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MEASUREMENT OF FAST NEUTRON INDUCED FISSION CROSS SECTION RATIOS OF ^{239}Pu AND ^{242}Pu RELATIVE TO ^{235}U

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Abstract: Fission cross section ratios of Pu-239 and Pu-242 relative to U-235 were measured in the energy range from 0.6 to 7 MeV using the 4.5 MV Dynamitron accelerator of Tohoku University. A fast timing back to back fission chamber was used to detect fission events. The measurement was carried out with time of flight (TOF) method. The corrections to the measured data were carefully applied and uncertainty was analyzed taking the correlation between error sources into account. The overall uncertainty of the present results was about 2 %. The present results for Pu-239/U-235 are slightly higher than other experimental data and JENDL-2. For Pu-242/U-235, the present data agree with those of Meadows and Kuprijanov et al.

(fission cross section, fast neutron, Pu-239, Pu-242, U-235, time of flight, back to back fission chamber)

Introduction

Fission cross sections induced by fast neutrons have primary importance in the design of fast reactors. Therefore, the accuracy required by reactor designers is as high as 2 %. However, except for Pu-239, the measured data are rather scarce. For instance, for Pu-242, only 7 data have been reported since 1970. In the case of Pu-239, although quite a few data have been reported, there are differences of more than 10 % among the measurements in the MeV energy range. This situation is reflected in the evaluation of nuclear data file. For instance, the evaluated data of Pu-242 from 2 to 7 MeV differ more than 10 % between ENDF/B-IV /1/ and JENDL-2 /2/ . Considering the situation mentioned above, we have measured fission cross sections of Pu-239 and Pu-242 relative to U-235 as a part of the measurement of fast neutron induced fission cross section of actinide elements using the Dynamitron accelerator at Fast Neutron Laboratory of Tohoku University. The measured energy range was from 0.6 to 7 MeV. Pulsed neutron mode and TOF method was used to improve S/N ratio and to clarify the background. A back to back parallel plate ionization chamber, which has better time resolution and is lighter in weight than the chamber previously used /3/, was employed to detect fission events.

Experiment

Fission samples were electroplated on the platinum plate of 0.3 mm thick with 36 mm in diameter and sintered into oxide to fix to the plate. The diameter of the deposit was 25 mm with thickness of 10 to 90 $\mu g/cm^2$. The numbers of atoms in the samples were determined by the low geometry alpha spectroscopy using a Si semiconductor detector. However, when the alpha activity of the isotope was too weak or the energies of alpha particles were too close to be separated with the spectrometer, the results of mass spectroscopy carried out at Japan Atomic Energy Research Institute (JAERI) or Oak Ridge National Laboratory (ORNL) were utilized. The results are summarized in Table 1 /4/.

The construction of the fission chamber is shown in Fig.1. The chamber is cylindrical and made of stainless steel with 44 mm in diameter, 32 mm in height and 1 mm inner wall thickness. However, the window from which neutrons enter and the electrode are 0.1mm thick to reduce the neutron scattering. Also coaxial cables are directly connected to the fission chamber. By these measures, scattered neutrons by the chamber materials were calculated to be reduced about a half of those by the previously used chamber.

To use in TOF mode, the time resolution of the fission chamber should be good, and this is realized by reducing the distance between the

Tablel Isotopic composition of fission sample

Sample	Areal Density / ($\mu g/cm^2$)	Isotopic Composition / %		Number of Atoms / 1×10^{17}
U-235	90.51	U-233	0.000005	0.0000005
		U-234	0.03	0.003
		U-235	99.91	10.01±0.01
		U-236	0.02	0.002
		U-238	0.04	0.004
Pu-239	9.801	U-235	0.02	0.0002
		U-236	0.002	0.00002
		Pu-238	0.009	0.00002
		Pu-239	96.74	1.034±0.005
		Pu-240	3.16	0.034
		Pu-241	0.06	0.0006
		Pu-242	0.003	0.00003
		Am-241	0.02	0.0002
Pu-242	10.46	Pu-238	0.003	0.00003
		Pu-239	0.02	0.0002
		Pu-240	0.08	0.0009
		Pu-241	0.04	0.0005
		Pu-242	99.84	1.126±0.009
		Am-241	0.02	0.0002

