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STUDIECENTRUM VOOR KERNENERGIE

CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

# INTERCOMPARISON OF FUNDAMENTAL FISSION RATES AND RATIOS IN THE INTERMEDIATE-ENERGY STANDARD NEUTRON FIELDS AT THE ITN-ΣΣ AND MOL-ΣΣ FACILITIES

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Summary. - Measurements of absolute fission rates and ratios have been performed in the ITN-ΣΣ Intermediate-Energy Standard Neutron field by means of fission chambers previously exposed in the sister facility MOL-ΣΣ. Absolute double gas-flow ionization fission chambers of the NBS type as well as single, sealed chambers developed at SACLAY were used; the latter ones are kept as references for future controls of the stability of central ITN-ΣΣ fission rates with respect to the two permanent and redundant chamber monitors applied for run-to-run flux level normalization.

The fission cross-section ratios measured at ITN-ΣΣ centre are (1.000 : 1.169 ± 0.027 : 0.0566 ± 0.0014 : 0.380 ± 0.011) for $^{235}$U : $^{239}$Pu : $^{238}$U : $^{237}$Np respectively; they agree with similar data obtained at MOL-ΣΣ centre.

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Résumé. - Des mesures absolues de taux de fission et d'indices de spectre ont été faites dans le champ neutronique standard ITN-ΣΣ au moyen de chambres à fission préalablement exposées dans la facilité jumelle MOL-ΣΣ.

Des chambres à fission doubles, à courant gazeux, du type développé au NBS, ainsi que des chambres scellées provenant de Saclay, ont été utilisées; ces dernières sont conservées comme références pour tout contrôle futur de la stabilité des taux de fission centraux dans ITN-ΣΣ par rapport aux deux chambres monitrices permanentes appliquées pour la normalisation précise du niveau de flux d'une irradiation à l'autre.

Les rapports des sections efficaces moyennes de fission au centre de l'assemblage ITN-ΣΣ sont respectivement (1.000 : 1.169 ± 0.027 : 0.0566 ± 0.0014 : 0.380 ± 0.011) pour $^{235}$U : $^{239}$U : $^{238}$U : $^{237}$Np. Ces rapports sont en accord avec les données correspondantes relevées au centre de l'assemblage MOL-ΣΣ.

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Samenvatting. - Absolute splijtingssnelheids- en spektrumindexmetingen werden in het ITN-ΣΣ standaard neutronenveld verricht met splijtingskamers die voorheen in de MOL-ΣΣ zusteropstelling werden bestraald.

Absolute dubbele splijtingskamers met gasstroom van het NBS-type en enkelvoudige verzegelde kamers ontwikkeld te Saclay werden gebruikt; deze laatsten worden bewaard als referentie voor latere kontrole van de stabiliteit van centrale ITN-ΣΣ splijtingssnelheden ten opzichte van twee permanente en meervoudige kamermonitoren aangewend voor de nauwkeurige normalizatie van het fluxniveau van bestraling tot bestraling.

De verhoudingen van de splijtingswerkzame-doorsneden gemeten in het centrum van ITN-ΣΣ bedragen (1.000 : 1.169 ± 0.027 : 0.0566 ± 0.0014 : 0.380 ± 0.011) respektievelijk voor $^{235}$U : $^{239}$Pu : $^{238}$U : $^{237}$Np; zij stemmen overeen met de gelijkaardige gegevens verkregen in het centrum van de MOL-ΣΣ opstelling.

# INTERCOMPARISON OF FUNDAMENTAL FISSION RATES AND RATIOS IN THE INTERMEDIATE-ENERGY STANDARD NEUTRON FIELDS AT THE ITN-ΣΣ AND MOL-ΣΣ FACILITIES

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# 1. INTRODUCTION

Intermediate-energy standard neutron fields [1] provide a unique capability for the development and standardization of experimental techniques and nuclear data needed for fast reactor physics and for fuels and materials dosimetry. Among a variety of applications, the calibration of instruments and activation detectors used for in-pile measurements of fission rates and burn-up is particularly vital. The establishment of high accuracy fission rate scales is furthermore one of the milestones in the utilization of standard neutron fields as benchmarks for cross-section validation; such approach is best examplified by the guidelines proposed at the IAEA panel on nuclear data for reactor neutron dosimetry [2].

