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MEASUREMENT OF ^{235}U FISSION CROSS SECTION AROUND 14 MeV

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Abstract: The neutron-induced fission cross section of U-235 was measured at five neutron energy points from 13.5 to 14.9 MeV by a newly developed detector which coupled a proton-recoil counter telescope with a fission chamber in back to back form. An experimental test of this detector was performed using the time correlated associated particle method. The experimental neutron sensitivity agreed with the analytical one within 1%. The fission cross section measurement was carried out for an U-235 sample with a purity of 99.91%. The overall uncertainties of the measurement were 2.5-2.8%. The present results agreed well in magnitude and in energy dependence , respectively, with the results by Cancé and by Czirr.

(fission cross section, ^{235}U, absolute, counter telescope, T(d,n) reaction)

Introduction

The neutron-induced fission cross section of U-235 is of prime importance as the standard cross section. For the measurement of the U-235 fission cross section, the time correlated associated particle (TCAP) method is the most reliable one. This method could provide the high accuracy (1-2%) around 14 MeV/1-4/. However, this method is difficult to apply for other energy regions.

The proton recoil counter telescope (PRCT) method have been used above a few MeV for determination of the neutron fluence in the fission cross section measurements/5-7/. The uncertainty by the PRCT method exceeds 1% in obtaining the neutron fluence and 2% in determining the fission cross section. But, this method is applicable in the wide energy range above 1 MeV.

We have measured the fission cross section ratios of various actinides relative to U-235 /8,9/. We have conducted the program to measure the absolute fission cross section of U-235 above a few MeV, and developed a detector which is suitable for the absolute measurement of U-235 fission cross section with the accuracy of 2-3%. In this paper, the outline of this detector and the experimental test of it are described. In the last section, a measurement of the fission cross section of U-235 around 14 MeV using this detector are explained with the results.

Counter telescope with fission chamber

The detector consists of a fission chamber for detecting fission events and a PRCT for determining neutron fluence at fission sample. The PRCT is coupled with the fission chamber in back to back form in order to minimize the uncertainty of the neutron fluence at the fission sample. The detector is shown in Fig.1.

A radiator of the PRCT is settled just behind a backing of the fission sample. The diameter of the radiator is almost same as that of the fission sample. A CsI(Tl) scintillator (1mm thick, 3cm in diameter), which is the main detector of the PRCT, is placed approximately 12cm apart from a radiator. The body of telescope is separated into three parts by apertures. Each part is functioned as a gas proportional counter with a platinum wire of 0.1mm in diameter. The signals from the gas counters are used as the gate signals for the scintillator signal. In this study, only two counters were used because the background in the recoiled proton spectrum could be reduced enough by the three-fold coincidence.

A fission chamber is a parallel plate ionization chamber which has an inner diameter of 60mm and a distance of 17mm between electrode and the sample. Cable connectors are attached far behind the sample and the radiator in order to reduce the scattered neutron which entered those. Due to the same reason, the walls of the detector are as thin as possible (see Fig.1). The stainless-steel as the detector material was used because no special material is required to reduce the background owing to the use of the coincidence. A mixture gas of Ar 95% and CO_2 5% flows continuously through the detector.

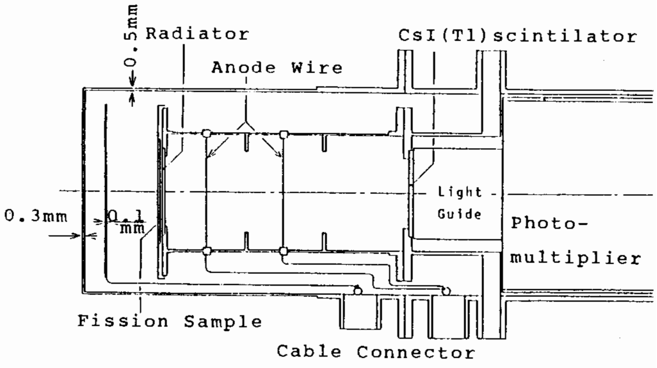

Fig.1. Counter telescope with fission chamber

Experimental test of telescope

The neutron fluence by the PRCT method is considered to be determined straightforwardly using the neutron sensitivity obtained analytically. But, in deriving the neutron fluence, an empirical procedure is required for obtaining the counts of recoiled protons/7,10/. Corrections are also needed/10/. In this study, an experimental test was performed to check the reliability of the detector, the data processing procedure and the correction methods by the TCAP method.

Experiment

The experimental arrangement is shown in Fig.2. The 14.7 MeV neutrons were produced via the T(d,n) reaction by 250 keV deuterons from the Cockcroft-Walton accelerator of Sub-Critical

Assembly Laboratory at Tohoku University. The deuteron beam was defined by a 3mm-diameter aperture on the neutron generation target.