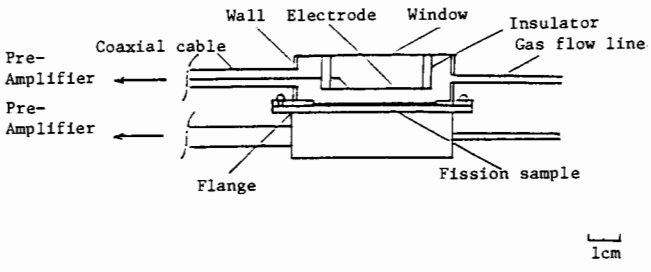

Fig.1 Construction of fission chamber

electrode and the fission sample. On the other hand, good separation between the pulses due to alpha particles and those due to fission fragments is obtained with wider spacing between the electrode and the fission sample. Based on the experiment of time resolution and energy resolution with changing the spacing, the spacing was determined to be 6 mm. In this case, the time resolution was about 7 to 10 nsec which was 10 times better than the chamber with 20 mm spacing. At the same time, the valley between the alpha pulses and the fission fragment pulses was 2.5 times wider than the chamber with the electrode spacing of 4mm. The ionization gas was continuously flowed during the experiment. Since the thickness of Pu sample was thin, the chamber was placed at the distance of about 3 cm from the neutron producing target. A LiF target evapolated on a copper or platinum plate was used to produce 0.6 to 1.8 MeV neutrons via $^{7}Li(p,n)^{3}He$ reaction. A T-Ti target with copper backing was used to produce 0.9 to 2.2 MeV neutrons via $T(p,n)^{3}He$ reaction. A D_2-gas target with copper gas cell was used in producing 4.0 to 6.9 MeV neutrons via $D(d,n)^{3}He$ reaction.

Prior to each fission rate measurement, the neutron energy spectrum was measured with TOF method. This measurement was composed of two parts; one was to determine the precise neutron energy emitted to 0°; the other was to check the existence of parasitic neutrons such as produced by Cu(p,n) reaction with copper backing. A NE213 scintillation counter of 2"ϕ x2" was placed on the axis of incident beam line 8m apart from the target. The detector was placed in a concrete shield structure and a cylindrical water tank was placed in front of the concrete to minimize the effect of room-returned neutrons and γ-background. The repetition rate of the pulsed beam was 0.5 or 1 MHz according to the flight time of source neutrons. A BF_3 monitor was placed at the position of 6 m from the target and 90° from the beam line. It was surrounded by boron loaded paraffin with cadmium cover.

Since the fission chamber was placed about 3 cm from the target, the neutron spectrum has larger energy spread and the average neutron energy is lower than the 0° neutrons measured by the NE213 counter. Therefore, the incident neutron energy and its spread at the position of the fission chamber were calculated by a Monte Carlo code prepared for this purpose based on the neutron spectrum and its spread measured with the NE213 counter. The appropriateness of the code was checked by comparing the calculated neutron spectrum and its spread of 0° neutrons with the measurement. Although the calculated energy spread was a little larger than the measured one, it was found that the neutron energy was very accurately calculated.

The fission rate measurement was composed of two runs for each energy point. One was the case where the standard fission sample was faced to the target, the other was the case where the sample whose cross section was to be measured was faced to the target by rotating the fission chamber 180 at the same position. In this experiment, TOF method was used in the fission rate measurement. While the foreground signal is generated during the neutron pulse passes through the chamber, the backgrounds due to alpha particles, spontaneous fissions and room-returned neutrons are time independent. Therefore, it is easy to separate the foreground component and the background component. The signals of TOF and fission fragment energy were accumulated on the two parameter pulse height analyzer.