Secondary intermediate-energy standard neutron fields of the ΣΣ type [3] are presently in operation at the NISUS facility [4], University of London Reactor Centre, England, at the Mol-ΣΣ facility [3], CEN/SCK, Belgium and at the ITN-ΣΣ facility [5], Bucharest, Rumania. These three facilities are implemented within 50 cm diameter spherical cavities hollowed out from conventional graphite thermal columns; the standard configuration is constituted by a ∿ 5 cm thick natural uranium spherical source, ∿ 25 cm outer diameter, surrounding a concentric natural boron carbide shell at the centre of which the standard neutron field is generated.

This paper documents joint ITN-MOL measurements of fundamental fission rates performed at the centre of the ITN-ΣΣ standard neutron field by means of fission chambers extensively used in the Mol-ΣΣ facility; two of these instruments are NBS-type double absolute fission chambers [6] which have also been previously exposed [7] in the NISUS facility; they are intercalibrated with the NBS (National Bureau of Standards, USA) absolute fission chambers used in the benchmark neutron fields selected for the US Interlaboratory LMFBR Reaction Rate (ILRR) programme [8].

# 2. EXPERIMENTAL DETAILS

## 2.1. Geometry for exposure of absolute fission chambers at the centre of the ITN-ΣΣ facility

The ITN-ΣΣ facility is presently implemented in the horizontal thermal column of the VVRS reactor [5] Institute of Atomic Physics (IFA), Bucharest. This graphite thermal column is cylindrical, with a diameter of 110 cm and a length of 300 cm; it is mounted on an iron table with wheels and can be pulled out in the reactor hall for manipulation of the experiments.

The 50 cm diameter spherical cavity is located at the centre of the thermal column; the upper part of the cavity block is fixed to a curved stainless steel plate which provides the mechanical basis for suspension of the ΣΣ shells and their removal by means of the reactor transfer bridge.

The natural uranium metal source consists of these concentric spherical shells with a total thickness and an outer diameter identical to the Mol-ΣΣ ones, e.g. respectively 5.0 and 24.5 cm (as in the new version of the Mol-ΣΣ facility, a set of additional uranium shells is available so that an array of different configurations may be realized). These uranium shells are provided with two identical access holes; one is drilled through the horizontal intersection plane of the two hemispherical parts and the other one lies on the vertical polar axis; appropriate plugs are available to close the holes or reduce their diameter to 17 mm. The boron carbide shells are the same as in Mol-ΣΣ.

On figure 1 is sketched the configuration used for exposure of the NBS-type absolute fission chambers. The fissionable layers are placed vertically and back-to-back at ITN-ΣΣ centre; the same deposits were irradiated in the horizontal midplane for Mol-ΣΣ experiments [9] [10], and the present geometry was selected, for reasons of convenience only, after complementary runs had verified the expected lack of any angular response sensitivity. The access hole is shielded against reentrant subcadmium neutrons by means of a collimating screen similar to the one applied and extensively assessed in Mol-ΣΣ [9].

## 2.2. Fission chambers : types, operation, performances

Three different types of fission chambers have been used for this work. Fission rate distributions along the vertical and horizontal polar axis have been registered with cylindrical, miniature chambers. The parallel plate absolute ionization fission chambers for central reaction rate measurements are either from the gas flow(x) NBS-type [6] or from the CEA (Saclay) sealed type [11], 20 mm outer diameter. Run-to-run flux level monitoring is achieved by two redundant, CEA-type sealed chambers: one is fixed against the wall of the ITN-ΣΣ graphite cavity and the other one on the axis of the thermal column, at about 60 cm from the cavity centre in direction of the reactor core.

The electronics attached to each chamber involve a low noise preamplifier and amplifier, three discriminator levels and three scalers. A multichannel analyser is furthermore used to record the fission fragment pulse-height distribution for each chamber as well as to properly set the three discriminators; the settings were done according to the procedure described by GRUNDL et