A Si surface barrier detector (SSD) was used to detect the associated alpha particles. In the associated alpha particle spectrum, backgrounds from the D(d,p)T and ^{3}He(d,p)^{4}He reactions could not be observed owing to the use of a virgin Ti-T target. The telescope was placed according to the associated neutron cone measured by a NE102A plastic scintillator, so that all the neutrons coincided with the detected alpha particles by SSD could enter the radiator. The angle variation of the associated neutron cone was certified to be negligible by the agreement of the neutron cone between before and after the sensitivity measurement.

The sensitivity measurement was performed separating to nine individual foreground runs and two kind of background runs; one for random coincidence proton spectrum and the other for recoiled proton spectrum by replacing the radiator with a carbon foil. The reaction rate in the target was suppressed so that the counting rate of the SSD was less than 7000cps. Fig.3 shows the recoiled proton spectrum in which the background was already subtracted.

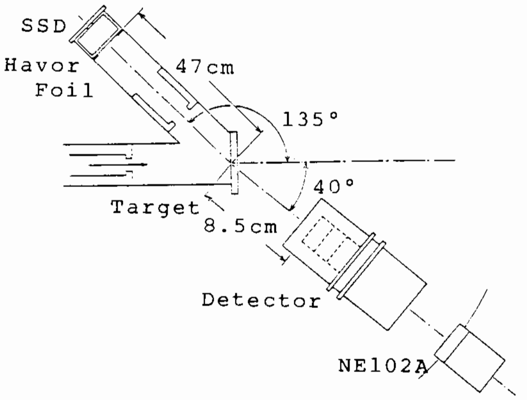

Fig.2. Experimental arrangement of sensitivity measurement

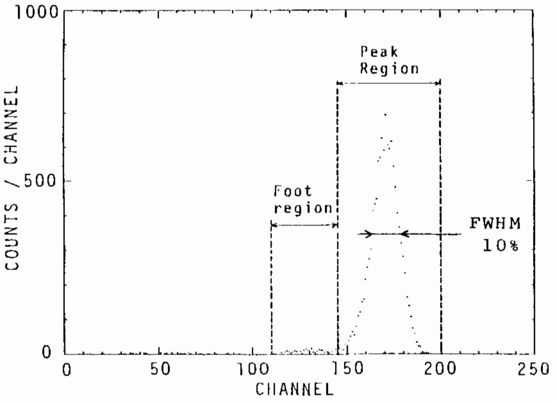

Fig.3. Measured recoiled proton spectrum

Experimental Sensitivity

The experimental neutron sensitivity EPS_{exp} was obtained by ;

$$EPS_{exp} = \frac{C_{AP}}{C_A\ (1-F_B)}\ F_{AT}\ F_P$$

where C_{AP} :count rate of recoiled protons coincided with associated alpha-particles

C_A :count rate of associated alpha particles

F_P :correction for interaction of recoiled protons

F_{AT} :correction for attenuation of source neutron

F_B :background fraction of associated alpha particles

The count rate C_{AP} was obtained by summing the counts over peak region in the recoiled proton spectrum from which the random coincidence spectrum was already subtracted. The separation point of the peak region to the foot region was chosen empirically. It became about 70% of the maximum pulse height (See Fig.3). The count rate C_A is the total counts of the alpha particles which is used as the gate signals to the recoiled proton signals. The background fraction F_B was 0.26%.

Correction factors

The correction factor F_P was defined as the ratio of the number of protons without interactions to the number of interacted protons remained in the peak region. In order to estimate this factor, a Monte Carlo code was developed. This code considers the interactions of recoiled protons with apertures, anode wires, counter gas, and polyethylene radiator. The output of this code is the perturbed energy spectrum of recoiled proton which enter the scintillator. Main contributor for the correction factor was the interaction with the anode wires.

The similar estimation has been reported by Siebert/10/. For their telescope against the 14 MeV neutron, the correction factors obtained by Siebert and our code was 1.012 and 1.015, respectively. The difference (0.3%) was within the statistical error of the calculation. The uncertainty by this correction is assumed to be 33%.

The estimation for this sensitivity experiment required the distributions of the energies and the angles of the neutrons which enter the radiator. These distributions could be obtained from the associated neutron cone measurements. Using this distribution, F_P became 1.020. The increase from the above cited value of 1.012 was caused by the increase of the neutrons which entered the center of the radiator.

The correction factors F_{AT} was estimated by a Monte Carlo code tracing the scattered neutron, as mentioned below. In this sensitivity experiment, the target backing, the target holder, the detector entrance window and the fission foil backing are taken into account. The value of this correction factor was 1.046.

