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INVESTIGATION OF BINARY AND TERNARY FISSION IN ^{238}U INDUCED BY MONOENERGETIC NEUTRONS

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Abstract: Absolute yields have been determined for 34 binary fission products, representing 29 mass chains created during the fission of ^{238}U with monoenergetic neutrons. The neutron energies used for fission ranged from 1.75 MeV to 5.8 MeV. Neutron fluences were determined with a calibrated fission chamber along with metal foils as secondary flux monitors. Fission product activities were measured with a large, hyperpure Ge detector by direct counting of an irradiated ^{238}U foil. In general, fission yields were determined from several counts at different cooling times, using a least-squares technique to unfold multi-component peaks.

The yields of tritons and long range alpha (LRA) particles in the ternary fission of ^{238}U induced by fast, monoenergetic neutrons were determined with a ΔE-E detector telescope technique. A thin ^{238}U deposit was bombarded with neutrons at energies between 3.6 MeV and 4.12 MeV in a vacuum chamber. Total fission events were monitored with a separate surface barrier detector. A mass versus energy spectrum was obtained of the light fragments using suitable electronics. The resolution of the system decreased with irradiation time due to neutron degradation of the ΔE detector, limiting the number of events that could be detected.

(fission, mass yields, ternary fission, ^{238}U, monoenergetic neutrons, detector telescope)

Introduction

A survey of the literature reveals a lack of data on fission product mass distributions for monoenergetic, neutron-induced binary fission of ^{238}U, particularly as a function of neutron energy. Several authors$^{1-4}$ have determined the relative or absolute yield of some fission fragments at neutron energies in the range 1.5 to 18 MeV. In most cases, however, there is only one reported value for the fission yield. With the development of the fast breeder programme, the user requirement for fast yield data has become more important and in some cases values with an uncertainty of ±1% for a fast reactor fission yield may be necessary.

It is hoped that investigations of the emission of the light charged particles from ternary fission would provide unique information about the fission process, particularly the moment of scission. In the case of ternary fission in fertile material, especially ^{238}U, there are very limited experimental data available and some of these are inconsistent with each other. Of particular practical importance are tritium production rates to the development of fast reactors because tritium may be produced at a greater rate in fast reactors than in thermal ones.

Experimental Techniques

Binary Fission

The investigation of mass distribution following the fission of ^{238}U was carried out using discs of natural uranium which contained 99.78% of ^{238}U. These discs were 10 mm diameter and 0.178 mm thick.

Neutrons for irradiation were produced from standard reactions i.e. ^{3}H(p,n)^{3}He or ^{2}H(d,n)^{3}He. Accelerated beams of protons or deuterons were available from the 3 MV Dynamitron accelerator in the Department of Physics Radiation Centre, University of Birmingham. Solid targets of tritium or deuterium absorbed in titanium were used, with the titanium evaporated onto a silver backing disc. During irradiations the silver backing was water cooled. Details of the target assembly and irradiation conditions are given in Ref. 6. The above reactions allowed neutrons in the energy range 1.75 to 6.0 MeV to be produced.

The primary neutron fluence monitor was a specially designed fission chamber^{5}. The ^{238}U foil, along with secondary fluence monitor foils was located in a recess in the fission chamber, and the foils and the chamber were located, 20-30 mm from the neutron-producing target. Monitor foils selected were indium and rhodium and a foil pack was constructed in the form of a multi-layer sandwich of thin discs wrapped in thin aluminium with the ^{238}U disc sandwiched between aluminium catcher foils to retain fission products ejected from the disc. The whole pack was covered with a cadmium sheet in order to prevent any thermal neutrons from reaching the ^{238}U disc.