When the D_2-gas target was used, the fission rate measurement with H_2-gas in stead of D_2-gas was also carried out. Since H_2-gas has the same stopping power as D_2-gas and it dose not generate neutrons, the parasitic neutrons generated in the target assembly can be estimated by this measurement. The current passing through the havar foil in front of the target assembly was used for normalization. The H_2-gas pressure was kept as same as D_2-gas pressure.

The counting statistics of fission event was 7000 to 15000 counts including background component in each run. It took from 30 minutes to 8 hours.

Data Analyses and Corrections

The fission cross section ratio was obtained by Eq. (1)

$$R_\sigma(E_0) = \frac{\sigma_{x,0}(E_0)}{\sigma_{s,0}(E_0)} = \frac{N_{s,0}}{N_{x,0}} R(E_0)\cdot F(E_0), \qquad (1)$$

where

$$\begin{aligned} F(E_0) = 1 &+ \frac{1}{N_{s,0}\,\sigma_{s,0}(E_0)}\Big[\sum_{i=1}^{n} N_{s,i}\sigma_{s,i}(E_0) \\ &+ \frac{1}{\phi(E_0)}\sum_{g=1}^{l}\phi(E_g)\sum_{i=0}^{n} N_{s,i}\sigma(E_g)\Big] \\ &- \frac{1}{R(E_0)N_{s,0}\sigma_{s,0}(E_0)}\Big[\sum_{j=1}^{m} N_{x,j}\sigma_{x,j}(E_0) \\ &+ \frac{1}{\phi(E_0)}\sum_{g=1}^{l}\phi(E_g)\sum_{j=0}^{m} N_{x,j}\sigma_{x,j}(E_g)\Big] \end{aligned} \qquad (2)$$

The meaning of each notation is as follows;

x : sample to be measured
s : standard sample
i : nuclide existing in the sample foil
  (i=0 : nuclide to be measured
    n : number of nuclides)
j : nuclide existing in the standard foil
  (j=0 : standard nuclide
    m : number of nuclides)
g : number of energy groups of incident neutrons
  (g=0 : source neutron energy
    l : number of energy groups)
R : fission cross section ratio to be ultimately obtained
  : fission cross section
N : number of atoms
  : incident neutron flux.

The second term of right-hand side of Eq.(2) is the correction for fissions due to impurity nuclides in the standard foil and the fourth term is that in the sample foil to be measured. The third term is the correction for fissions in the standard foil due to parasitic and scattered neutrons in the flux and the fifth term is that in the sample foil to be measured.

In the following, the correction terms and the method of corrections will be explained.

(1) Correction for fissions due to room returned neutrons and spontaneous fission: this was carried out by subtracting time independent com-

ponent from time dependent part of TOF spectrum.
(2) Extrapolation of fission counts obscured by the alpha pulses: since the tail of the pulse height spectrum of fission fragment showed almost flat distribution, we extrapolated the average counts per channel to zero pulse height.
(3) Self-absorption of fission fragments in the sample foil: this was estimated by the formula given by Carlson /5/. In applying this formula, the angular distribution of fission fragments were assumed to be isotropic, since the data were not available for the distribution. The thickness of the sample foil was obtained by assuming the sample was dioxide. The mean free path of the fission fragment was obtained by the empirical formula of Kahn et al. /6/. The kinetic energy of the fission fragment was taken as 88.5MeV /7/.
(4) Anisotropy of emitted angle of source neutrons and inhomogeneity of the thickness of sample foil: they were corrected by a Monte Carlo code mentioned in the section of Experiment. The appropriateness of the code was verified by measuring the fission cross section ratio between two U-235 foils with different inhomogeneity.
(5) Fissions due to impurity nuclides in the fission sample: this effect was calculated by the second and the fourth term of Eq.(2). The fission cross sections used were those in JENDL-2.
(6) Fissions by the parasitic neutrons in the incident neutrons: for Ti-T and LiF target, this effect was corrected by the Monte Carlo code mentioned above. For D_2-gas target, the result of the measurement with H_2-gas target was used.
(7) Fissions due to construction materials of the target assembly and the fission chamber: this effect was corrected by the Monte Carlo code which calculates the effect of multiple scattered neutron energy spectrum /8/. The used cross sections were those in ENDF/B-IV /1/ or ENDL-84 /9/. The appropriateness of the code was verified by comparing the measured and the calculated neutron spectrum.
(8) The attenuation of incident neutron flux due to platinum backing:
(9) The difference in solid angle between the forward and backward sample from the neutron target:
(10) The anisotropy of emission angle of fission fragments:

by taking the geometric mean of the two runs carried out at each energy point, the corrections in item (8) and (9) were canceled and the correction in item (10) was approximately canceled /10/. The corrections are shown in Table 2.

The errors were estimated by considering the following error sources.
(1) The number of atoms of U-235 and the nuclide to be measured ($N_{s,0}$,$N_{x,0}$): for these quantities the following items were considered.
i). absolute counting rate of alpha particles,
ii). relative alpha decay rate,
iii). half lives of alpha decay,
iv). detection efficiency of the alpha counter.
(2) Fission rate ratio ($R(E_0)$): for this quantity, the following items were considered.
i). statistical error of fission counts by source neutrons,
ii). statistical error of fission counts by room returned neutrons and spontaneous fissions,
iii). statistical error of fissions by parasitic neutrons (in the case of D_2-gas target)
iv). error in the estimated counts by the extrapolation to zero pulse height,
v). error in the estimated counts by the self-absorbed fission fragments,
vi). error in the corrections associated with angular dependence of source neutrons and the inhomogeneity of the thickness of the fission sample.
(3) Correction coefficient ($F(E_0)$): for this quantity, the following items were considered.
i). the number of atoms of all the impurity nuclides contained in the sample,
ii). fission ratio,
iii). fission cross sections of impurity nuclides in the sample,
iv). parasitic neutron flux (in the case of LiF and Ti-T target),
v). scattered neutron flux by the construction materials of the target assembly and the fission chamber.

In estimating the error, the correlation between the error sources were considered whenever applicable and covariances were derived. The summary of the estimated errors are shown in Table 3.

Table2 Corrections applied in obtaing fission ratios

Sample	Correction / %											
	Extrapolation	Self-absorption		Inhomogeneousness		Background	Parasitic Neutron			Scattered Neutron		Impurity
		Fore	Rear	Fore	Rear		LiF	T-Ti	D_2-Gas	Chamber	Target	
U-235C	0.1~1.6	0.9~1.9	0.0~0.2	1.1~5.2	1.2~5.1	0.1~1.0	0.4~16	0.0~1.6	0.0~9.1	0.8~1.8	0.7~3.3	0.0~0.1
Pu-239	0.2~0.8	0.5~1.3	0.0	0.3~1.1	0.4~1.0	0.1~0.7	1.4~11	0.1~1.5	0.0~8.8	0.8~1.7	0.7~1.0	2.0~2.9
Pu-242	0.2~0.8	0.4~1.3	0.0	0.1~0.4	0.1~0.4	1.4~9.9	0.0~12	0.0~0.6	0.0~6.2	0.8~1.7	0.6~1.6	0.2~0.4

Table3 Sources of uncertainty

Sample	Error / %			
	Number of Atoms	Fission Reaction Rate Ratio	Correction Factor	Result
U-235	1.1	-	-	-
Pu-239	0.5	1.1 ~ 2.3	0.1 ~ 0.6	1.7 ~ 2.7
Pu-242	0.8	1.0 ~ 2.3	0.0 ~ 2.1	1.8 ~ 2.7

