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MEASUREMENT OF THE  $^{235}\text{U}/^{238}\text{U}$  FISSION CROSS SECTION

RATIO IN THE  $^{235}\text{U}$  FISSION NEUTRON SPECTRUM

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MEASUREMENT OF THE  $^{235}\text{U}/^{238}\text{U}$  FISSION CROSS SECTION  
RATIO IN THE  $^{235}\text{U}$  FISSION NEUTRON SPECTRUM

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## ABSTRACT

Fission cross section ratio of  $^{235}\text{U}$  to  $^{238}\text{U}$  has been measured in the fast neutron field generated by the  $^{235}\text{U}$  fission plate installed on the thermal column of the Tehran Research Reactor (TRR) with a Makrofol solid state nuclear track detector. The experiments were carried out with a set of total six enriched  $^{235}\text{U}$  and depleted  $^{238}\text{U}$  deposits with different masses and Makrofol films of 0.025 mm and 0.060 mm thicknesses. The chemically etched tracks were counted by an optical microscope. No significant differences were observed with the thin and the thick films. The results showed that the average fission cross section ratio is  $3.83 \pm 0.25$ .

### 1. INTRODUCTION

The fast neutron field installed on the thermal column cavity of the Tehran Research Reactor (TRR) is generated by an  $^{235}\text{U}$  fission plate. The plate 0.25 mm thick and 230 mm in diameter is made of 93% enriched uranium with the uniformity of about 10%. The plate has been supplied by the U.K. Atomic Energy Research Establishment (AERE), Harwell, and installed during 1978. Aluminium, iron and copper were tried for fission plate cladding. Because of some difficulties with aluminium soldering, and those of the long half-life (44.5 days) of  $^{56}\text{Fe}$ , copper has finally been chosen.

The mechanism of fast neutron generation is that the neutrons originated from the reactor core, are slowed down in the graphite thermal column and are collimated after passing through the rectangular hole of 100 x 100 x 850 mm. The well-thermalized collimated neutron beam impinges on the fission plate and produces fission neutrons. In order to reduce the gamma dose rate from the fission plate, the lead stacking, 150 mm thick, have been used.

This report describes the measurement of  $^{235}\text{U}/^{238}\text{U}$  fission cross section ratio in the TRR fast neutron field with Makrofol solid state nuclear track detector (SSNTD).

### 2. EXPERIMENTAL MEASUREMENTS

A selection of six deposits was exposed at the TRR fast neutron field. The deposits were specifically prepared for the University of Tehran at the U.K. AERE, Harwell. The deposits were 20-mm-diam vacuum evaporated on backings of 32.0-mm-diam polished platinum, 0.125 mm thick. The isotopic concentration and mass assay of the fissionable deposits exposed at the TRR fast neutron field, supplied by the AERE, are shown in Table I. The diameter of each deposit was determined from the autoradiography and from the microscope traverse along the fission source.

The detectors were 12 circular Makrofol KG films with the diameter slightly greater than that of the backing of the deposits to make the removing easier. Six of the films were 0.025 mm thick (thin films) and the rest were 0.060 mm thick (thick films). The Makrofol SSNTDs were etched simultaneously at

Table I. The fissionable deposits exposed at the TRR fast neutron field

Foil Identification and Principal Isotope	Deposit Diameter (mm)	Isotope Concentration (at %)	Mass of Principal Isotope ( $10^{-9}$ kg)
UT-51 $^{235}\text{U}$	$20.0 \pm 0.1$	$^{235}\text{U}$ :93.197; $^{234}\text{U}$ :1.179 $^{236}\text{U}$ : 0.250; $^{238}\text{U}$ :5.435	$79.7 \pm 0.4$
UT-52 $^{235}\text{U}$	$20.1 \pm 0.1$	"	$264.8 \pm 1.3$
UT-53 $^{235}\text{U}$	$20.1 \pm 0.1$	"	$729.5 \pm 4.0$
UT-81 $^{238}\text{U}$	$20.0 \pm 0.1$	$^{238}\text{U}$ :99.995; $^{235}\text{U}$ :0.005	$81.9 \pm 2.5$
UT-82 $^{238}\text{U}$	$20.1 \pm 0.1$	"	$276.1 \pm 8.3$
UT-83 $^{238}\text{U}$	$20.4 \pm 0.1$	"	$777 \pm 23$

328 K for 95 min in the solution of 25% (6.25 N) NaOH. The etched films were then counted by an optical microscope with an overall mag. of 400.