Since fission product yields were determined by a purely instrumental technique the detection efficiency for both short- and long-lived fission products was optimised by using both short (20 min to 3h) and long (6-24h) irradiation times at each neutron energy. The gamma ray spectometer system used in this work was based on a hyperpure Ge detector with an efficiency of 15%. The absolute efficiency of the detector had previously been determined by us. Fission yields determined were cumulative yields. Following irradiation, the sample pack was dis-assembled and the ^{238}U disc and the two Al catcher foils were separately counted with the Ge detector. Several counts at varying intervals of time were made for each in order to maximise the information obtained on the fission products. The complex γ-ray spectra were analysed with the SAMPO80 program, modified to run on a Hewlett-Packard 2100A computer^{6}. Peak areas obtained with this program were corrected for both self-absorption and coincidence summing effects of the γ-rays^{7}. To enhance the statistical accuracy in the determination of the fission product γ-ray activities, a large number of 4096-channel spectra were recorded over a sufficient period of time to encompass the wide range of half-lives involved. Measured γ-ray activities were assigned to the relevant fission product from their energies and half-lives.

Ternary Fission

A ΔE-E counter telescope of two silicon surface barrier detectors was used to identify the particles emitted in ternary fission and to measure their yield and energy by means of analogue signal processing. The telescope was located inside a vacuum chamber specially designed for this experiment.

The experimental arrangement within the vacuum chamber is illustrated in Fig. 1. The chamber was 40 cm diameter and was provided with two, independent, radial rotating arms, A and B, which were pivoted about the centre. Detectors could be mounted on these arms and clamped in appropriate position. The neutron-producing target assembly was also located inside the chamber at the end of a Dynamitron beam line. The distance of the neutron target from the centre of the chamber was adjustable. At the centre of the chamber was located a ^{238}U fission foil on a special mount. The centres of the neutron-producing target and the ^{238}U fission foil were

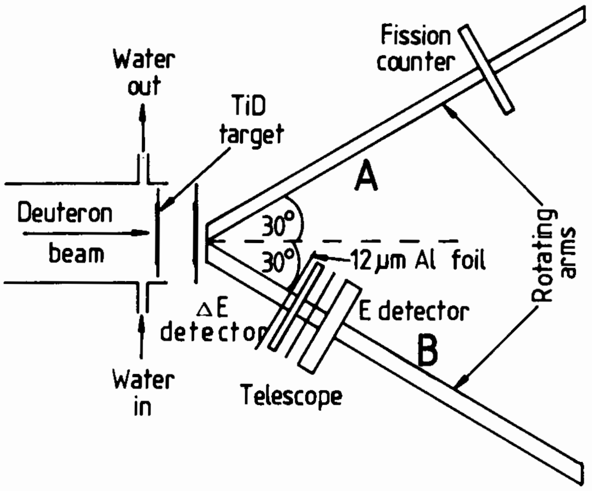

Fig 1 Schematic diagram of the experimental arrangement in the vacuum chamber for the ternary fission of ^{238}U.

aligned on the axis of the chamber and adjusted to be at the same height. The arms carrying the detectors were placed at 30° to the beam direction, either side of the fission foil. One arm carried the ΔE−E telescope whilst the other one carried a surface barrier fission detector for determining the total fission events. The detectors were perpendicular to the arm and were carefully adjusted so that their centres were at the same height as those of the neutron target and fission foil. Throughout the experiment the chamber was maintained at a pressure of 10^{-5} torr.

The neutron-producing reaction used was $^2H(d,n)^3He$ and the target assembly was identical to that used in the binary experiments. Neutron energies of 3.6 and 4.1 MeV were used.

The ^{238}U foil was prepared by electrospraying highly depleted ^{238}U onto a 7µm thick gold coated, polymide foil to form a deposit 500 µg/cm² thick and 20 mm diameter. The complete target was mounted on an aluminium annulus. The thickness of the ^{238}U deposit used was a compromise between a target thin enough not to seriously degrade the energy of the emitted low mass fragments but thick enough to produce a reasonable number of fission events. The neutron target to ^{238}U foil distance was 15 mm.

The electronics used with the telescope to obtain the requisite mass vs energy spectrum were standard and have been previously described[8]. The key to the mass resolution is a Particle Identification Unit (PIU)[9]. The principle of operation of the PIU is based on the relationship between the type of charged particle and the rate of energy loss during its passage through matter, and the following relationship applies:

$$\Delta E . E'_T = \text{constant}.MZ^2$$

where

ΔE (−dE/dx.t) is the energy deposited in a thin transmission detector (of thickness, t)

E'_T is the energy deposited in a stopping (E) detector

M is the mass of the particle

Z is the charge on the particle.

