Nuclear Data for Science and Technology (1988 MITO), 123- 126, Copyright © 1988 JAERI.

MEASUREMENT OF ABSOLUTE FISSION RATE OF NATURAL, DEPLETED URANIUM AND THORIUM AND MICROSCOPIC CROSS SECTION RATIOS IN THE RADIAL BEAM OF THE RA3.

M. Bovisio de Ricabarra, L. Cohen de Porto, D. Waisman, G. H. Ricabarra

Argentine National Atomic Commission
Avda. Libertador 8250, Buenos Aires, Argentina

Abstract: The capability to measure absolute fission rates per nuclei has been established to a precision level of around ± 2%. Thin deposits of ThO2 and U3O8 were prepared by molecular plating and the isotopic masses determined by two techniques. A miniature fission chamber and SSNTD fission fragment detection techniques were used in the radial neutron beam of the RA3. Fission fragments selfabsorption in the deposits are corrected. The results of this investigation are: 1. Optical efficiency of SSNTD detectors. 2. The microscopic integral fission cross section ratio of Th232 to U238 by two independent techniques. 3. Experimental ratios of natural, depleted uranium and thorium fission rate per μg to Mn56 (Mn thermal flux monitor) desintegration rate per μg. The agreement with calculated results is excellent.

(fission fragments detection, integral cross section, fission tracks, neutron spectra, neutron beam, Th232, U238, In115)

Introduction

The objective of this work is the determination of fission rates of U238, natural uranium and Th232 at the radial beam of the RA3 reactor. This facility will be considered by us as a controlled environment benchmark neutron field.

Individual fission fragments are directly recorded as they emerge from a thin deposit of fissionable material. The measurement problem has several important features :

1. Preparation and mass assay of thin fissionable deposits.
2. Fission fragments detection was based in two techniques: a)Miniature fission chamber detection of fission fragments. b)Solid state nuclear track detection of fission fragments.
3. Experiments of fission rate measurements at the radial neutron beam of the RA3.
4. Formalism used for interpretation and data reduction of experimental results.
5. Final results and comparison with calculated values which validates the assumption of the near fission spectrum shape of the fast neutron beam.

Preparation of Thin Fissionable Deposits

All fissionable sources under consideration in this paper are 12.45mm diameter oxide deposits laid down on polished aluminum disks 24.8mm in diameter and 0.1 to 0.2mm thick.

Molecular plating[1, 2, 3] was chosen to ensure adecuate deposit uniformity, for accurate track counting*. Uranium and thorium deposits were 100 to 200 μg/cm^2 thick. Deposits homogeneity was controlled by microscopic inspection. The deposits adherence was controlled by α counting after a process which included soft but firm friction, α counting revealed losses smaller than 1%. Actual manipulation for deposits irradiation is much less than this process, anyhow the possibility of weight losses was controlled by α or gamma counting before and after irradiation.

Mass Assay of Uranium Deposits

The uranium deposits masses were assayed by absolute α emission rate determination.

A depleted uranium deposit (UDE1, 23ppm U235, monel backing) and several natural uranium deposits (UN0, UN2, UN7, UN10) were measured in a 2π gas flow counter working in the chamber region. Several gases were tried and a propane-butane mixture was selected because a reasonable separation of U234 and U238 α peaks was obtained. The electronic for 2π α counting was standard and the events were registered in a 4096 MCA. The integration bias was taken at channel 100 and counts below the bias, ≤0.5%, were obtained by extrapolation of the flat distribution from channel 100 to 200. The background, around 0.006d/s, was substracted and amounts to 0.3% for the less active deposits.

In addition a thin natural uranium deposit (<100μg/cm^2) was measured in a semiconductor α spetrometer and the α desintegration ratio (U234+U235+U238)/U238=2.054±0.5%, was determined.

To get the absolute α desintegration rate corrections, for α selfabsorption in the deposit, f_α, and for the small back-scattering in the backing, f_B, were applied. The α selfabsorption was taken into account using the $(1-t/2R_{U3O8})$ approximation, t is the deposit thickness and the α range in U3O8, R_{U3O8}=10.10mg/cm^2, was calculated by:

$$1/R_{U3O8} = W_U/R_U + W_O/R_O \qquad (1)$$

where W_U, W_O are the weight fractions of U and O in U3O8 and the α ranges, R_U, R_O, were taken from Littmark and Ziegler[4]. The α backscattering correction was taken from Hutchinson et al.[5], where a backscattering study for different backings was made, using a specially designed 1π detector.