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(x) Flow of P-10 gas (90 % argon - 10 % methane) al. [6]; a lower bias is put at $V_L = 0.36\ V_{PEAK}$, an upper bias at $V_U=0.54\ V_{PEAK}$ and a gain control bias at $V_{GC}=1.4\ V_{PEAK}$, where $V_{PEAK}$ is the position in volts of the maximum in the fission fragment pulse-height distribution; the corresponding count rates are labelled $S_L$, $S_U$ and $S_{GC}$ respectively. The NBS-type fission chambers were operated by means of an electronic chain transported from Mol to Bucharest; in one irradiation, this electronics was replaced by ITN modules and, although this change resulted in some resolution loss, no fission rate difference could be observed, as expected (a similar consistency check was done during NBS-type fission chamber measurements at the NISUS facility [7]). All fission chambers used for absolute central fission rate measurements had been previously irradiated at the centre of the Mol-ΣΣ facility; the details of these runs are documented in [10] for NBS-type fission chambers and in [12] for CEA-type chambers. The performance of all chambers in the present experiment was generally as excellent as observed in Mol-ΣΣ; an exception occurred however in the first exposures of the NBS chambers: the number of small pulses due to gamma ray pile-up in the chamber manifold was such that the performance check ratio $S_L/S_U$ for natural uranium departed from its nominal value by an amount too important to afford an accurate correction; this effect was eliminated by improved electrical insulation in the remaining runs.

### 2.3. Derivation of unperturbed absolute fission rates from chamber pulse rates and appraisal of uncertainties

In Table I are gathered the main data relevant to the thin fissionable deposits used for absolute fission rate measurements at the centre of the ITN-ΣΣ standard neutron field. The masses of the NBS deposits have been determined at NBS by alpha and fission counting comparisons to the reference deposits the mass assay of which is documented in [6], Table II. The masses of the CEA (Saclay) deposits have been derived from calibrations in the Mol-ΣΣ standard neutron field and are tied directly to the results [9] of interlaboratory fission chamber comparisons involving NBS, GfK (Kernforschungszentrum Karlsrühe), RCN (Reactor Centrum Nederland, Petten) and Mol.

A flat extrapolation to zero of the fission pulse height distribution is assumed valid [6] for all chambers and fissionable deposits, e.g. the actually measured distribution is replaced by a horizontal line drawn from the minimum in the valley between the fission fragment peak and the alpha-electron-electronic noise. Under such interpretation, the extrapolation to zero (ETZ) correction may alternatively be defined from the lower ($V_L$) or the upper ($V_U$) discriminator level; the second definition is used in this work, in contrast to [6]. It must thus be understood that all ETZ values quoted in Table I contain a sizeable fraction of valid fission pulses, e.g. the pulses registered between $V_L$ and $V_U$: this is the reason why, for a given deposit thickness, the correction is much higher than in [6] and its fractional uncertainty much lower; the corresponding absolute fission rate and its uncertainty are not affected by the choice of definition. In terms of the linear dependence of the ETZ on fissionable deposit thickness ([6] Fig. 4), it is noted that the actual correction for the CEA chambers as derived from the $S_L/S_U$ ratios is about twice as expected; this is the case also for the experiences performed with these chambers [12] in three other neutron fields at Mol. This effect is believed to be caused by imperfect collection of the ionization in these chambers. No additional uncertainty is associated with such deviation because it was the same for the mass assay calibration runs by fission fragment counting [12].

The fission fragments undetected as a result of absorption in the deposits themselves are accounted for, as in [6], by applying a correction of 0.69 % per 100 μg/cm$^2$ for $UO_2$ and $NpO_2$ and 0.66 % per 100 μg/cm$^2$ for $PuO_2$, with an estimated uncertainty equal to 25 % of the correction (but never lower than 0.35 %). This absorption correction ranges here from 0.4 % up to 3.7 %.

Dead-time losses in the present measurements are small. The count rates range from ∿ 65 cps up to ∿ 350 cps for the absolute chambers at a nominal reactor power of 300 kW, while the count rate of both monitors is ∿ 1000 cps. With MOL electronics, the dead-time is 4.4 μs for the $V_L$ bias and 4.0 μs for the $V_U$ bias. It is assumed that dead-time is proportional to the pulse width at half maximum in the main amplifier so that the Mol values are multiplied by 2 or 4 for the ITN electronics, depending on the selection of time constants. A complementary run at a reactor power of 50 kW has been done and the results agree well with the 300 kW irradiations.

The corrections for fission of impurity isotopes in the deposits are generally small and are the same as in Mol-ΣΣ. The same statement applies to the remaining corrections related to the instrumental perturbation of the neutron field : access hole correction (0.995 ± 0.005 for $^{235}U$ and $^{239}Pu$) and correction for neutron scattering and absorption in the chamber structure, especially the backing of the fissionable deposit (1.006 ± 0.003 or 1.000 ± 0.001, depending on whether the principal isotope is fertile or fissile).

