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YIELDS AND ENERGIES OF DELAYED NEUTRONS FROM FAST FISSION IN URANIUM AND PLUTONIUM

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Abstract: The properties of delayed neutrons from the aggregate of fragments produced by fission in uranium or plutonium are of interest not only in reactor physics but also for comparison with the summed properties of individual precursors. We report measurements on delayed neutrons from neutron-induced fast fission in natural uranium, where the interest was in absolute and fractional yields, and in ^{239}Pu, where the interest was in energies. The primary neutrons were from an accelerator target containing tritium or beryllium bombarded by modulated beams of protons or deuterons of various energies; delayed neutrons were measured when the accelerator beam was off. For yield measurements, the fission rate in the sample was deduced from that in a ^{238}U fission chamber mounted close to it, and the delayed neutrons were counted in a calibrated precision long counter; the sample was shuttled automatically between its irradiation and counting positions. In separate measurements, a ^{3}He spectrometer provided the energies of delayed-neutrons from ^{239}Pu.

(Fission, uranium, plutonium, delayed neutrons, yields, energies)

Accelerator-based neutron sources used to produce fission

At the 1986 Specialists' Meeting on Delayed Neutrons[1], we reported the use of the Birmingham 3MV Dynamitron accelerator in measurements of delayed-neutron yields from fast fission in natural uranium[2]. For technical reasons concerned with the effect of modulation on the energy of the accelerator beam, the neutrons producing fission were obtained by bombardment of a thick beryllium target with a modest beam of deuterons (up to 20μA at 1 to 3 MeV) instead of a much higher current of protons on tritium. We have now extended the measurements at a deuteron energy of 2.5 MeV to a second uranium sample of a larger diameter but smaller mass than the first; the first sample had a diameter of 25 mm, a thickness of 5 mm, and a mass of 48.38g, the second a diameter of 44.58 mm, a thickness of 0.3 mm, and a mass of 9.36 g.

Energy spectra and angular distribution of neutrons from the ^{9}Be(d,n)^{10}B reaction have been reported by Inada et al[3], and some of their results are given here as Figures 1 and 2 to indicate the situation at 2.5 MeV, an

