background run was done with the chambers without sample and reference deposits, and we observed few background counts. Details about the corrections and the error estimation are given elsewhere[11].

In Figure 3, the present result is compared with the evaluated values in JENDL-3[12] and ENDF/B-VI[13]. Since the energy resolution of their data is much higher than that of the present experiment, we processed the eveluated data values by multiplying a resolution function of a Gaussian with 40% of its full width at half maximum. In this figure, shown are the processed values of both files. From this figure, it can be seen that (1) the evalu-

ated values in both JENDL-3 and ENDF/B-VI are from 3 to 4 times smaller than the present result. (2) The gross shape of the present result is very similar to that of ENDF/B-VI in all range and to that of JENDL-3 below 120eV. Above 120eV, the cross section in JENDL-3 is flat and differs from the present result due to the neglection of resonance treatment in the evaluation.

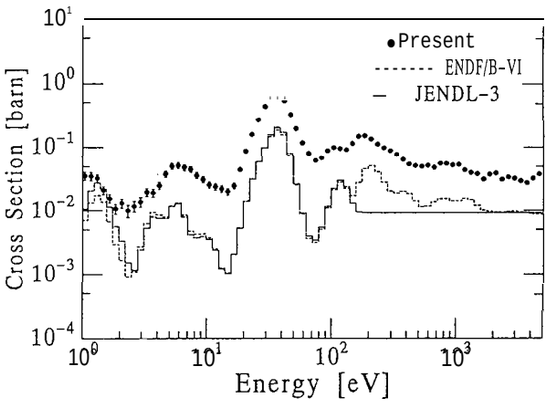



Figure 3 Experimentally obtained cross section of the $^{237}$Np(n, f) reaction compared with those in evaluated data files.

Reevaluation of the $^{237}$Np(n, f) cross section in JENDL-3 and ENDF/B-VI in the intermediate and resonance regions is recommended. If this cross section increases factor 3, the efficiency of the transmutation of $^{237}$Np bythe fission 'n these regions would improve almost the same factor.

Obviously the effectiveness of the fission in the fast region is superior to that in the intermediate and resonance regions for the transmutation of $^{237}$Np, however precise data of its fission cross section in these regions and in the thermal region is required to design any trasmutation systems of $^{237}$Np. We are now trying to measure the thermal fission cross section for $^{237}$Np with the same chambers and the standard thermal neutron field described below.

## THERMAL NEUTRON CAPTURE CROSS SECTION MEASUREMENT FOR NP-237

The thermal neutron cross section for the $^{237}$Np(n,$\gamma$) $^{238}$Np reaction was measured by the activation method in a standard thermal neutron field at the heavy water thermal neutron facility of Kyoto University Reactor (KUR). The configuration of this facility is shown in Figure 4. The thermal neutron spectrum at this facility was measured with a chopper and was shown to have a pure Maxwellian shape[14]. This facility is regarded as one of the best standard thermal neutron spectrum fields in the world.

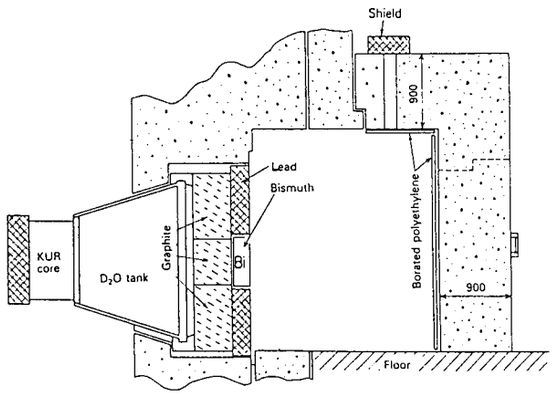



Figure 4 Standard thermal neutron filed at the heavy water facility of Kyoto University Reactor, KUR.

The neptunium samples were prepared by that we dropped a droplet of the solution of $^{237}NpO_2$ in hydrochloric acid on a filter paper and dried it. The gamma rays from $^{233}$Pa and $^{238}$Np in an irradiated sample were simultaneously measured with a calibrated HPGe detector. Since $^{233}$Pa is in radioactive equilibrium to $^{237}$Np, the mass of $^{237}$Np in a sample can be obtained from the radioactivity of $^{233}$Pa.

Thermal neutron flux was monitored with a gold foil of 3mm in diameter and 50μm thick and with a wire segment of aluminum-gold alloy (gold: 0.0314% in weight) 0.5mm in diameter. The gamma rays from $^{198}$Au were measured with the HPGe detector. As the cross section for the $^{197}$Au(n, 7 )$^{198}$Au reaction at 2200m/s of neutron velocity, we referred the data, 98.65±0.09b, from the Mughabghab's book[15].

The experiment was independently repeated six times for nine samples and the results are summarized in Table 1. The present value is about 17% smaller than those in JENDL-3 and ENDF/B-VI. Reevaluation of the thermal neutron cross section for the $^{237}$Np(n, 7 ) reaction is recommended.