The statistical uncertainties have always been kept below ± 0.2 % by running the reactor long enough. The run-to-run flux level normalization factors are deemed accurate to ± 0.5 % or better on the basis of external error appraisal and reproducibility arguments; this applies to the period of the present experiments, but has not been established yet on a sustained, long-term scale.

Uncertainties due to chamber positioning in ITN-ΣΣ are at present slightly higher than in Mol-ΣΣ because the manipulation of the assembly is somewhat more complex; in the worst case, e.g. threshold fission foils, such uncertainties however do not exceed ± 0.5 %.

## 3. RESULTS

All the results of the present work are contained in Tables II and III and in Fig. 2.

Owing to the two types of access hole available in the ITN-ΣΣ facility, fission rate traverses could be performed both in the horizontal plane along the thermal column axis and on the vertical polar axis, through the assembly centre. Note that each experimental point on Fig. 2 involves an independent irradiation, as the reactor must be shut down and the thermal column extracted before any modification of positioning can be achieved; the quality of these data thus provides further support to the assessment of a run-to-run flux level normalization accuracy better than ± 0.5 %. Fig. 2 furthermore calls for the following comments.

- The fission rate gradients along the horizontal axis are identical to the ones observed in Mol-ΣΣ [3]; they are related to the overall thermal flux gradient within the thermal column : less fissions are generated in the part of the source looking outwards (e.g. in direction of the access hole); the impact of this source non-uniformity on central $^{235}$U and $^{239}$Pu fission rate traverses is partly smeared out by multiple cavity crossings at intermediate neutron energies.
- The fission rate gradients along the vertical polar axis are caused by a decrease of the source strength at its bottom(*) (opposite to the access hole): thermal neutron absorption by the thick aluminium support of the uranium sphere (Fig. 1) is the reason for this effect; in future, this support plate will be replaced by an annulus as in Mol-ΣΣ and the vertical gradient at centre will consequently be flattened to negligible proportions.
- The neutronic centre of ITN-ΣΣ might be slightly displaced downwards of its geometrical centre due to support absorption; the displacement, if any, is difficult to quantify because of possible systematic positioning uncertainties of the order of ± 2 - 3 mm.

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(*) Thermal neutron flux maps in the empty cavity are symmetric about the centre in the vertical plane

- The perturbation of the fission rates for $^{235}$U and $^{239}$Pu due to subcadmium neutrons entering the central zone through the shielded access hole is undetectable, except eventually close to this hole in the case of $^{239}$Pu ($\sim$ 0.3 eV resonance [7]); this agrees with previous extensive investigations in Mol-ΣΣ [9].

The absolute fission rates per nucleus observed at ITN-ΣΣ centre and gathered in Table II generally result from a few independent irradiations; excellent agreement was always found between the different runs, within assigned reproducibility uncertainties. For $^{235}$U and $^{239}$Pu, the two different types of instruments used agree with each other.

It is interesting to compare absolute fission rate scales in Mol-ΣΣ and ITN-ΣΣ at a same power of the driving reactor, e.g. BR1 and VVRS respectively. For both piles, the maximum reactor power is $\sim$ 3 MW.
The fission rates per nucleus of $^{235}$U at 3 MW are as follows :

Mol-ΣΣ : $2.8 \times 10^{-15}$
ITN-ΣΣ : $6.2 \times 10^{-15}$ fission/nucleus.s

The factor two difference in flux level between the two facilities is mostly due to the shortest distance from the reactor core for ITN-ΣΣ; increased core neutron leakage is the penalty : the cadmium ratio for gold at infinite dilution exceeds $10^4$ in the Mol cavity when empty, and is of the order of 200 in the ITN cavity. Although the influence of core leakage is negligible for fission rates, this might not remain true for high-energy threshold reactions and possible background corrections should be investigated. It is worth mentioning here that the maximum power of the VVRS reactor is being increased to 15 MW and an appropriate manipulation cell for the ITN-ΣΣ facility is under completion; these facts render the ITN standard particularly useful for all applications requiring comparatively high neutron fluences.

The data in Table III establish the effective neutronic identity of the Mol-ΣΣ and ITN-ΣΣ standard neutron fields.