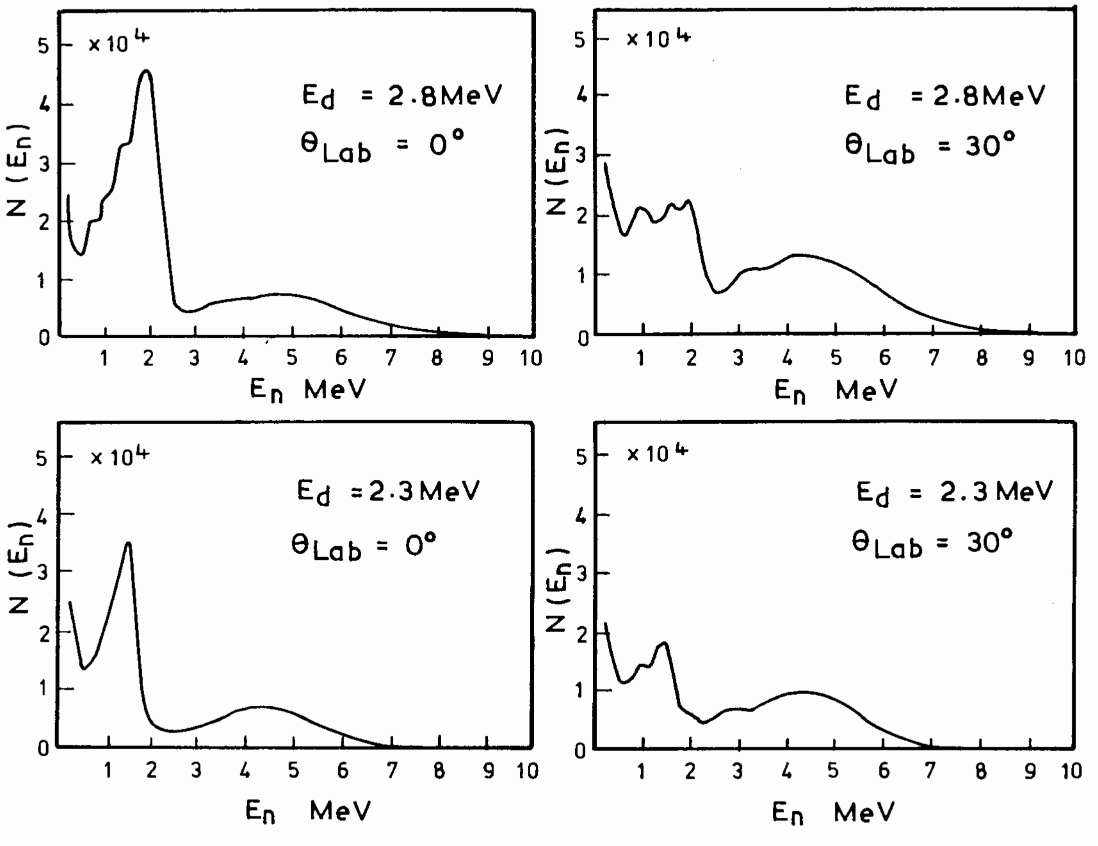


Fig. 1 Energy spectra of neutrons from ^{9}Be(d,n)^{10}B (after Inada et al[3]).

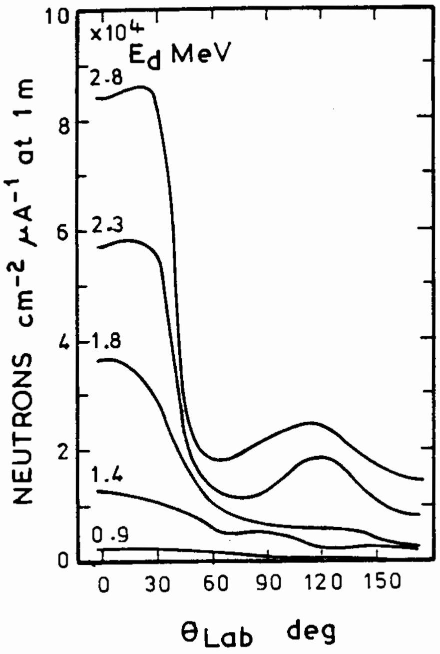


Fig. 2 Angular distribution of fast neutron fluence from $^{9}Be(d,n)^{10}B$ (after Inada et al[3]).

energy which they did not actually use. At this energy, the emitted neutrons have a mean energy of approximately 2.4 MeV, and therefore irradiation by them is likely to produce a delayed neutron yield from natural uranium reasonably similar to that produced by prompt neutrons from fission. However, our intention is to measure yields from fission produced by monoenergetic primary neutrons at various energies, and for this the $T(p,n)^{3}He$ reaction is appropriate. Indeed, we have used it extensively already with gas or solid targets for measurements of the dependence of delayed-neutron energies on the energy of the neutrons producing fission[4,5]; the gas target was used some years ago on the 3MV Van de Graaff accelerator IBIS at Harwell and the solid one (tritium absorbed in titanium on a backing of copper or silver) has been on the Birmingham Dynamitron.

Whichever reaction is used, the beam of charged particles has to be modulated to allow delayed neutrons to be measured after irradiation of the sample. In our case the modulation is via the extraction potential of the ion source and/or a mechanical chopper, and it is essential that there should be no break-through during the measuring period. Mechanical chopping of the beam after acceleration avoids the change in energy mentioned earlier but our first chopper was not designed to accept high powers; a new system has now been constructed to do so. The modulation cycle for yield measurements has been 40s irradiation followed by 40s counting, with small delays between the two; for energy spectra, the corresponding times have been 0.8s with delays of 0.1s.