The signal from the PIU ("mass" signal) and the sum of the signals from the E and ΔE detectors ("total energy" signal) were input to two ADCs operating in coincidence. The output of these two ADCs was then displayed via a MCA system as a two dimensional, mass vs energy spectrum.

The ΔE detector was fully depleted, with a thickness of about 50 µm and an active area of 75 mm². The E detector had a thickness of 800 µm and an active area of 400 mm² and an initial energy resolution of 36 keV for 6.05 MeV α particles. The fission detector was a partially depleted detector, with an active area of 50 mm² and thickness of 100 µm.

The telescope was located 25 to 50 mm from the ^{238}U foil whilst the fission detector was 120 mm distant. A 12 µm Al foil was placed between the ^{238}U target and the ΔE detector to prevent binary fission fragments from reaching the ΔE detector and so degrading its energy resolution. The detector resolution was monitored throughout the course of an irradiation by the use of an annular ^{241}Am source permanently mounted between the ΔE and E detectors.

Irradiations at each of the two neutron energies investigated (3.6 and 4.1 MeV) were carried out for about 36 hours with a flux of approximately 5 x 10^7n/cm²/s.

The complete counting system was checked initially by replacing the neutron source with a ^{252}Cf source and detecting the light charged particles emitted in the spontaneous ternary fission of this nuclide. For the neutron irradiations the discriminators on the timing single channel analysers were set at levels sufficient to prevent counting of particles produced by events other than ternary fission i.e. (n,particle) reactions, but low enough to allow most of the light ternary fragments to be detected.

Results and Discussion

Binary Fission

Absolute fission yields were based on a determination of the saturation activity, S, defined from the standard activation equation[10]. The absolute, saturation disintegration rate was determined from the corrected peak area, counting time, counting efficiency, absolute emission intensities, possible precursor activity and irradiation conditions (time, flux and flux variations). Fission yields could then be calculated from the simple formula

Fission Yield = Saturation Activity/Fission Rate

Fission rate refers to the count rate of the fission chamber or indirectly from the activity of the indium or rhodium monitor foils. Fission yields so determined were corrected for the following:

i) variations in the neutron flux during irradiation,
ii) decay of precursors of the nuclide under observation during the irradiation and cooling periods, and
iii) independent yields of members of the decay chain under investigation.

One of the major sources of error in determining the fission yields from γ-ray spectra lies in the values taken for the absolute γ-emission intensities. This is an inherent error dependent on the best available, published data. To minimise the overall experimental error data for each of the fission products was taken from more than 20 spectra at each neutron energy and a weighted, least squares analysis applied to these data to determine the activity for each fission product at a particular γ-ray energy. The fission product yields obtained in this way are given in Table 1 for three neutron energies. Comparison of this data with that of Nagy et al[4] indicate that our data is generally in good agreement with theirs.

Ternary Fission

During the experiments a considerable number of background counts were recorded by the telescope close to the triton and alpha particle regions. A two-dimensional spectrum of energy vs mass recorded in the manner described above is shown in Fig. 2. Figure 2(a) represents a raw spectrum including background counts whilst Fig. 2(b) is the same spectrum after removing these background counts. The spectrum was recorded continuously throughout the irradiation but was stored about every 2 hours. These spectra indicated a relatively constant value for the ratio of tritons to alpha particles, but progressive

Table 1. Recommended fission product values based on the experimental values determined at three neutron energies