The specimen holder has been made of three cadmium plates, 1 mm thick, in the shape of equilateral triangles, each side 100 mm long. One of the plates which is placed between the other two, has three holes, 32 mm in diameter, the same as of the backings. The deposits are positioned back-to-back with the Makrofol SSNTD placed on their fissionable materials. This sandwich of deposit and SSNTD is placed between two cadmium plates of specimen holder. This arrangement prevented the deposits to slip and also held the SSNTD in intimate contact with the deposits. Figure 1 shows the schematic view of the specimen holder with two uranium deposits and two Makrofol films. The cadmium specimen holder is mounted on the main support, the plexiglass plate, next to the fission plate. The gold foil used for reactor power monitoring is placed at the top corner of the plexiglass plate.

### 3. RESULTS AND DISCUSSION

The first part of the experiments was to determine the thermal neutron flux within the TRR cavity in order to choose the suitable reactor power and irradiation time. The thermal neutron flux was measured using gold foils. The results showed that the mean cadmium ratio in the cavity is  $R_{-2}(\text{Cd}) = 253 \pm 4.1\%$  and that the thermal neutron flux is  $(2.61 \pm 0.01) \times 10^{12} \text{ m}^{-2} \text{ s}^{-1}$  for reactor power of 1 MW.

Several experiments were carried out in the TRR fast neutron field with the Makrofol SSNTD. In these experiments three pairs of enriched and depleted uranium deposits and Makrofol films were placed in the specimen holder.

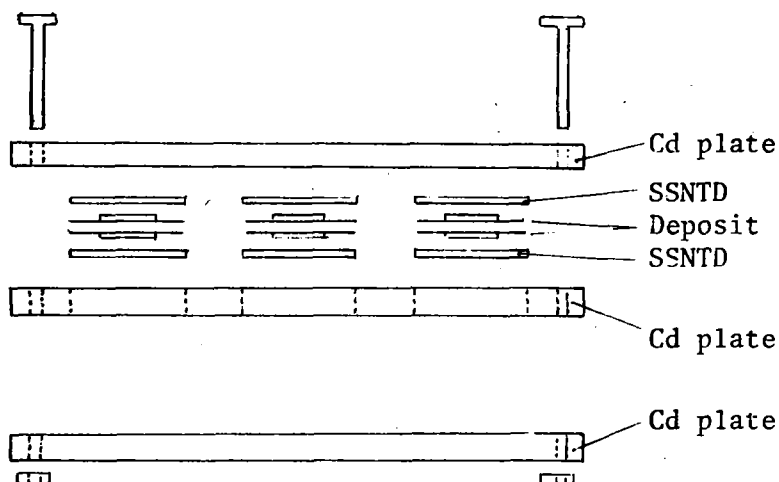


Fig. 1 The schematic view of the specimen holder with two uranium deposits and two Makrofol films.

The experiments were carried out at low reactor power of 100 kW for 60 min to avoid track pile-up and excessive overlap. This led to track density of about  $4 \times 10^8$  tracks/m<sup>2</sup> with the statistical error of about 2% for the thickest enriched deposit.

The correction typically applied to the isotopic fission rates are summarized in Table II. These corrections include absorption of fission fragments in the deposits, fission in other isotopes, neutron scattering and absorption, and flux perturbation by the Makrofol.

The fission fragment absorption was calculated<sup>(1)</sup> on the basis of 0.69% per  $10^{-5}$  kg U/m<sup>2</sup> in UO<sub>2</sub>. The corrections for fission in isotopes other than the principal one are based on the fact that<sup>(2)</sup> in the fast neutron field the tracks are produced only by fission of <sup>235</sup>U and <sup>238</sup>U. Considering the isotopic concentration of the deposits and taking the average fission cross section<sup>(3)</sup> of  $1.233 \pm 0.52\%$  and  $0.2967 \pm 3.25\%$  for the <sup>235</sup>U and <sup>238</sup>U, respectively, the corrections are 0.9863 and 0.9998 for the <sup>235</sup>U and <sup>238</sup>U, respectively. The effect of neutron scattering and absorption due to the thermal column cavity wall was assumed to be negligible since a well-collimated neutron beam impinges on the fission plate, and that due to platinum backing of the deposits is small and could be neglected. It has been shown<sup>(4)</sup> that the perturbation of flux caused by the hydrogen in the Makrofol is about 1%. Hence, the corrections are  $0.995 \pm 0.005$  and  $1.005 \pm 0.005$  for the <sup>235</sup>U and <sup>238</sup>U, respectively.