Measurement of numbers of fissions and delayed neutrons in yield studies

To obtain delayed neutron yields, it is obviously necessary to measure the number of fissions produced in the sample and the number of delayed neutrons emitted. The former has been obtained by monitoring the irradiating neutron field with a parallel-plate ^{238}U fission chamber mounted as close as possible to the natural uranium sample, and the latter by a calibrated precision long counter of the de-Pangher type. Although the effective mass of the thin ^{238}U coating on the cathode of the fission chamber was provided by the manufacturer, it was also measured by comparing the chamber's efficiency with that of the long counter. Both were exposed for many hours to an ^{241}Am-Be source, the long counter having been calibrated previously by the National Physical Laboratory, Teddington, U.K. and also subjected to a series of measurements in the Birmingham Radiation Centre's low-scatter cell of its effective centre for ^{241}Am-Be, ^{252}Cf, and accelerator sources of different energies. The neutron flux distribution in the irradiating field was measured over the volume occupied in the experiment by the uranium sample and the parallel plate fission chamber. Movement of the chamber itself provided the distribution along the extrapolated axis of the deuteron beam; Figure 3 shows the results obtained when the inverse of the square root of the fissions in the chamber per unit beam charge is plotted against the distance of the chamber from the accelerator target. This information allows the fissions in the sample to be computed from those in the chamber when the relative effective masses are known. The flux distribution in the plane of the sample was obtained by separate measurements with a small (6 mm diameter) ^{238}U fission chamber scanned automatically by stepping motors; the ratio of counts in the small and large (parallel-plate) chambers shown in Figure 4 enabled corrections to be made for any difference in flux shapes across the sample and the monitoring fission chamber. With the thin sample, where the diameter is very similar to that of the chamber cathode, this correction is negligible; even for the smaller diameter sample, it is less than 2%.

In the yield measurements, the uranium sample was shuttled automatically over a distance of 0.8 m in less than 0.5 s by a pneumatically-driven arm from its irradiation position close to the accelerator target to its counting position about 115 mm from the face of the inner polythene cylinder in the long counter; Figure 5 illustrates the arrangement. The long counter had its efficiency measured for this geometry by a long series of comparisons of delayed-neutron counts with the uranium sample either held in its irradiation position or shuttled.

On the 6-group model of aggregated precursors, the emission rate of delayed neutrons produced by an infinitely recurring periodic irradiation is:

$$FY \sum_{i=1}^{6} a_i \frac{(1-e^{-\lambda_i t_b})}{(1-e^{-\lambda_i \tau})} e^{-\lambda_i t_d} e^{-\lambda_i t}$$

where

Y	is the yield of delayed neutrons per fission.
F	is the fission rate in the sample during irradiation.
t	is the time relative to the start of the count period.
τ	is the beam-modulation period.
t_b	is the irradiation time.
t_d	is the delay between the end of irradiation and the start of counting.
λ_i	is the decay constant of precursor group i.
a_i	is the relative yield of precursor group i.

This expression allows the yield Y to be extracted from the measured time-dependence of delayed-neutron intensity if the relative yields and decay constants of the six groups are assumed. Alternatively, group constants such as relative yields for assumed decay constants, can be extracted by the best fitting of predicted and experimental decay data.