The α emission per U μg was calculated using U238 half life[6] and the experimental total to U238 α desintegration ratio. Final results are:

Table 1. Alpha Counting Uranium Mass Assay

Source	f_α	f_B	α (d/s)	U (μg)
UDE1	0.989	1.015	2.114±.007	338.7±.6%
UN0	0.977	1.000	6.106±.025	492.8±.8%
UN2	0.987	1.000	3.516±.022	280.9±.8%
UN7	0.994	1.000	1.756±.006	139.3±.6%
UN10	0.987	1.000	3.419±.016	273.0±.7%

*In collaboration with LUE Lab., CNEA.

Mass Assay of Thorium Deposits

A 0.25mm thick thorium monitor foil (THM3) and the molecular plated deposits (TH12, TH13, TH14) were packed together and irradiated 4 days in the RA3 thermal column ($\phi_{th}=10^{10}$ n/cm^{2}s). After Th233 (22.3 min) decay the 26.95 days Pa233 activity of TH12, TH13, TH14 was measured relative to the same activity of THM3, in a Ge(Li) gamma spectrometer.

The THM3 activity was about 100 times higher than the deposits activity, therefore to minimize random summing error in the 'shape' amplifier, a 1 mm lead absorber was used to suppress U233 X rays and the amplifier time constant was set equal to 1.5μs. To minimize live time error (<1%) the ADC upper and lower level discriminators were set to count only 300, 312, 340, 376, 399 and 416 keV Pa233 photopeaks.

The 312keV photopeak activity ratio, TH13/THM3, was measured at 5.5cm distance from the Ge(Li) detector. Previous measurements at differents distances, from 2.5 to 14.5cm, shown that there were not random summing and live time errors. To obtain the true TH13/THM3 activity ratio the THM3 activity was corrected for thermal neutron flux self-depression, G_{th}, and selfabsorption of the 312keV photopeak, f_g, in the thorium thick foil. $G_{th}=0.993$ includes neutron depression in the foil and border effect[7]. $f_g=0.937$ was calculated assuming exponential approximation, $f_g=(1-e^{-\mu_g d})/\mu_g d$.
A theoretical calculation shows that the error in assuming this approximation is less than 0.5% at 5.5cm from the detector. The 312 keV photonic absorption coefficient, μg, was obtained by measuring the activity ratio with and without an equally thick thorium absorber foil. Finally from the measured ratio and the monitor mass (21.400mg, 99.8% Th) the TH13 mass was determined.

Then TH12 and TH14 masses were determined relative to TH13 by measuring the 312keV photopeak activity ratio at 0.5 and 2.5cm distance from the Ge(Li) detector.

The gamma activity ratios were measured before and after each experience at the RA3 neutron beam. The average ratios and deduced masses are:

Table 2. Gamma Counting Thorium Mass Assay

Source	d(cm)	counting ratio	Th mass (mg)
THM3	5.5	1.	21.357
TH13	5.5	8.812E-3±0.8%	0.1882±1.0%
TH12	2.5	1.1257±0.6%*	0.2119±1.2%
TH14	2.5	0.8488±0.7%*	0.1597±1.2%
TH12	0.5	1.1304±0.6%*	0.2128±1.2%
TH14	0.5	0.8573±0.7%*	0.1614±1.2%

*ratio to secondary monitor (TH13)

Fission Fragments Detection Techniques

a) Miniature Fission Chamber Measurements

The miniature fission chamber is of paralell plate type. The grounded plate is the aluminum electroplated deposit. The chamber has an anode disk paralell to the fissionable deposit and a cylinder which forms the side wall of the chamber. This anode arrangement prevent loss of electric field strength at the active volume edges. To reduce neutron absorption and scattering electrodes and side wall are made of aluminum (fig.1).