The combined uncertainty ascribed to the observed number of tracks per unit area (Table III) arises from track counting statistics, background, exposure timing, irradiation positioning, imperfect contact between deposit and SSNTD, radial uniformity of the fission sources, and calibration of the microscope field of view.

Other possible of systematic errors such as integrity of the deposits, flux

Table II: Typical corrections applied to the isotopic fission rates.

Correction	Isotope					
	$^{235}\text{U}$			$^{238}\text{U}$		
	Foil					
	UT-51	UT-52	UT-53	UT-81	UT-82	UT-83
Absorption in deposit	1.0018±.0035	1.0058±.0035	1.0159±.0040	1.0018±.0035	1.0061±.0035	1.0171±.0043
Fission in other isotopes	.9863±.0005	.9863±.0005	.9863±.0005	.9993±.0005	.9998±.0005	.9998±.0005
Neutron scattering and absorption	1.000 ±.001	1.000 ±.001	1.000 ±.001	1.000 ±.001	1.000 ±.001	1.000 ±.001
Flux perturbation by Makrofol	.995 ±.005	.995 ±.005	.995 ±.005	1.005 ±.005	.005 ±.005	.005 ±.005
Combined uncertainty of corrections	±.62%	±.62%	±.62%	±.62%	±.62%	±.66%

Table III. Track densities of Makrofol SSNTD irradiated at the TRR fast neutron field.

Detector	Film Number	Foil Identification and Principal Isotope	Field of View ( $10^{-8} \text{ m}^2$ )	Number of Frames	Total Count	$10^4 \text{ track/m}^2$				
						Observed	Relative Statistical Error (%)	Combined Uncertainty of Track Densities (%)		
Thin Makrofol (0.025 mm)	151	UT-51	$^{235}\text{U}$	9.2416	221	1060	5191	$\pm 3.1$	$\pm 3.3$	
	181	UT-81	$^{238}\text{U}$	9.2416	210	264	1360	$\pm 6.2$	$\pm 6.3$	
	152	UT-52	$^{235}\text{U}$	9.2416	132	2424	19871	$\pm 2.0$	$\pm 2.3$	
	182	UT-82	$^{238}\text{U}$	9.2416	121	642	5741	$\pm 3.9$	$\pm 4.0$	
	153	UT-53	$^{235}\text{U}$	3.3489	210	2933	41705	$\pm 1.8$	$\pm 2.1$	
	183	UT-83	$^{238}\text{U}$	3.3489	210	704	10010	$\pm 3.8$	$\pm 4.0$	
	Thick Makrofol (0.060 mm)	251	UT-51	$^{235}\text{U}$	9.2416	195	831	4611	$\pm 3.5$	$\pm 3.6$
		281	UT-81	$^{238}\text{U}$	9.2416	196	218	1204	$\pm 6.8$	$\pm 6.9$
252		UT-52	$^{235}\text{U}$	9.2416	208	3444	17916	$\pm 1.7$	$\pm 2.0$	
282		UT-82	$^{238}\text{U}$	9.2416	210	945	4869	$\pm 3.3$	$\pm 3.5$	
253		UT-53	$^{235}\text{U}$	3.3489	224	3131	41738	$\pm 1.8$	$\pm 2.1$	
283		UT-83	$^{238}\text{U}$	9.2416	121	1133	10132	$\pm 3.0$	$\pm 3.2$	



perturbation by the specimen holder, track overlap, variation of track density with etching time and temperature, and uncertainty in the etchant concentration are all assumed to be negligible. The summary of possible sources of systematic errors are given in Table IV. The results of the  $^{235}\text{U}/^{238}\text{U}$  fission ratio are shown in Table V. The uncertainties (1 sd) assigned to the  $^{235}\text{U}/^{238}\text{U}$  fission ratios include:

1. Uncertainties in mass per unit area (Table I)
  - a. mass assay errors
  - b. deposit area
2. Uncertainties in corrections (Table II)
  - a. absorption in deposit
  - b. fission in other isotopes
  - c. neutron scattering and absorption
  - d. flux perturbation by Makrofol
3. Uncertainties in count ratios (Table V)
  - a. counting statistics
  - b. background
  - c. calibration of the microscope field of view

Table IV. Possible sources of systematic errors.