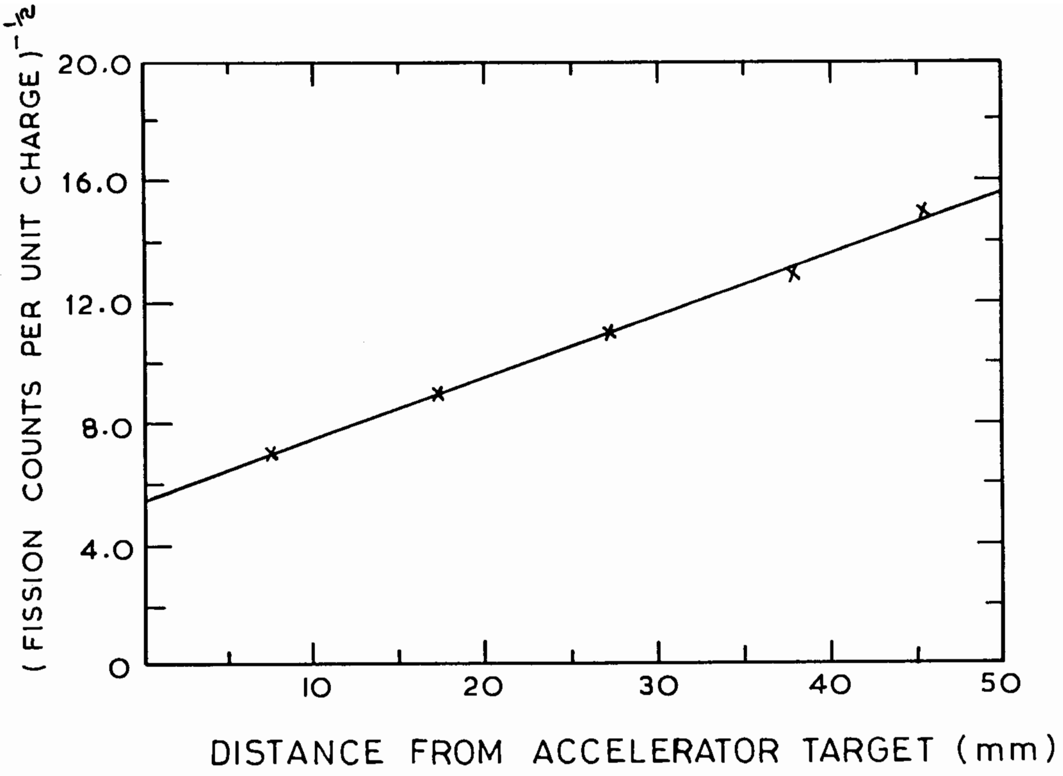


Fig. 3 Fission chamber output at different distances from accelerator target.

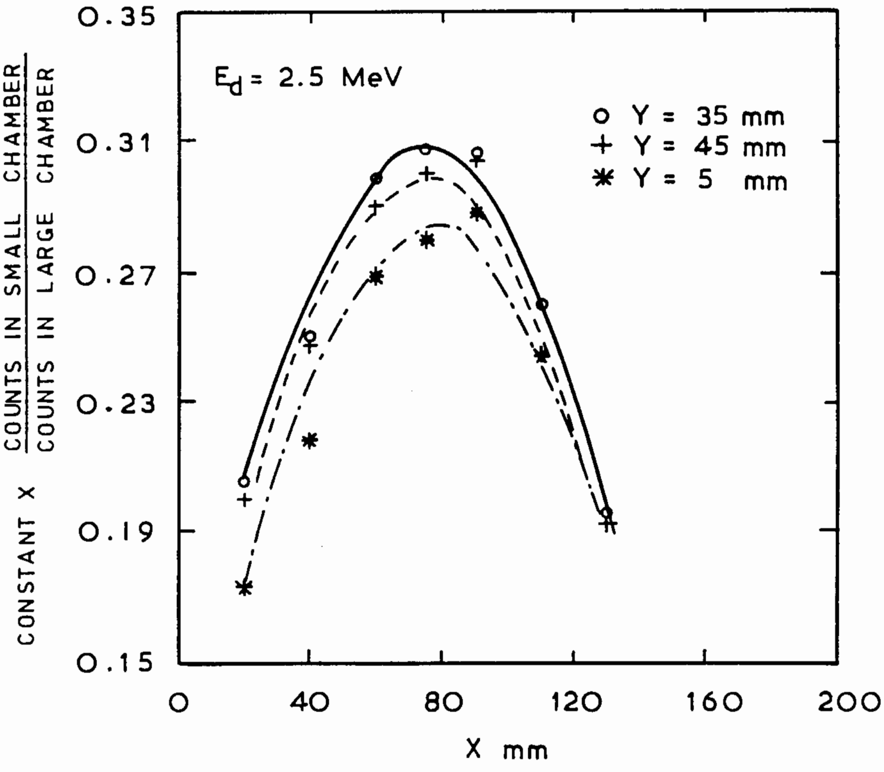


Fig. 4 Neutron flux distributions in the plane of the uranium sample.