Table 1 Thermal neutron capture cross section for $^{237}Np$, $\sigma_\gamma$(2200m/s)

| | |
|---|---|
| Present value | 154.7±2.9 b |
| JENDL-3 | 181.0 |
| ENDF/B-VI | 181.0 |
| Mughabghab | 175.9±2.9 |

## RESONANCE NEUTRON CAPTURE INTEGRAL MEASUREMENT FOR Np-237

The resonance integral for the $^{237}Np$ (n, $\gamma$) $^{238}Np$ reaction was measured by the activation method in a standard l/E neutron field at the central cavity in the internal graphite reflector between the two divided cores of Kinki University Reactor (UTR-KINKI). The configuration of this reactor is illustrated in Figure 5. By unfolding multi-foil activation data and by the sand-with foil method, we demonstrated that the neutron spectrum at the central cavity of UTR-KINKI was close to l/E from 1eV to a few keV[16], as seen in Figure 6. By making use of this position, we have measured resonance integrals for several reactions so far[17].

The preparation of the neptunium samples and the method of the gamma ray measurement are as same as to those in the thermal cross section measurement. A gold foil of 12.7mm in deameter and 50pm thick was covered by a cadmium filter 0.5mm thick and was irradiated with the neptunium sample in the centrl cavity of UTR-KINKI. The correction factors for the self-shielding of the neutron resonance in the gold foil was derived by the Monte Carlo code VIM[18]. As the resonance integral for the $^{197}Au$(n, $\gamma$) $^{198}Au$ reaction, we referred the data

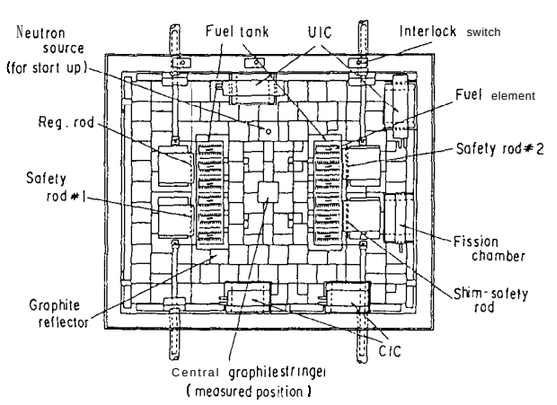
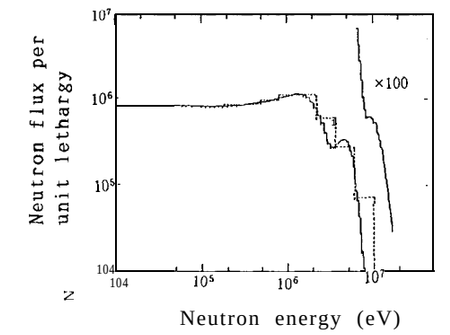



Figure 5 Standard l/E neutron field in Kinki University Reactor UTR-KINKI.



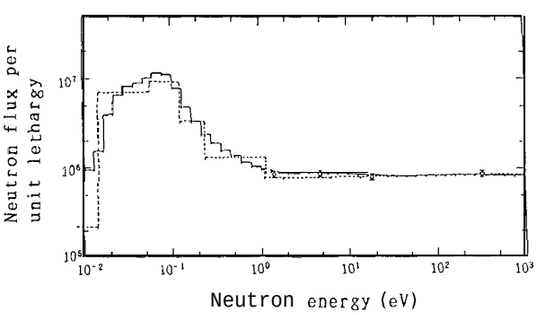




Figure 6 Measured and calculated neutron spectrum at the standard l/E neutron field in UTR-KINKI.
○ □ △ ▽ : Measured by the sandwich foil method.
: Measured by the multi-foil unfolding method.
Calculated by SRAC.

1550±28 b from the Mughabghab's book[15].

The present result, which is not the final data yet, is shown in Table 2. The error is estimated about 60 b. The present value is close to that given in the Mughabghab's book and agrees with those in JENDL-3 and ENDF/B-VI in the estimated error.

Table 2 Resonance neutron capture integral for $^{237}Np$

| | |
|---|---|
| Present value | 646±55 |
| JENDL-3 | 663 |
| ENDF/B-VI | 655 |
| Mughabghab | 640±50 |

## CONCLUSION

Three important cross sections for $^{237}Np$, the fission cross section in the intermediate and resonance region, the

thermal neutron capture cross section and the resonance neutron capture integral, were measured by making use of three standard or reference neutron fields. The absolute values of the fission cross section are larger by from 3 to 4 times larger than the evaluated data in JENDL-3 and ENDF/B-VI. The thermal neutron capture cross section became 154.7±2.9 b and is about 17% smaller than those in both files. The preliminary data for the resonance neutron capture integral, 646 b reasonably agrees with those in both files.

We are now trying to measure other cross sections for higher actinoids or minor transuranium nuclides as precise as possible by making use of standard or reference neutron fields and rather classical detectors, to supply more reliable nuclear data for designing transmutation systems.

ACKNOWLEDGEMENT

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