KERNFORSCHUNGSZENTRUM

KARLSRUHE

Oktober 1966

KFK 450
IAEA-CN-23/7

Institut für Angewandte Kernphysik

The Fission Cross-Sections of Some Plutonium Isotopes in the Neutron Energy Range 5 - 150 keV

W. B. Gilboy, G. Knoll

GESELLSCHAFT FUR KERNFORSCHUNG M. B. H.

KARLSRUHE

KERNFORSCHUNGSZENTRUM KARLSRUHE

October 1966

KFK 450
CN-23/7

Institut für Angewandte Kernphysik

The Fission Cross-Sections of some Plutonium Isotopes in the Neutron Energy Range 5-150 keV

W. B. Gilboy and G. F. Knoll

Gesellschaft für Kernforschung m.b.H. Karlsruhe

INTERNATIONAL ATOMIC ENERGY AGENCY

CONFERENCE ON NUCLEAR DATA -
MICROSCOPIC CROSS SECTIONS AND OTHER
DATA BASIC FOR REACTORS

Paris, October 17 - 21, 1966

CN-23/7

THE FISSION CROSS-SECTIONS OF SOME PLUTONIUM ISOTOPES IN THE NEUTRON ENERGY RANGE 5-150 keV

W. B. Gilboy[1)] and G. F. Knoll[2)]
Kernforschungszentrum Karlsruhe[3)]

1. Introduction

In the prototype fast reactors presently being planned a large fraction of the neutron population lies in the energy interval between 5-150 keV. Due to technical difficulties however much of the required neutron data in this region is only known to limited accuracy. X The present work was undertaken to provide improved fission cross-section data on ^{239}Pu and ^{240}Pu, which sensitively effect the behaviour of plutonium-fuelled fast neutron assemblies. In order to avoid the necessity of measuring the neutron flux in this rather difficult energy region the fission cross-section ratios ^{239}Pu/^{235}U and ^{240}Pu/^{235}U are actually measured. Plutonium cross-sections can be derived from these ratios by reference to the best available values of the ^{235}U fission cross-section. This indirect approach was chosen since ^{235}U is much more suitable for absolute cross-section measurements and some accurate ^{235}U results which have recently been published [1,2] may be used for this normalization. X

1) Now at A.W.R.E. Aldermaston, Berks. England.

2) Now at University of Michigan, Ann Arbor, Mich. U.S.A.

3) Work performed within the Association in the Field of Fast Reactors between the European Atomic Energy Community and Gesellschaft für Kernforschung m.b.H. Karlsruhe

2. Apparatus and Method

The pulsed and bunched proton beam from the Karlsruhe 3 MeV Van de Graaff was used to provide 1 ns duration neutron bursts at 1 Mc/s repetition rate via the $Li^7(p,n)Be^7$ reaction. Thick lithium metal targets were employed to give a wide spread in proton energies and a consequent continuous ("white") spectrum of neutrons so that cross-section ratios could be simultaneously measured over a wide energy range which considerable quickened the rate of data acquisition. Two fission counters, containing ^{235}U and ^{239}Pu (or ^{240}Pu) respectively, were placed symmetrically about the pulsed neutron source in order to sample identical fluxes. Runs were repeated with the counters interchanged to average out any slight source asymmetries. Flight paths from 7.5 to 30 cm were chosen to give these best compromise between neutron energy resolution and fission counting rate. With typical proton currents of about 6 μA runs of between 8 and 16 hours were required to attain reasonable statistics.

The fission samples, manufactured in C.B.N.M. at Geel by the electro-spray technique, are detailed in Table I. For ^{235}U and ^{239}Pu a nominal thickness of 1 mgm/cm^2 was specified to keep the self-absorption corrections fairly small, but a thinner sample of ^{240}Pu was necessitated as only a small amount of this material was available.