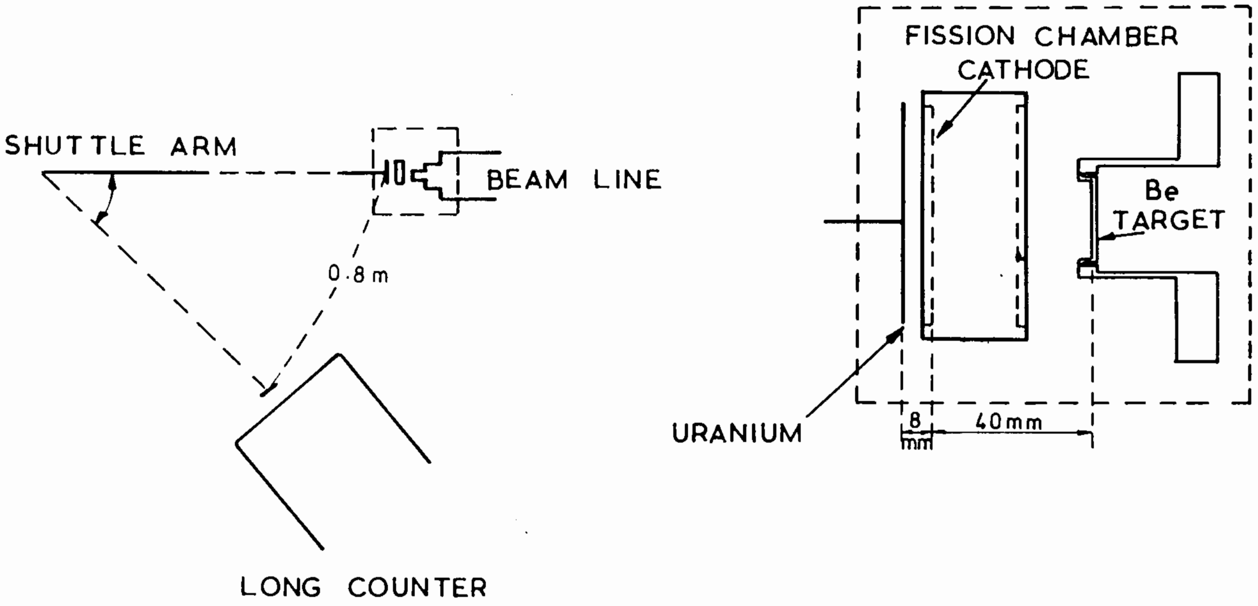


Fig. 5 Arrangement for yield measurements with (inset) enlarged view of accelerator target region.

Figure 6 shows the time dependence of the delayed neutron counts from each of the uranium samples. These data are being linked with the long-counter efficiency and the number of fissions in the sample, obtained as described earlier, to obtain the values of absolute and fractional yields shown in Tables 1 and 2 show the current situation.

Tablel: Absolute yields of delayed neutrons from fast fission in natural uranium

Sample 1 (25 mm dia.): 0.0495±0.0003

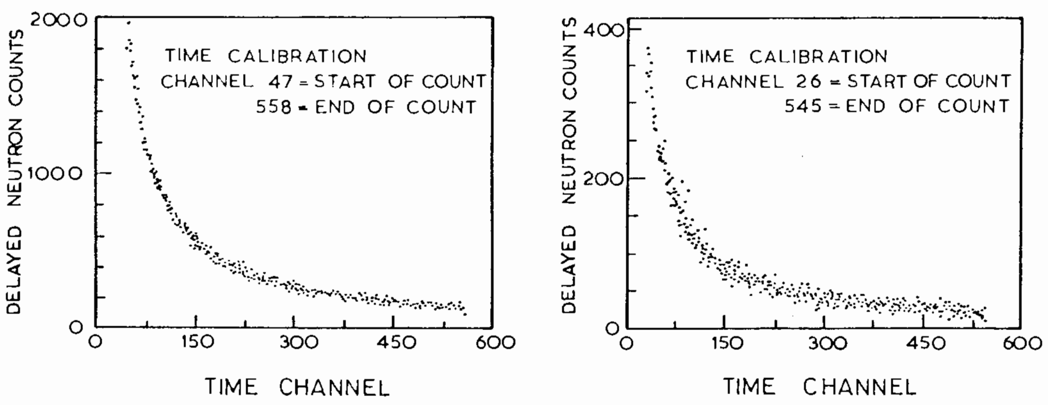
Sample 2 (44.58 mm dia.): Analysis in progress.

Primary neutrons from 2.5 MeV deuterons on Be. Published fractional yields[6] used in the analysis.