Fission Product	Neutron Energy (keV) 1722	3726	5982
^{85m}Kr	0.71 ± 0.07	0.80 ± 0.02	1.06 ± 0.14
^{87}Kr	1.22 ± 0.15	1.38 ± 0.16	1.54 ± 0.20
^{88}Kr	1.61 ± 0.18	1.77 ± 0.19	2.04 ± 0.23
^{89}Rb	3.0 ± 0.4	2.9 ± 0.5	3.2 ± 0.4
^{91}Sr(Y)	3.8 ± 0.4	3.59 ± 0.22	3.6 ± 0.3
^{92}Sr	4.4 ± 0.5	4.9 ± 0.5	3.7 ± 0.4
^{93}Y	5.2 ± 0.5	5.2 ± 0.6	4.3 ± 0.5
^{94}Y	4.4 ± 0.5	4.6 ± 0.6	4.9 ± 0.5
^{95}Zr	5.4 ± 0.6	5.8 ± 0.6	5.4 ± 0.6
^{97}Zr(Nb)	5.3 ± 0.4	5.7 ± 0.4	5.4 ± 0.4
^{101}Tc	7.8 ± 0.9	7.5 ± 0.8	6.3 ± 0.7
^{103}Ru	6.7 ± 0.7	6.3 ± 0.6	-
^{104}Tc	4.8 ± 0.5	4.1 ± 0.5	3.9 ± 0.5
^{105}Ru(Rh)	4.54 ± 0.26	4.2 ± 0.4	4.33 ± 0.29
^{107}Rh	0.67 ± 0.20	0.81 ± 0.18	1.17 ± 0.15
^{129}Sb	0.44 ± 0.07	0.62 ± 0.08	1.11 ± 0.12
^{131}I	3.6 ± 0.4	3.7 ± 0.4	3.7 ± 0.4
^{132}I	4.98 ± 0.29	4.6 ± 0.4	4.74 ± 0.22
^{133}I	7.1 ± 0.7	6.5 ± 0.7	6.9 ± 0.8
^{134}Te	6.6 ± 0.7	6.1 ± 0.8	5.3 ± 0.6
^{135}I	7.1 ± 0.4	7.0 ± 0.6	6.3 ± 0.4
^{138}Cs(Xe)	6.2 ± 0.7	5.6 ± 0.4	6.0 ± 0.5
^{140}Ba(La)	5.7 ± 0.4	5.6 ± 0.4	5.9 ± 0.4
^{141}Bu	5.0 ± 0.7	5.0 ± 0.8	5.2 ± 0.6
^{142}La	5.0 ± 0.6	5.1 ± 0.5	4.6 ± 0.5
^{143}Ce	4.6 ± 0.6	4.4 ± 0.5	4.6 ± 0.6
^{146}Ce	3.4 ± 0.3	3.6 ± 0.3	3.6 ± 0.3
^{147}Nd	2.6 ± 0.4	2.8 ± 0.3	2.9 ± 0.3
^{149}Nd	1.65 ± 0.25	1.84 ± 0.22	1.61 ± 0.20