The fissile samples were mounted in Xenon gas scintillation counters to detect induced fissions. The very short decay time of scintillations in Xenon ($\sim$1ns) helps to reduce pile-up of α-pulses from the highly active plutonium and gives fast output pulses which enable the neutron energies to be determined by time-of-flight. The fission counters were fabricated out of stainless steel to the design shown in Fig. 1. To reduce potential poisoning of the gas scintillator no wavelength shifter was used and since the pure Xenon scintillates in the ultra-violet end of the spectrum both the window of the chamber and the photomultiplier (56 UVP) were made of quartz. The distance between the fission sample and the chamber window was 5 cm which gives a total flight path of at least 10 cm for neutrons backscattered from the window and the multiplier. This reduced the time correlated backgrounds at the fission sample to a low level. Using thin lithium targets these backscattered neutrons could be time resolved and a typical value of $\sim$1.5% of the primary neutron intensity was measured which has only a very slight effect on the ratios over the energy range studied. As a further check on this point thick target runs were made at several different flight paths and with various maximum neutron energies (by altering the accelerator energy) which should alter the effects of time correlated backgrounds. The ratios measured with these different dispositions were always in good agreement showing that spectral distortions due to scattered neutrons were small.

Due to the intense α-activity ($\sim 10^8$ sec^{-1}) in the plutonium chambers it was impossible to take slow (~ 1 μ sec) outputs from these counters in order to measure pulse-height spectra for setting bias levels. Instead fast outputs (~ 5 ns) were used to trigger fast discriminators which operated the START channels of a nanosecond time-sorter; STOP pulses were derived from a beam pick-up aerial near the neutron producing target. In this way time spectra from the two fission chambers were recorded simultaneously on identical time scales. The spectra from both counters were routed to different parts of the memory of a CAE 510 on-line computer in which the data was accumulated and inspected. The raw time spectra were recorded on punched paper tape for later off-line analysis.

Using a single fast bias level the overall time resolution for neutron events was about 3 ns but for some of the measurements a dual fast bias system was adopted which improved the resolution to about 1.5 ns. Figure 2 shows a typical time spectrum from the ^{235}U chamber with the neutron induced counts collected into 10% lethargy intervals which illustrates the relative neutron source intensity in the range 5-150 keV. The small γ-ray peak was found to be due mainly to γ-rays from the lithium target interacting in the quartz window and photo-multiplier. This γ-peak was useful for fixing the zero of the time scale. The counters were usually biased so that there were almost no counts due to α-pile-up events and the random backgrounds under the time correlated events were found to be due mainly to room scattered neutrons most of which had energies above the cadmium cut-off. In the case of ^{240}Pu the random background also contained spontaneous fission events. This clear time discrimination of most types of background events was an important aid to accurate background removal.

The ratio of the counting rates in the Pu and U fission counters as a function of neutron energy can be derived directly from the background subtracted time spectra and is related to the fission cross-section ratio by a factor containing the relative numbers of fissile nuclei in the two counters multiplied by the ratio of their bias levels. These bias factors were obtained in several different ways which are described in the following sections. These factors were fed into a data analysis computer programme which subtracted backgrounds and combined the various runs according to their statistical weights. The programme also calculated the neutron energies and divided the data up into equal lethargy intervals of any desired width. The programme output tabulated the final cross-section ratios together with their statistical errors as a function of the mean neutron energy in each lethargy interval. The results are shown in Figures 3 and 4.

3. Fission Counter Bias Factors

3.1 ^{239}Pu/^{235}U: Method A

As only fast output pulses were available from the Pu-counters fission fragment pulse height spectra were obtained by measuring integral bias curves for equal steps in the threshold settings of the fast discriminators on both detectors. For this purpose the fission rates were greatly increased by surrounding each counter by polythene to moderate neutrons into the energy region near thermal where both isotopes have large fission cross-sections. Pulse height spectra were derived from these integral counting rates by differentiation. Since the two counters contain closely similar fissile deposits the fission spectra for both chambers are expected to be almost identical and this was borne out by the measurements. The spectra from the two counters were normalised to each other and the relative efficiency for counting fissions over the bias levels used in the ratio measurements were calculated by integrating areas under the normalised distribution. Due to the greater α-pile-up the Pu chamber was usually biased about twice as high as the U-chamber. The relative numbers of ^{239}Pu and ^{235}U atoms were calculated from the sample masses with allowance for the other isotopes. The Pu-masses in Table I are given to 5% and are only provisional pending a more accurate assay by destructive analysis. Ratio measurements based on this method of defining the biases are shown in Figure 3.