Result of sensitivity measurement

The average sensitivity from the nine foreground runs was $1.138\text{x}10^{-5}$ counts/neutron. The total uncertainty of the experimental sensitivity was 2.0%. The analytical sensitivity was $1.127\text{x}10^{-5}$ counts/neutron obtained by integrating the equation of Bame/11/. The (n,p) cross section and its anisotropy was taken from ENDF/B-IV/12/ and Hopkins and Breit/13/. The experimental and analytical sensitivity agrees within 1%.

Measurement of fission cross section

The fission cross section of U-235 was measured using the mono-energy neutrons produced via the T(d,n) reaction by Dynamitron accelerator in Tohoku University. The U-235 sample (25mm in diameter and 94 microgram/cm^2) was prepared by the electro-deposition on a platinum plate.

Sample assay

The isotopic composition in the sample was determined by the mass spectrometry at Oak Ridge National Laboratory. It was also obtained from the measured spectrum, by a low geometry alpha spectrometer, by using the peak separation technique with the shape function of Garcia-Toraño and Aceña/14/. The fraction of U-235 agreed within 0.5% with the result by the mass spectrometry. This difference of 0.5% was taken into account in the uncertainty of the number of U-235 atoms. The number of atoms of the sample was obtained from the total activity, the isotopic composition and the half lives/15,16/.

Table 1 shows the isotopic composition, the relative activity and the number of atoms of this sample. The uncertainty of the number of U-235 atoms was 1.0%. Main contributors of this uncertainty were the statistics (0.8%), the efficiency of the low geometry detector (0.5%) and the isotopic composition error (0.5%).

Table 1

Isotope	Isotopic composition (%)	Relative activity (%)	Number of atoms ($\times 10^{17}$)
U-233	< 1ppm	<0.24	-
U-234	0.0298	45.84	0.003
U-235	99.912	53.66	10.40
U-236	0.0165	0.27	0.002
U-238	0.0414	-	0.004

Measurement

Fig.4 shows the experimental arrangement in this measurement. The fission sample and the radiator (28.04 mg/cm^2) were placed approximately 15cm apart from the neutron target, where an assumption of the isotropic neutron emission from the point source would be adequate in the calculation of the neutron sensitivity. Also, by taking this distance, the inhomogeneity of the deposit thickness could be neglected.

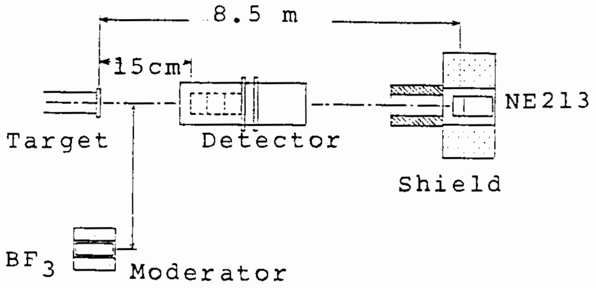

Fig.4. Experimental arrangement of fission cross section measurement

The energies of neutrons which irradiated the fission sample were varied by rotating the detector around the target from 0 to 143 degree. The neutron energies and its spreads on the fission sample were calculated by the kinematics equation considering the T(d,n) cross section /17/, the stopping power of Ti/18/ and the geometrical factors. No parasitic neutron was observed in the neutron spectra measured by the NE213 liquid scintillator on the deuteron beam line with the n-γ discrimination circuit owing to the use of the fresh Ti-T target. The recoiled proton spectrum and the fission fragment spectrum were accumulated simultaneously. A typical fission fragment spectrum is shown in Fig.5.

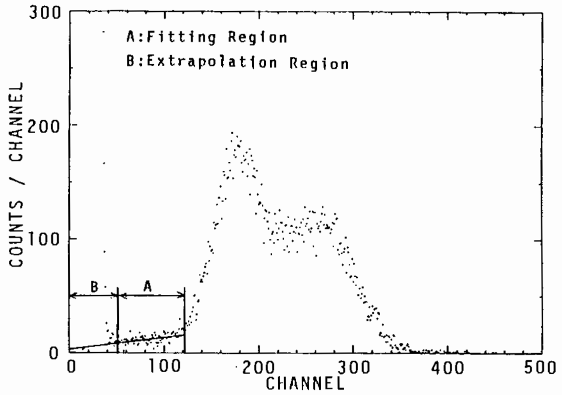

Fig.5. Typical fission fragment spectrum

Each measurement consisted of foregrounds run and two background runs. One background run was performed without the radiator (with the carbon foil). The fraction of this background relative to the foreground was about 0.4%. The other background run was carried out with a iron bar of 32cm long to shadow the direct neutrons. This background was about 6% of the foreground. In order to place the shadow bar, the detector should be moved backward. The effect of this movement was verified to be negligible by measurement of position dependence of this background. A pulse generator was employed to evaluate the loss of recoiled protons in the coincidence circuit. The correction factor for this effect was 1.010.