The chamber is operated with continous flow of argon. As the maximun number of pulses expected from natural uranium deposits was about 30 c/s no fast electronic was needed and the pulses from the shape amplifier were directly recorded into a 1024 MCA. The pulse height distribution has a very low plateau up to channel 200. The fission data obtained is the integral from channel 40 to 1024 plus zero extrapolation correction(≃0.8% for 200μg/cm^2).

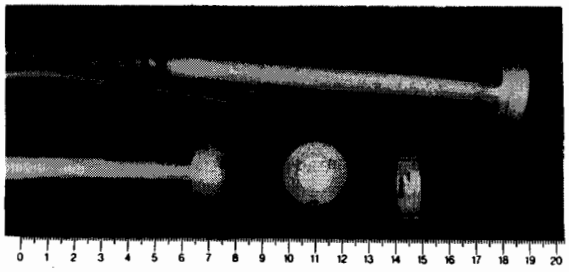

Fig.1 Miniature Fission Chamber

b) Solid State Nuclear Track Detector Measurements

Good optical quality and low fossil tracks background mica was selected for SSNTD detectors.

Mica disks of 23mm diameter and 50μm thick were used. The mica disks were heated up to 580 °C during 24 hours, for annealing of the fossil tracks. The remaining background tracks were enlarged by a 7 hours preetching in HF 49% for easy discrimination from the 'true' tracks[8].

The mica detector in close contact with the fissionable deposit was placed between an aluminum back plate and a ring holder secured by screws. This stack was irradiated at the RA3 neutron beam simultaneously with the fission chamber.

After irradiation the mica detectors were etched with HF 49% for 1 hour to obtain 5μm diameter tracks. The tracks counting was made by two observers and the agreement was better than 1.5%. Counting statistics was 10000 tracks for natural uranium and 5000 tracks for thorium. The tracks were counted along 6 to 10 detector diameters by each observer. Total fission tracks were obtained by 2π integration of the radial tracks distribution.

As we made simultaneous irradiation of fission chamber and SSNTD, by comparing the results from both detectors we get the mica optical efficiency of Table 3.

Table 3. SSNTD Optical Efficiency

EXP	Foil	f_f	f_s	fis/(s•μg) x100	Mica Eff.
CTR1 F.Ch	UN0	.967	.989	7.525±0.3%	1.002
CTR1 SSNTD	UN7	.991	.997	7.533±1.0%	±1.3%
CTR3 F.Ch	UN2	.981	.989	8.235±1.2%	0.996
CTR3 SSNTD	UN10	.982	.997	8.205±1.0%	±1.7%
CTR4 F.Ch	TH13	.988	.993	0.4086±1.7%	0.986
CTR4 SSNTD	TH12	.987	.996	0.4030±1.6%	±2.6%

c) Corrections to Fission Rates Measurements

Fission chamber and SSNTD measurements are nearly 100% efficient, however small corrections for fission absorption in the deposit, f_f, and flux perturbation in the neutron beam, f_s, must be applied to obtain absolute fission rates.

The fission fragments absorption is taken to be $f_f=1-t/2R$. The average fission fragments range in U3O8, R=7.1mg/cm^2, was calculated using equation(1), where ranges in U and O, R_U and R_O were taken from Littmark and Ziegler[4]. For Th there

is not fission fragments range information. So the fission fragments range in ThO_2, R=7.5mg/cm^2, was obtained from the U_3O_8 range using the Bragg-Kleeman[9] rule. We assume 20% error in the ranges, this means <0.3% error in f_f for a 200μg/cm^2 thick foil.

The second correction takes into account the small perturbation produced by the chamber deposit and the SSNTD stack absorption and scattering in the neutron beam (~0.3% in most cases).

Experiments in the Radial Neutron Beam

Experiments were made in the radial neutron beam of the Argentine MTR Research Reactor, RA3. Description of the beam has been made in a previous publication[10].

The miniature fission chamber and/or SSNTD stack were mounted on a dispositive outside the neutrography room and behind an auxiliary shielding. A carriage moves the dispositive inside the room to the center of the neutron beam (fig.2).