3.2 ^{239}Pu/^{235}U: Method B

Both of these isotopes have large fission cross-sections near thermal energies and consequently they can be accurately measured. In particular the ratios of these cross-sections have been measured to about 2% accuracy very recently [3] over the range 16-550 meV. Therefore if the present two fission detectors are compared in the same low energy flux their relative bias levels can be calculated from a measurement of their relative counting rates and a knowledge of the fission cross-section ratio at the energy used. This method actualy gives the ratio of the products <Number of fissile nuclei x Fission counting efficiency> so it also obviates the need to know the masses of the two fissile samples. The fission chambers were irradiated with neutrons of precisely known energies from the crystal spectrometer on the Karlsruhe FR 2 reactor and the counting rate ratio measured for 41 meV neutrons. Another series of Pu/U counting ratios were measured between 41 meV and 271 meV to check for second order neutrons and these produced a small "ghost" peak at 75 meV corresponding to the strong 300 meV resonance in ^{239}Pu. A 2% correction was estimated for the 41 meV ratio due to second order neutrons; higher orders were negligible. The cross-section ratio at 41 meV was derived by using some older measurements [4] normalised to the new data in reference [3]. In order to conveniently refer to the standard bias ratio defined by the 41 meV measurement a relative bias "indicator" was devised. This consisted of a polythene moderator containing a Po-Be neutron source which was placed over each of the counters in turn and the fission rate ratio measured shortly before the 41 meV standardization runs were made. For later runs at the Van de Graaff the same moderated source was used in identical geometry and from the fission rate ratio with this source the new relative bias conditions could be defined with respect to the 41 meV standard. The bias indicator method was used to check the bias conditions during a further set of accelerator runs and the results are shown in Figure 3.

3.3 ^{240}Pu/^{235}U

^{240}Pu has a very small low energy fission cross-section and has a higher specific α-activity than ^{239}Pu. For these reasons it was difficult or impossible to apply the methods described above for fixing the relative bias levels. However, ^{240}Pu is spontaneously fissile and the sample used had a total spontaneous fission rate of $\sim$2.4 sec^{-1}. With the bias set to cut out α-pulses a spontaneous fission rate of about 1 sec^{-1} was observed. Since this is proportional to the amount of ^{240}Pu in the sample it can be shown that it is unnecessary to know either the mass of ^{240}Pu or the efficiency for counting fissions over the bias if the induced fission rate is always measured relative to the spontaneous fission rate. This method also required a knowledge of the spontaneous fission half-life which has been measured to $\pm$ 1% by Watt et al [5]. In this case the absolute efficiency for counting fissions with the ^{235}U counter has to be measured and this was done in the following way. The moderated Po-Be source was used to give a large number of fission events in the ^{235}U chamber and a slow output from the counter was analysed in a multi-channel analyser to display the fission spectrum. This spectrum was fitted with a semi-theoretical spectrum derived by Kahn et al [6] in order to extrapolate to zero pulse height; a correction was also applied for self-absorption in the fissile layer. Once the total fission rate is known for the Po-Be source in its standard moderator geometry the total fission rate at any other time can be calculated for the same source. The counting rate actually observed with this source gives the fission counting efficiency for the biassing conditions at the time of the measurement. The only other quantity required for the overall bias factor is the mass of the ^{235}U sample which was accurately measured in Geel by careful weighing techniques. The small fission cross-section of ^{240}Pu below 150 keV gave a rather small counting rate and long runs were made at flight paths as short as 7.5 cm in order to achieve reasonable statistics.

4. Discussion of Results

The ^{239}Pu results are shown in Figure 3 together with ratios measured by Allen and Ferguson [7] and by White et al [2,8]. The data are presented in 10% lethargy intervals and no marked structure is apparent beyond the expected statistical fluctuations. The statistical errors rise from about 1% at the higher energies to 5% at 5 keV. There is a further systematic uncertainty in the upper set of points of about 3% due to uncertainties in the bias factors derived from matching the fission pulse height spectra. Another scale uncertainty of 5% arises from the ^{239}Pu mass determination but this should be greatly reduced when the results of the final assays are available. Method B is independent of this mass measurement and an overall uncertainty in the scale factor of about 4% is estimated stemming mainly from the statistical errors in the bias indicator counts. Taking into account the statistical and systematic uncertainties the two sets of ^{239}Pu results agree reasonably well. A weighted mean of these two sets is in excellent agreement with the work of White and agrees well in slope with the measurements of Allen and Ferguson, although lying about 5% lower.