Fission cross section derivation and correction

The fission cross section was derived by the following equation;

$$\sigma_f = \frac{2\ \mathrm{EPS}\ C_F}{N\ C_P\ \ln[(S^2+R^2)^{1/2}/S]} F_{SA}\ F_P\ F_{TS}\ F_{CS}\ F_{AT}$$

where C_F :count rate of fission
C_P :count rate of recoiled proton
N :areal density of sample
S :distance from target to sample
R :radius of sample
EPS:analytical efficiency of telescope
F_{SA}:correction for self absorption of sample
F_P :correction for interaction of recoiled proton
F_{TS}:correction for scattered neutron by target material
F_{CS}:correction for scattered neutron by chamber material
F_{AT}:correction for attenuation of source neutron

The counts of the recoiled protons were obtained by the same procedure mentioned above, after correcting the coincidence loss. The fission counts were obtained by adding the extrapolated counts (about 1.5%) to zero pulse height. The extrapolations were made by the fitting with the exponential function (See Fig.5).

The code mentioned above was applied to calculate the correction factor for the interaction of the recoiled protons (F_P:0.988). For the correction for the scattered neutrons, we developed an another Monte Carlo code which traced the scattered neutron and calculated the energies and the angles of the direct and the scattered neutrons which entered the radiator and the fission sample. This code was proved to be adequate from the agreement of the neutron spectrum and the fraction of the scattered neutrons calculated by MCNP/19/ and this code. The correction factors by the target assembly (F_{TS}:0.990-0.993) and the chamber material (F_{CS}:0.970-0.979) were estimated separately.

The effects of the self-absorption (F_{SA}: 1.023-1.024) in the fission sample were estimated by the method of Carlson/20/ considering the fission fragment anisotropy/21/. The correction for the neutron attenuation (F_{AT}:0.979) in the sample backing was also taken into account.

Result

The total uncertainties is from 2.5 to 2.8%. Main contributors are the sample assay (1.0%), the (n,p) scattering cross section and its anisotropy (1.2%) and the fission count statistics including the extrapolation and the backgrounds(1.0 to 1.5%).

Table 2
Result of fission cross section measurement

Neutron energy(MeV)	Fission cross section(b)	Uncertainty (%)
13.51±0.15	1.995±0.050	±2.5
13.97±0.04	2.040±0.054	±2.6
14.10±0.05	2.057±0.058	±2.8
14.49±0.17	2.119±0.058	±2.7
14.90±0.29	2.114±0.058	±2.7

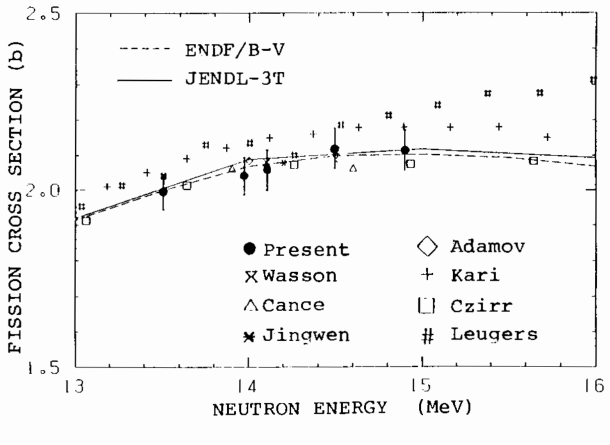

Fig.6. Result of fission cross section measurement of U-235 around 14 MeV

The present results of this measurement are given in Table 2 and Fig.6. This results agree very well in magnitude with the results by using the TCAP method by Cancé (2.062±0.039b for 13.9MeV)/2/, Jingwen (2.078±0.040b for 14.2 MeV)/4/, Wasson (2.080±0.030b for 14.1 MeV)/1/ and Adamov (2.084±0.034b for 14.0 MeV)/3/ considering the experimental uncertainties.

Fig.6 indicates that the energy dependence of the present results have good agreement with those by the shape measurements of Czirr/7/ and Kari/22/, and the evaluated data files of ENDF/B-V/23/ and JENDL-3T*/24/.

Summary

A proton recoil telescope coupled with a fission chamber was developed for measuring the fission cross sections and was tested using the time correlated associated particle method. The experimental neutron sensitivity of the telescope agreed with the analytical one within 1%. This experiment test was proved the applicability of this telescope and the validity of the data processing procedure and the correction methods to determine the neutron fluence. The fission cross sections of U-235 were measured at five energy points around 14 Mev. The results were obtained with the experimental uncertainty of 2.8% or less. The present results agreed well both in magnitude and in energy dependence with those of the recent measurements.

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* JENDL-3T is a temporary file for testing the evaluated data file which are for JENDL-3.