## 4. CONCLUSIONS

In terms of the well established need for international standardization of fundamental reaction rate measurements, the present work is the first practical step taken to link the Rumanian fast reactor physics and neutron dosimetry programme with similar programmes in Western Europe and the U.S.. More specifically with regard to the applications of ΣΣ-type neutron fields, such link enhances the ability to share and co-ordinate current and future efforts by providing :

- adequate demonstration of the effective identity of the standard as realized at the different sites;
- an accurate, absolute fission rate scale consistent for the facilities Mol-ΣΣ, ITN-ΣΣ, NISUS and other relevant benchmarks; such consistency has been achieved by systematic application of a same transfer instrument, e.g. NBS-type fission chambers.

These achievements are particularly useful from the standpoint of the current standardization of reactor neutron dosimetry because the ITN-ΣΣ standard neutron field can be operated at fluences about an order of magnitude higher than its sister facilities MOL-ΣΣ and NISUS.

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## TABLE I

Fissionable deposits exposed at the centre of
the ITN-ΣΣ standard neutron field

| Deposit identification and principal isotope | | Mass of principal isotope ($\mu g/cm^2$) | Extrapolation-to-zero (ETZ)(f) | |
|---|---|---|---|---|
| | | | nominal (g) | actual(h) |
| NBS Deposits(a) | | | | |
| 28N-5-2 | $^{238}$U(natural) | 541.5 (± 1.4%)(d) | 1.040 ± 0.01 | (1.064)(i) |
| 28HD-5-1 | $^{238}$U(99.938%) | 514 (± 2.2%)(d) | 1.037 ± 0.01 | 1.043 ± 0.013 |
| 25S-2-3 | $^{235}$U(99.75%) | 175 (± 1.4%)(d) | 1.015 ± 0.005 | 1.016 ± 0.005 |
| 49I-1-1 | $^{239}$Pu(99.11%) | 83.0 (± 1.4%)(d) | 1.006 ± 0.004 | 1.009 ± 0.004 |
| 37-5-1 | $^{237}$Np(99.3%) | 497 (± 1.8%)(d) | 1.035 ± 0.01 | 1.036 ± 0.01 |
| CEA (Saclay) deposits(b) | | | | |
| 23(c) | $^{235}$U(99.89%) | 50.4 (± 1.6%)(e) | 1.005 ± 0.004 | 1.014 ± 0.005 |
| 24(c) | $^{239}$Pu(99.88%) | 53.1 (± 1.8%)(e) | 1.005 ± 0.004 | 1.014 ± 0.005 |

(a) 12.7 mm diameter fissionable oxides vacuum evaporated on platinum backing 19 mm diameter 0.13 mm thick.

(b) 15 mm diameter fissionable oxides vacuum evaporated on Zircaloy backings 20 mm diameter 0.1 mm thick.

(c) Refers to the fission chamber (sealed) rather than to the deposit itself.

(d) Based on the latest comparisons with the NBS reference deposits [6].

(e) Based on calibrations in MOL-ΣΣ [12].

(f) Correction for fragments below the upper discriminator level $V_U$; about twice the correction for fragments below the alpha cut-off (see text).

(g) Derived from ETZ values given in Fig. 4 of [6] after accounting for difference in definition compared to this paper.

(h) Derived from observed $S_L/S_U$ ratios according to recommendations of [6].

(i) Affected by gamma ray pile-up (see text).

## TABLE II

Absolute fission rates relative to principal monitor(a) for ITN-ΣΣ

| Isotope | Fissionable deposit(b) | Fission rate per monitor count, x $10^{19}$ (fission/s. nuclide . cps) |
|---|---|---|
| $^{235}U$ | NBS 25S-2-3 | 6.542 (± 1.7 %) |
| | CEA 23 | 6.604 (± 1.8 %) |
| $^{239}Pu$ | NBS 49I-1-1 | 7.678 (± 1.7 %) |
| | CEA 24 | 7.674 (± 2.0 %) |
| $^{238}U$ | NBS 28HD-5-1 | 0.3715 (± 2.5 %) |
| $^{237}Np$ | NBS 37-5-1 | 2.479 (± 2.7 %) |

(a) Fission chamber fixed against cavity wall. Count rate at 300 kW : ∿ 950 cps.

(b) No data given for foil 28N-5-2 because of gamma-ray pile-up effects.