The results for ^{240}Pu are given in Figure 4 with the error flags showing the estimated overall errors. Since the induced fission rate for the ^{240}Pu sample was rather small the counts were combined into 30% lethargy intervals to improve the statistics. Data is only shown down to 15 keV as the statistical errors at lower energies were very large. Included in the assigned errors is a scale factor uncertainty of 5% which is dominated by the statistical errors on the spontaneous fission counts taken to determine the bias. The data is in good agreement with measurements by White et al [2, 9] which are representative of results by other workers [10, 11]. The present results extend to lower energies than previous measurements and indicate a second fission threshold at about 10 keV. Using the ^{235}U fission cross-section [1, 2] the ^{240}Pu fission cross-section derived from the ratios is in good qualitative agreement with a theoretical curve of De Vroey et al [10] which describes the cross-section below 200 keV as being mainly due to p-wave neutrons.

Table I: Details of Fissile Samples

Sample	Total Weight mgm[a]	Density[b] mg cm^{-2}	Isotopic Analysis atom %
Uranium 235	23.62 ± 0.03 (U_3O_8)	1.204	^{235}U ii.505; ^{234}U 0.168 ^{236}U 0.026; ^{238}U 0.301
Plutonium 239	21.59 ± 1.08 (PuO_2)	1.100	^{239}Pu 99.979; Pu^{240} 0.020 ^{241}Pu 0.001
Plutonium 240	5.70 ± 0.25 (PuO_2)	0.290	^{240}Pu 99.60; ^{239}Pu 0.31 ^{241}Pu 0.07; ^{242}Pu 0.02

a Chemical purity of ≥ 99.9% in each case

b Area of fissile deposit 19.63 cm^2 (5.0 cm diameter circle)

References

[1] White, P. H., J. Nuclear Energy A/B 19 (1965) 325

[2] Perkin, J. L., White, P. H., Fieldhouse, P., Axton, E. J., Cross, P., Robertson, J. C., J. Nuclear Energy A/B 19 (1965) 424

[3] White, P. H., Reichelt, J. M. A., Warner, G. P. (see Proceedings of this Conference)

[4] Hughes, D. J., Schwartz, R. B., Brookhaven National Laboratory Report BNL 325, Second Edition (1960)

[5] Watt, D. E., Bannister, F. J., Laidler, J.B., Brown, F., Phys. Rev. 126 (1962) 264

[6] Kahn, S., Harman, R., Forgue, V., Nucl. Sci. Eng. 23 (1965) 8

[7] Allen, W. D., Ferguson, A. T. G., Proc. Phys. Soc. A70 (1957) 573

[8] White, P. H., Hodgkinson, J. G., Wall, G. J., Physics and Chemistry of Fission, Vol, I, I.A.E.A., Vienna (1965)

[9] Ruddick, P., White, P. H., J. Nuclear Energy A/B 18 (1964) 561

[10] De Vroey, M., Ferguson, A. T. G., Starfelt, N., Physics and Chemistry of Fission, Vol. I, I.A.E.A. Vienna (1965)