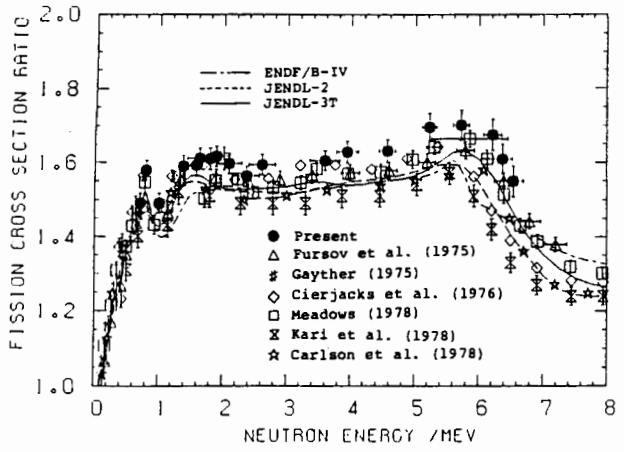

Fig.2 Fission cross section ratio of $^{239}Pu/^{235}U$

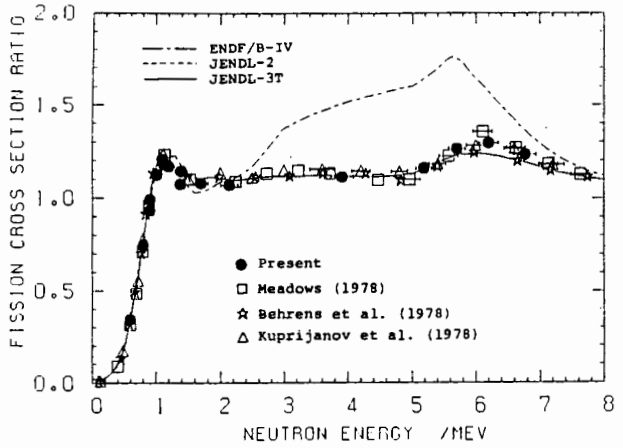

Fig.3 Fission cross section ratio of $^{242}Pu/^{235}U$

Result and Discussion

The results of fission ratio of Pu-239 and Pu-242 relative to U-235 are shown in Fig.2 and 3, respectively. In the figures, the present results are compared with the several reported data and the evaluations. For Pu-239, error is less than 2 % below 5 MeV, but above that energy, the error increases significantly. This is due to the fact that above 5 MeV, the error accompanied with the correction due to anisotropy of emission angle of source neutrons and inhomogeneity of thickness of the sample foil increases.

The present data show similar tendency in shape with those being compared, but they are some 5 % higher than the evaluations. However, the present data are within the quoted errors with those by Cierjacks et al./11/ and Meadows /12/. As for Pu-242, the error is a little larger in low MeV range than for Pu-239. In this experiment, a 2.5 Ci Ti-T target was used and it was found that the corrections due to parasitic neutrons and scattered neutrons by the construction materials were rather large, therefore, the measurement for Pu-239 was carried out with a 8 Ci Ti-T target with improved design. The present results generally show good agreement with those recently reported /12/ /13/, although compared closely with those of Behrens et al. /14/ which are the bases of JENDL evaluation /15/, they are about 4 % lower between 1 and 2 MeV and 4 % higher around 6 MeV.

Recently, we have measured the fission cross section of Pu-240 relative to U-235. They also show good agreement with the experimental data recently reported. Therefore, it is difficult to consider that the systematic disagreement in Pu-239 comes from the process of the measurement or the data handling. Since the Pu-239 sample contains relatively large amount of Pu-240 (3.16 %) which cannot be analyzed by alpha spectroscopy, the small change in the isotopic composition can have relatively large effect. The re-measurement of the mass spectrometry is now under way at JAERI, and the present result might be revised by the re-measurement.

Summary

The fission cross section ratios of Pu-239 and Pu-242 relative to U-235 were measured in the energy range from 0.6 to 7 MeV. The overall uncertainty of the present experiment was about 2 %. For Pu-239/U-235, the present results were about 5 % higher than the evaluated data (JENDL-2), however, there might be an ambiguity in the isotopic composition of Pu-239 sample. For Pu-242/U-235, the present results showed rather good agreement with other data recently measured, though there were differences of about 4 % with JENDL evaluation in certain energy range.

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