## TABLE III

Fission cross-section ratios for ΣΣ

| Ratio | MOL-ΣΣ | | ITN-ΣΣ |
|---|---|---|---|
| | Interlaboratory [9] | NBS chamber [10] | this work |
| $\frac{\bar{\sigma}_f(^{238}U)}{\bar{\sigma}_f(^{235}U)}$ | 0.0561 (± 1.5 %) | 0.0564 (± 2.5 %) | 0.0566 (± 2.5 %) |
| $\frac{\bar{\sigma}_f(^{239}Pu)}{\bar{\sigma}_f(^{235}U)}$ | 1.167 (± 2.0 %) | 1.173 (± 2.1 %) | 1.169 (± 2.3 %) |
| $\frac{\bar{\sigma}_f(^{237}Np)}{\bar{\sigma}_f(^{235}U)}$ | 0.388 (± 2.5 %) | 0.381 (± 2.9 %) | 0.380 (± 3.0 %) |

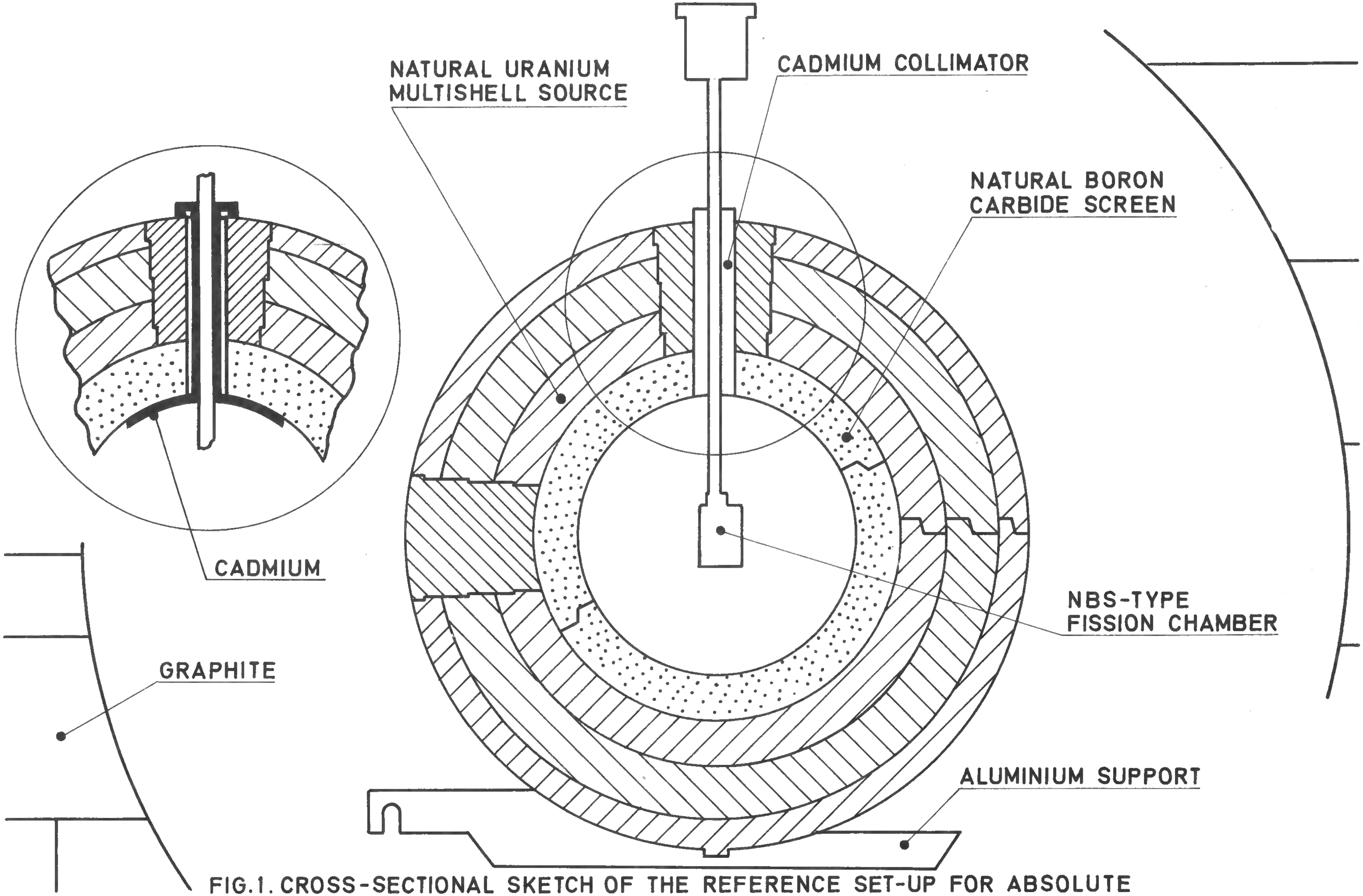

FIG.1. CROSS-SECTIONAL SKETCH OF THE REFERENCE SET-UP FOR ABSOLUTE FISSION CHAMBER FISSION RATE MEASUREMENTS IN ITN-ΣΣ

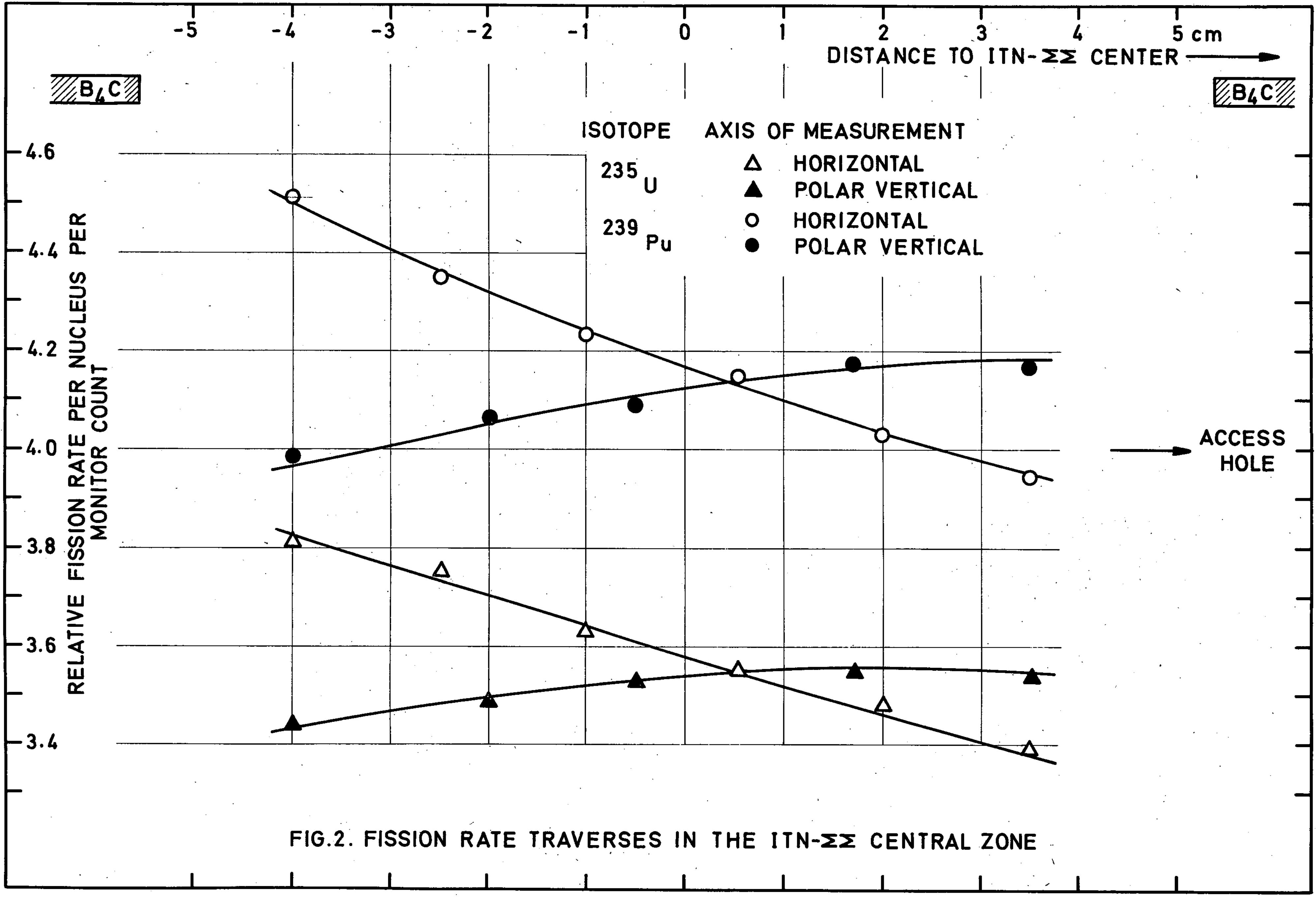

FIG.2. FISSION RATE TRAVERSES IN THE ITN-ΣΣ CENTRAL ZONE