[11] Nesterov, V. G., Smirenkin, G. N., J. Nuclear Energy A/B 16 (1962) 51

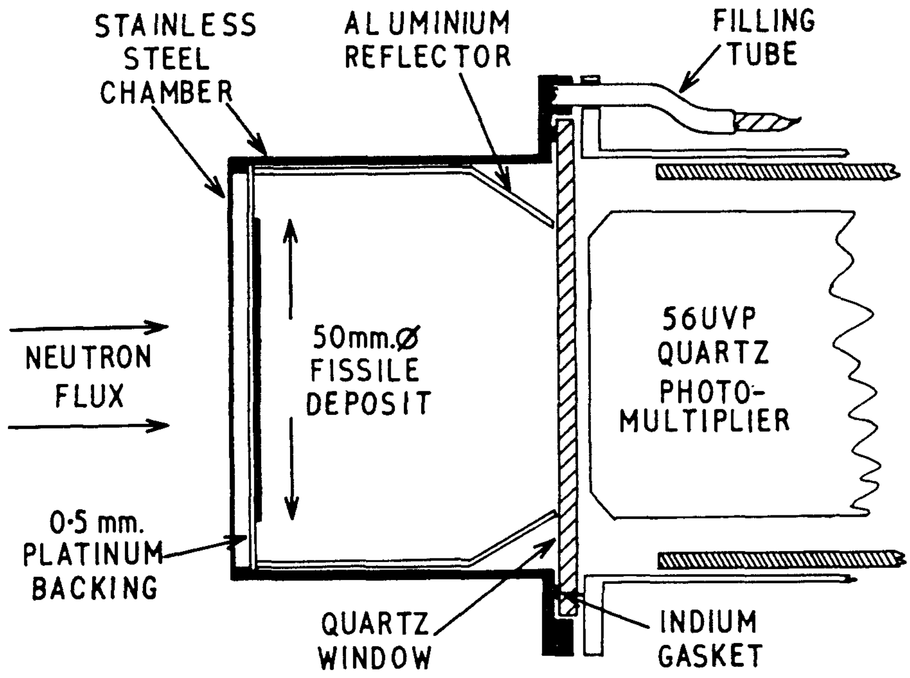

FIGURE 1. GAS SCINTILLATION FISSION COUNTER.