Description	Error (%)
1. Mass assay (UT-51)	±0.50
2. Deposit area (UT-51)	±1.0
3. Background	±1.0
4. Exposure timing	nil
5. Irradiation positioning	nil
6. Imperfect contact between deposit and SSNTD	nil
7. Radial uniformity of fission sources	nil
8. Calibration of the microscope field of view	
Large graticule	±0.33
Small graticule	±0.55

Table V.  $^{235}\text{U}/^{238}\text{U}$  fission ratio measured in the TRR fission-neutron spectrum.

Detector	Foil Identification and Principal Isotope	Count Ratio	$^{235}\text{U}/^{238}\text{U}$ Fission Ratio
Thin Makrofol (0.025 mm)	UT-51 $^{235}\text{U}$ UT-81 $^{238}\text{U}$	3.8172±7.1%	3.78±0.30
	UT-52 $^{235}\text{U}$ UT-82 $^{238}\text{U}$	3.4609±4.6%	3.48±0.20
	UT-53 $^{235}\text{U}$ UT-83 $^{238}\text{U}$	4.1663±4.5%	4.15±0.23
	UT-51 $^{235}\text{U}$ UT-81 $^{238}\text{U}$	3.8297±7.8%	3.79±0.32
	UT-52 $^{235}\text{U}$ UT-82 $^{238}\text{U}$	3.6799±4.5%	3.70±0.19
	UT-53 $^{235}\text{U}$ UT-83 $^{238}\text{U}$	4.1193±3.8%	4.10±0.20
Average			3.83±0.25

Table V shows that there is no significant difference with the thin and the thick films, and that the average fission cross section ratio is  $3.83 \pm 0.25$ .

The comparison of the  $^{235}\text{U}/^{238}\text{U}$  fission ratios in the  $^{235}\text{U}$  thermal fission spectrum of the TRR cavity with similar estimates  $^{235}\text{U}$  (S-10) is shown in Table VI. The results show that the agreement between the present work and other experimentally measured fission ratios is to within 3%.

The correction for fission in other isotopes (Table II) depends on the assumption of the fission cross section for  $^{235}\text{U}$  and  $^{238}\text{U}$ . The values of cross sections of six different references (Table VI) were used to estimate this correction. The results showed that the sensitivity of the average  $^{235}\text{U}/^{238}\text{U}$  fission ratio to the values of cross sections used in the correction is less than 0.4%.

#### 4. CONCLUSIONS

The  $^{235}\text{U}/^{238}\text{U}$  fission cross section ratio was measured in the TRR fast neutron field using Makrofol SSNTD. The track detectors were irradiated with the UT enriched and depleted deposits, and after the etching they were

Table VI. Comparison of the  $^{235}\text{U}/^{238}\text{U}$  fission cross section ratio in the  $^{235}\text{U}$  fission neutron spectrum.

Authors	$^{235}\text{U}/^{238}\text{U}$ fission ratio
Grundl, 1968 <sup>(5)</sup>	3.85±0.23
Fabry et al., 1970 <sup>(6)</sup>	3.78±0.18
Grundl, 1972 <sup>(7)</sup>	3.71±0.17
McElroy and Kellogg, 1975 <sup>(8)</sup>	3.82±0.24
Fabry et al., 1978 <sup>(9)</sup>	3.94±0.08
Cullen et al., 1982 <sup>(10)</sup>	4.05*
This work	3.83±0.25

\*  $\sigma_f(E)$ : ENDF/B-V

counted by an optical microscope. The average  $^{235}\text{U}/^{238}\text{U}$  fission cross section ratio in this fission-neutron spectrum was found to be 3.83±0.25. Better results can almost certainly be achieved by counting the whole area of the exposed track detector. This is time consuming and difficult, but appears necessary if the best results are to be obtained.

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