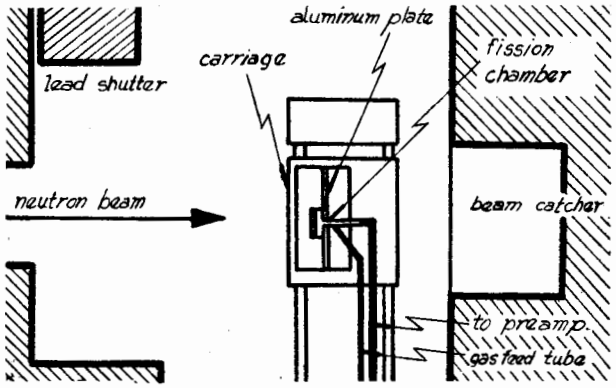

Fig.2 Irradiation Facility

Two types of experiments were made: CTR and CAM. In the first experiments (CTR1, CTR3, CTR4) simultaneous neutron irradiation of SSNTD and fission chamber were made. In CTR1 and CTR3 arrangement, a pair of natural uranium deposits, one in the miniature fission chamber and another in the SSNTD stack, were iradiated during 40 min. In fig. 3 the arrangement for experiment CTR4 is shown. Thorium deposits are placed in the fission chamber (TH13) and in the SSNTD stack (TH12), back to back, with a uranium depleted deposit (UDE1) and irradiated during 6 hours.

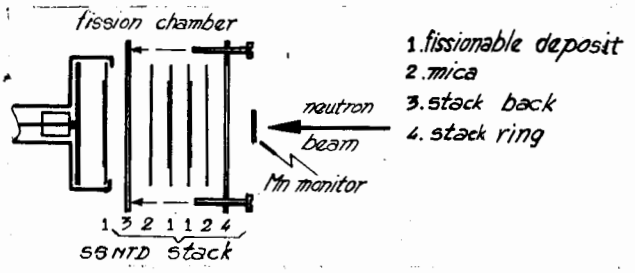

Fig.3 CTR4 Irradiation Arrangement

In CTR experiments chamber fission rates were registered and SSNTD detectors tracks were counted after etching as already described. In Table 3 are the SSNTD optical efficience results obtained from CTR experiments.

In CAM experiments the deposits were irradiated and measured by means of the fission chamber.

To intercalibrate different experiments of CTR and CAM series Mn foils were used as monitor. The neutron capture induced Mn56 activity was measured in a 4πβ proportional gas flow counter. The 4πβ counting was corrected for β absorption in the foil (s_β=0.73)[11], extrapolated to irradiation end and reduced to saturation activity.

From the measured normalized fision rates per U μg, the ratio of depleted to natural uranium, UDEP/UNAT was obtained:

Table 4. Experimental Depleted to Natural Uranium Fission Rate Ratio.

EXP	Foil	f_f	f_s	fis/(s•μg)** x100	UDEP/UNAT
CAM2	UN0	.967	.999	8.049±0.5%	
CAM2	UDE1	.984	.997	1.578±1.0%*	(0.196)*
CAM3	UN0	.967	.999	8.408±0.4%	
CAM3	UDE1	.984	.997	1.564±1.0%	0.186±1.4%
CTR3	UN2	.981	.989	8.034±1.2%	
CTR3	UDE1	.984	.997	1.552±2.4%	0.193±2.7%

**normalized to Mn monitor (A_∞ = 1 d/(s•μg))
*not included in final results because UDE1 monitor has not been directly measured.

The fission rate ratio of thorium to depleted uranium as obtained from fission chamber measurements (F.Ch) and SSNTD counting are in Table 5.

Table 5. Experimental Thorium to Depleted Uranium Fission Rate Ratio.

EXP	Foil	f_f	f_s	fis/(s•μg)* x100	TH/UDEP
CAM3 F.Ch	UDE1	.984	.997	1.564±1.0%	
CTR3 F.Ch	UDE1	.984	.997	1.552±2.4%	0.2675
CAM4 F.Ch	TH14	.990	.999	.4166±2.1%	±1.7%
CTR4 F.Ch	TH13	.988	.993	.4169±1.7%	
CTR4 SSNTD	TH12	.987	.996	.4112±1.6%	0.2667
CTR4 SSNTD	UDE1	.984	.999	1.542±0.9%	±2.2%

*normalized to Mn monitor (A_∞ = 1 d/(s•μg))

An additional measurement of $In115(n,n')In115^m$ reaction relative to a Mn monitor was made on a mock-up of the fission chamber used for CAM and CTR experiments. The 336keV and 847keV photopeaks of $In115^m$ (4.486h) and Mn56 (2.5785h) were measured in a efficiency, E_g, calibrated Ge(Li) gamma spectrometer (Table 6).