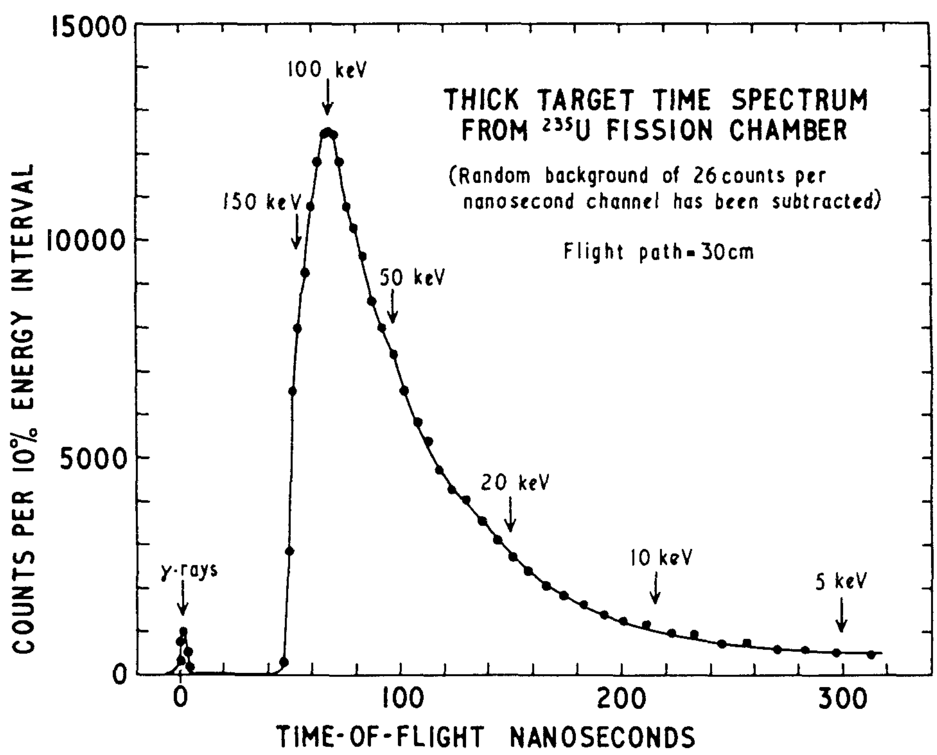

FIGURE 2. TYPICAL THICK TARGET TIME SPECTRUM FROM ^{235}U FISSION COUNTER

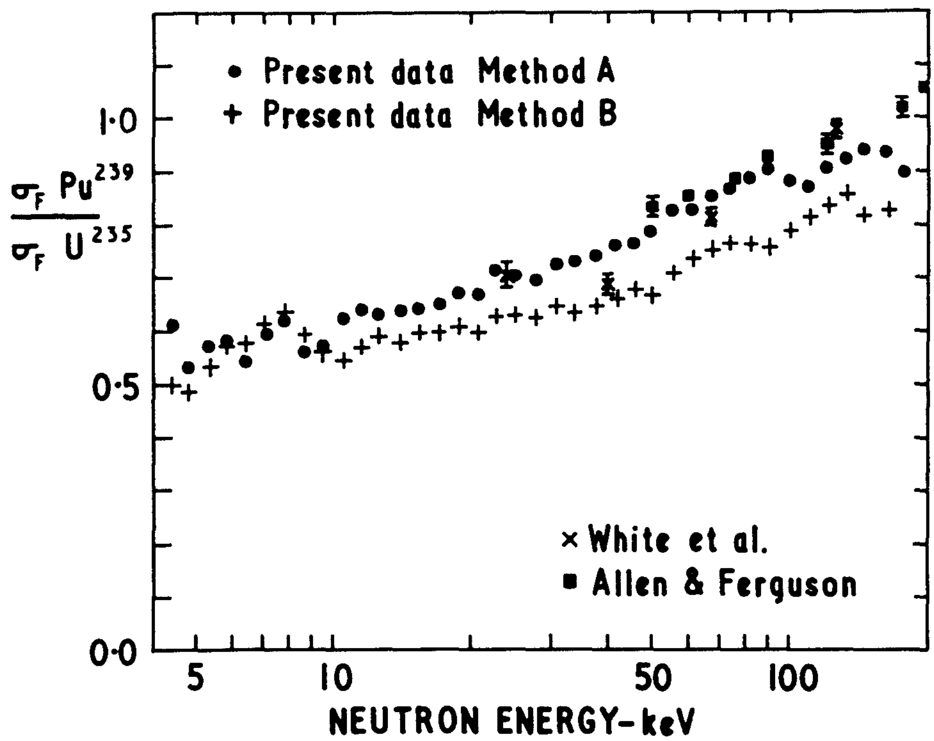

FIGURE 3. $^{239}Pu/^{235}U$ FISSION CROSS-SECTION RATIO at 10 % LETHARGY INTERVALS

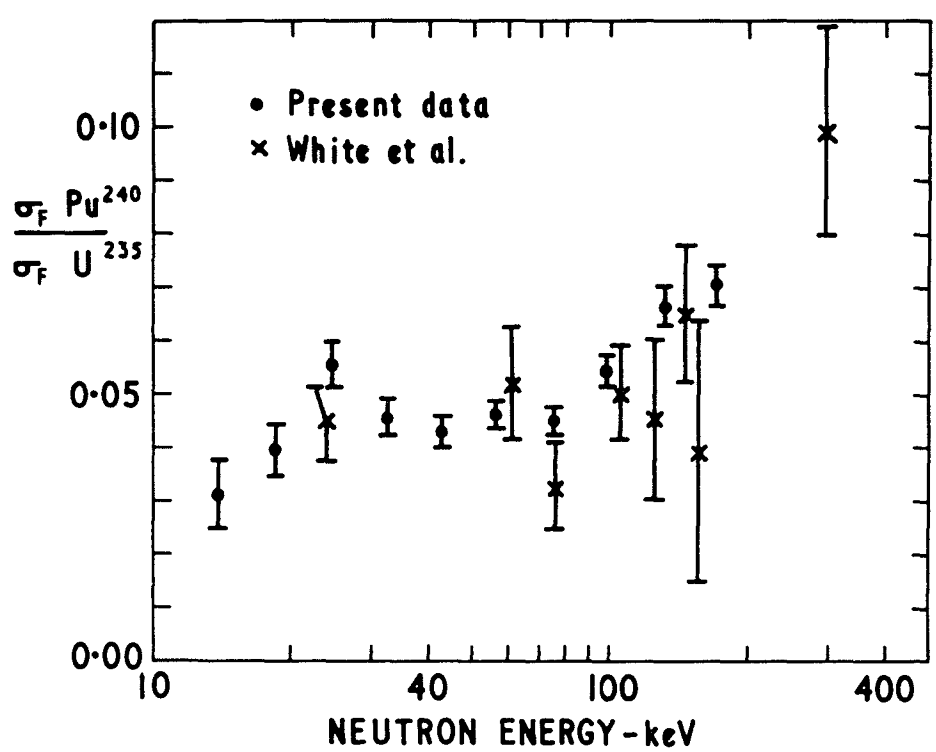

FIGURE 4. $^{240}Pu/^{235}U$ FISSION CROSS-SECTION RATIO AT 30 % LETHARGY INTERVALS