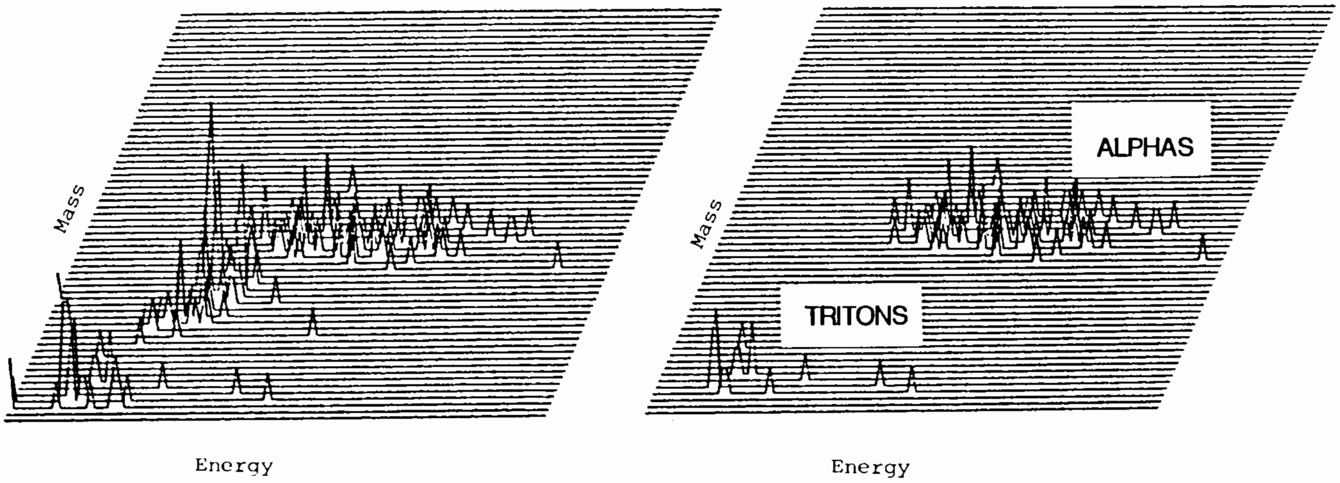

Fig 2 Mass vs energy spectrum for ternary fission products from ^{238}U obtained using the Particle Identification Unit (see text). The left-hand diagram represents a raw spectrum, including background counts. The right-hand one shows the same spectrum after removing the background.

neutron damage to the E detector resulted in a blurring of the boundaries between the two particles. Consequently, data from the later irradiation periods were not included in the final results.

Because the total fission detector and the telescope were at different distances from the ^{238}U foil, it was necessary to correct for differences in the geometrical counting efficiencies for these two detector systems. The correction was made by placing identical sheets of mica adjacent to the ΔE detector and to the fission detector. The mica records the fission events as tracks which can be counted after a suitable etching procedure[7]. From the track density found in each mica sheet, the ratio of these densities along with the known angular anisotropy for neutron-induced fission of ^{238}U[7] could be used to determine this correction factor.

In Tables 2 and 3 are summarised all the available data regarding yields of tritium and α particles from fast neutron-induced fission of ^{238}U. In the case of tritium the only reported value obtained experimentally is that of Buzzelli et al[11], using a fast reactor neutron spectrum and their reported value of 9 x 10^{-4} tritons/fission event is four times greater than that theoretically calculated[13]. The mean value from the two neutron energies for our

Table 2. Summary of values for the ratio of binary fission events to triton emission in the neutron-induced ternary fission of ^{238}U

Neutron energy (MeV)	Binary events/ tritons emitted	Method of measurement	Reference
Fast fission spec.*	434.7 ± 14	radiochemical	Buzzelli *et al*[11]
Fast fission spec.**	1000 ± 29	"	"
Fast fission spec.	384	"	"
Fission spectrum	9000	calculated	Rider[12]
Fission spectrum	3840	"	ANL[13]
3.6	5650 ± 1410	detector telescope	This work
4.1	4220 ± 1050	"	"

* Location of irradiation at lower blanket, 42 cm from core mid-plane

** Location of irradiation at core, 5 cm from core mid-plane.

Table 3. Summary of values for the ratio of binary fission events to α-particles emitted in the neutron-induced ternary fission of ^{238}U

Neutron energy (MeV)	Binary events/ α-particles emitted	Method of measurement	Reference
Fission spec.	645	calculated	Rider[12]
2.5	1103 ± 28	experimental	Nagy *et al*[14]
2.5	600	"	Solovena[15]
2.5	4550 ± 350	"	Drapchinskii *et al*[16]
3.6	884 ± 86	detector telescope	This work
4.1	869 ± 98	"	"
14	795 ± 35	experimental	Nagy *et al*[14]
14	3750 ± 270	"	Drapchinskii *et al*[16]
14	1050 ± 100	"	Perfilov *et al*[17]

data is seen to be close to this theoretical value (2.0 ± 0.4 x 10^{-4} t/f). The high values reported by Buzzelli *et al* may be due to tritium contamination from other sources in the reactor used.

It is difficult to establish any neutron energy dependence of the observed yield due to lack of published data for other neutron energies. Our results, however, for two neutron energies indicate that the triton yield does increase with increasing energy. For the yield of α particles in ternary fission several data obtained experimentally by other authors[14-17] along with the results obtained in this work are shown in Table 5. The reported data are statistically inconsistent and there is considerable spread in the values reported for the same neutron energy. Bearing this in mind there would appear to be no marked trend of α-particle yield as a function of neutron energy.

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