Table 6. Experimental $In115^m$/Mn56 Ratio

EXP	Foil	fg*	E_g	d/(s•μg)	In/Mn
FAS1	In	0.975	.017	0.0211±6%	0.0202±6%
FAS1	Mn	1.000	.0068	1.0430±2%	
FAS2	In	0.975	.017	0.0201±6%	0.0199±6%
FAS2	Mn	1.000	.0068	1.0100±2%	

*gamma selfabsorption in the foil

Errors quoted in these tables are obtained by cuadrature sum of independent errors components:
1) Statistics of α and fission products counting

(1σ) and systematic errors in α and fission fragments absorption corrections. 2) Statistics of gamma counting uncertainty (1σ) and gamma selfabsorption for thorium mass assay. 3) Statistics gamma counting errors (1σ) and gamma efficiency calibration errors for $In115^{m}/Mn56$ ratio.

Formalism for Data Reduction

The neutron beam is assumed to have three main components: a maxwellian thermal region, a 1/E epithermal region and a fast region with a neutron fission U235 shape (200keV<E<14MeV).

For the thermal and epithermal region Westcott formalism was applied[12]. Activation and fission rates can be expressed as $\mu_2 \cdot \phi_W$, where ϕ_W is the Westcott flux and $\mu_2 = N\hat{\sigma}$, N is the number of atoms per μg of the measured isotope and $\hat{\sigma}$ is the effective neutron cross section:

$$\hat{\sigma} = g_T \, \sigma_0 \, G_{th} \, F_{res}$$

$$F_{res} = 1 + r\sqrt{T/T_0} \cdot S_0 G_R / g_T G_{th}$$

σ_0 is the thermal cross section (E_0=0.025eV), g_T is the non-1/v factor and F_{res} takes into account the resonance activation as defined by Westcott[12].

The fast fission rate and the inelastic scattering in In115 is $\mu_1 \cdot \phi_f$, where ϕ_f is the fast flux and $\mu_1 = N\tilde{\sigma}$, $\tilde{\sigma}$ is the neutron average cross section in U235 fission spectrum. The fission rates in natural, UNAT, and depleted, UDEP, uranium are:

$$UNAT = \mu_2(nat) \, \phi_W \, (1+\mu_1(nat)/\mu_2(nat) \cdot \phi_f/\phi_W) \quad (2)$$

$$UDEP = \mu_1(dep) \, \phi_f \, (1+\mu_2(dep)/\mu_1(dep) \cdot \phi_W/\phi_f) \quad (3)$$

μ_2 was calculated with g_T=0.9757 and σ_{f0}=582.2b±.2% for U235[6] and N, U235 atoms per U μg in natural (0.72%) or depleted (23ppm) uranium. For μ_1, U238 $\tilde{\sigma}_f$=305mb±3%[13] and U235 $\tilde{\sigma}_f$=1.21b[6], were used.

From the measured ratio UDEP/UNAT=0.190±2% and equations (2) and (3) ϕ_f/ϕ_W=3.11±4% was obtained.

Th232 and In115 μ_1 were calculated with $\tilde{\sigma}_f$=81 mb±7% and $\tilde{\sigma}$=189mb±4%[13].

Mn56 μ_2 was calculated with σ_0=13.41b±.3%[14], G_{th}=1.0 and F_{res}=1.03±1%.

Final Experimental Results and Conclusions

A capability to measure absolute fission rate to a precision level around 2% has been obtained.

The mass assay of thorium and uranium has been done carefully and the details of the technique has been described precisely. The uncertainty in Th232 mass assay is around 1.2%, which is lower than quoted errors in literature[15]. The mass assay technique is a non destructive one and could be applied before and after each fission fragments detection experiment.