Table 2: Fractional Yields of delayed neutrons from fast fission in natural uranium

Precursor group	Fractional Yield (%) Sample 1	 Sample 2
1	1.5±0.9	1.4±0.5
2	15.6±2.8	12.6±1.4
3	21.6±3.8	14.3±1.7
4	39.6±6.6	36.2±5.2
5	14.2±3.9	27.9±8.1
6	7.5*	7.5*

* Taken from published data.



(a) 44.58 mm uranium (b) 25 mm uranium

Fig. 6 Time dependence of delayed-neutron counts from natural uranium.

Measurement of delayed-neutron energies

All our energy measurements mainly for fast fission in ^{235}U, have been undertaken with two ^{3}He spectrometers, each based on a cylindrical gridded ionisation chamber filled with a mixture of helium-3, argon, and methane at partial pressures quoted initially by the manufacturers as 4.2 atm., 2.1 atm. and 0.3 atm. respectively for the first chamber (Technion) and 6.0 atm., 3.0 atm., 0.45 atm. for the second (Seforad). Some of the work was presented at the Specialists' Meeting and the main feature then was the effect of changes in the energy of the neutrons causing fission on the energy spectra of the delayed neutrons. A comparison made of data obtained by single-parameter (pulse height) and dual-parameter (pulse height and rise-time) recording systems showed differences in spectral shape but only at energies below about 250 keV. Since that meeting, the unfolding procedure developed for single-parameter measurements has been applied to a pulse-height distribution obtained in 1980 with the Seforad detector for fission in ^{239}Pu produced by 2.2 MeV neutrons from bombardment of a tritium gas target with 3 MeV protons: the plutonium (^{239}Pu) sample was a cylinder 26.8 mm long with a radius of 6.2 mm and a weight of 62.9 g. There would have been contamination of the neutron spectrum by prompt neutrons from multiplication in the sample, and two methods have been used to correct for it. Firstly the multiplication has been calculated and linked with the counter efficiency for prompt neutrons to give the fraction of counts attributable to them; secondly it has been assumed that all counts at energies above 1.6 MeV are due to prompt neutrons, and the contribution below that energy has then been calculated from the known prompt spectrum. The uncertainties in each method have not been fully analysed and therefore the spectrum in Figure 7, obtained by the second method, should be regarded as preliminary.

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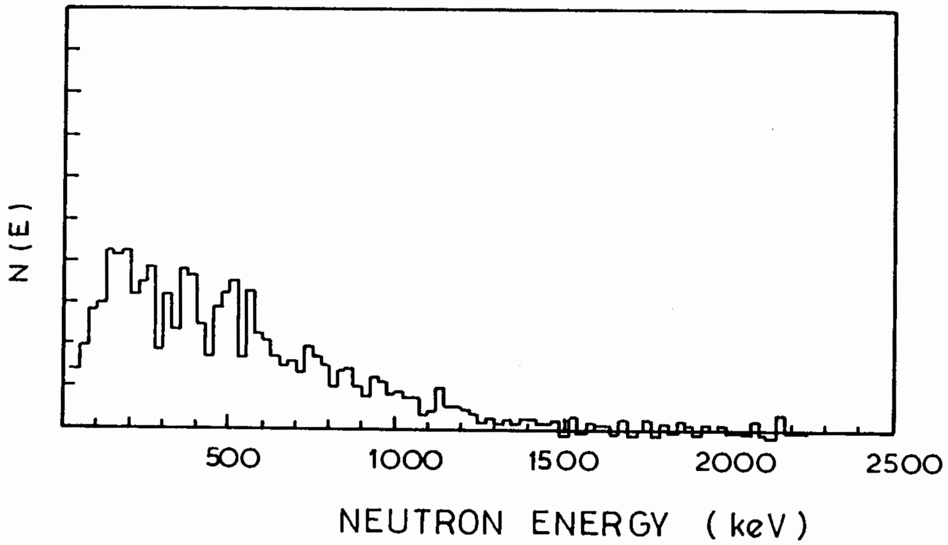

Fig. 7 Delayed neutron spectrum from fast fission in ^{239}Pu.