Simultaneous irradiation of SSNTD and miniature fission chamber has allowed to obtain both fission ratios and optical efficiency of SSNTD detectors. The mica optical efficiency obtained agrees with previous results[16].

Table 5 shows the experimental ratio of thorium to depleted uranium per μg measured by fission chamber and SSNTD. The agreement is better than quoted errors, indirectly showing the quality of thorium and uranium mass assay.

From results of Table 5 and 6 the experimental ratios of $\tilde{\sigma}_f$(Th232) to $\tilde{\sigma}_f$(U238) and $\tilde{\sigma}_{n,n'}$(In115) were obtained and shown in Table 7. The agreement with evaluated experimental data by Fabry et al.[13] is excellent.

Table 7. Measured Ratios of Microscopic Integral Cross Sections.

	$\frac{\tilde{\sigma}_f(Th232)}{\tilde{\sigma}_f(U238)}$	$\frac{\tilde{\sigma}_f(Th232)}{\tilde{\sigma}_{n,n'}(In115)}$
present results	0.264±2%	0.419±7%
recommended[13]	0.266±7%	0.428±8%

In addition the experimental ratios of UNAT, UDEP, Th and In to Mn and the calculated values using Fabry et al.[13] experimental evaluated integral cross sections are compared in Table 8. The agreement within quoted errors shows the consistency of results obtained by Mn monitor intercalibration of several irradiation experiments.

Table 8.Experimental and Calculated Ratios

Ratio	Experimental	Calculated
UNAT/Mn	0.0822±3%	0.0846±2%
UDEP/Mn	0.0156±3%	0.0159±2%
Th/Mn	0.00416±2%	0.00428±7%
In/Mn	0.0201±6%	0.0202±6%

REFERENCES

1. W. Parker, H. Bildstein, N. Getoff: Nucl. Instr. Meth. 26, 55(1964)
2. G. Dupetit: "Electroplating of Np237 from Isobutyl Alcohol Solution", Instr. Nucl. Phys. Research., Amsterdam-Holland (1966)
3. G. Dupetit, H. Vicens: private communication (1986)
4. U. Littmark, J.F. Ziegler: "Handbook Range Distribution of Energetics Ions in All Elements", Vol.6, Pergamon Press, New York (1980)
5. J.M. Hutchinson, C.R. Nass, D.H. Walker, W. Mann: Int. J. Appl. Radiat. Isotopes 19, 517(1968)
6. IAEA: "Handbook on Nucl. Act. Cross Sections", Tech. Rep. Series Nº 273, IAEA, Vienna (1987)
7. K.H. Beckurts, K. Wirtz: "Neutron Physics", Springer-Verlag, New York (1964)
8. M.D.B. Ricabarra, D. Waisman: CNEA 453 (1979)
9. R.D. Evans: "The Atomic Nucleus", Mc Graw-Hill Book Co.Inc., New York (1955)
10. M.T. Bang, M.D.B. Ricabarra, G.H. Ricabarra: PEO1-06-99-0801-0005/0 (1981)
11. H.Z. Meister: Naturforsch 13a, 722(1958)
12. C.H. Westcott, W.H. Walker, T.K. Alexander: 2nd Int. Conf. Peaceful Uses Atomic Energy, IAEA, 16, 70(1958)
13. A. Fabry, W.N. McElroy, J.A. Grundl, G.E. Jansen: "Neutron Cross Sections for Reactor Dosimetry", Vol.I(Review Papers), IAEA-280, 233, Vienna(1978)
14. K. Axton: U.K. Nucl. Data Progress Rep., UKNDC (85)P112, 90(1984)
15. J.A. Grundl, D.M. Gilliam: "Fission Cross Sections Measurements in Reactor Physics and Dosimetry Benchmarks", The Cf252 Fission Neutron Spectrum (Proc. IAEA Consultant Meeting Smolenice, Czechoslovakia, 1983)(H.Lemmel, H.Cullen, Ed) INDC(NDS)-146, 241 (1983)
16. J.H. Roberts, F.H. Ruddy, R. Gold: Nucl. Tracks, 8, 365(1984)