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MEASUREMENTS OF INTEGRAL CROSS SECTION RATIOS IN TWO DOSIMETRY BENCHMARK NEUTRON FIELDS

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ABSTRACT

In the frame of a current interlaboratory effort devoted to the standardization of fuels and materials neutron dosimetry, the $^{103}Rh(n,n')^{103m}Rh$ and $^{58}Ni(n,p)^{58}Co$ integral cross sections have been accurately measured relatively to the $^{115}In(n,n')^{115m}In$ cross section in the ^{235}U thermal fission neutron spectrum and in the MOL-$\sum\sum$ Intermediate-Energy Standard Neutron field. In this last neutron field, the data are related also to the $^{235}U(n,f)$ cross section.

The measurements are extensively documented and the results briefly compared to literature. Most noticeably, decisive support is provided for the selection of a specific $^{103}Rh(n,n')^{103m}Rh$ differential-energy cross section among the existing, conflicting data.

1. INTRODUCTION

The growing interest in realistic interlaboratory exchanges of fuels and materials irradiation effects data that can be properly correlated and applied necessitates a fast improvement and standardization [1] of the metrology capabilities for neutron spectral flux and fluence characterization and for radiation damage dose specification.
The development of recommendations [2] for the damage functions of materials and for the definition of displacement or energy deposition dose rates is an important milestone in such standardization effort.
Appropriate emphasis also has to be and is increasingly placed onto the establishment of consistent and reliable dosimetry cross section files [3][4] for activation foils used to monitor and/or calibrate material testing experiments.
A basic requirement for these nuclear data files is their ability to reproduce integral reaction rates observed in neutron flux spectra representative of the environments of practical engineering interest. Standard neutron fields are particularly ideal benchmarks [3] for the integral consistency testing of the dosimetry reactions featuring the best known differential-energy cross sections.
For the other, numerous reactions, integral measurements in benchmark and standard neutron fields provide a promising and economical approach [1][3] to make suitable selections or adjustements [5] in the usually scarce and/or discrepant differential nuclear data [6][7].

This joint work has been performed within such frame and was undertaken as the result of

- the mutual interest of the IAEA and MOL authorities into the standardization of reactor neutron dosimetry
- the availability at MOL of two of the few relevant European dosimetry benchmark neutron fields
- the existence at the SEIBERSDORF laboratory, IAEA, of an established, high level expertise for accurate, absolute activity measurements; direct interlaboratory comparison with the techniques applied at MOL was thus useful, especially more as these MOL techniques are currently intercompared with the ones developed in a number of European and U.S. laboratories involved

in neutron dosimetry; on another hand, the IAEA expertise is an unique and unchallenged one in the case of the important dosimetry reaction $^{103}Rh(n,n')^{103m}Rh$ [8].

The $^{103}Rh(n,n')^{103m}Rh$ and $^{58}Ni(n,p)^{58}Co$ integral cross sections have been accurately determined relatively to the $^{115}In(n,n')^{115m}In$ cross section in the two standard neutron fields available : the thermal neutron-induced fission neutron spectrum of uranium-235 and the MOL-ΣΣ Intermediate-Energy Secondary Standard Neutron Field [9], which simulates the core neutron spectrum of a fast breeder reactor. The indium cross section is subject to separate, high accuracy measurements relatively to the $^{235}U(n,f)$ cross section in these two neutron fields, as well as to an absolute measurement in the fission spectrum.

2. THE STANDARD NEUTRON FIELDS

2.1. General implementation and brief physical description of the experimental configurations

The BR1, graphite moderated, natural uranium reactor [10] at MOL is equipped with two large, high quality graphite thermal columns. A spherical cavity has been hollowed out in each of these thermal columns for the purposes of the project "Standard Neutron Spectra" [11][12]. The cavity of the horizontal thermal column has a diameter of 50 cm and the cavity of the vertical column has a diameter of 1 meter. Among a number of applications, these two cavities are used for the production of thermal neutron-induced fission neutron spectra; in the 50 cm cavity is generated the intermediate-energy secondary standard neutron field ΣΣ, extensively described elsewhere [9].

Briefly, ΣΣ is a 5 cm thick, 24.5 cm outer diameter natural uranium spherical shell placed at center of the 50 cm diameter cavity; the low energy tail of the neutron spectrum is properly shaped by a 1.5 cm thick vibrocompacted boron carbide shell with a 11 cm diameter central exposure zone.

A number of source-detector configurations have been developed at MOL for integral measurements in fission neutron spectra [13][14][15][16]. The present work involves the application of two slightly different assemblies,

patterned according to the design principles of the NBS Cavity Fission Spectrum Neutron Source [17]; this design was selected for two major reasons :

- the optimisation realized in terms of the neutron field perturbation by neutron scattering and absorption in the assembly structural components and the availability at NBS of a code [18] to compute this perturbation.
- the interest to achieve some degree of standardization in the production of cavity fission neutron spectra.

Fig. 1 displays one of the source-detector assemblies. Two bare discs of 93% enriched uranium, diam. 19 x 0.1 mm, are coaxially separated by 8.36 mm and locked into contact of a 1 mm thick cadmium box containing at its center a stack of activation foils, here diam. 10 x 0.1 mm for rhodium and nickel, diam. 10 x 0.127 mm for indium. The cadmium thickness was optimized through a new series of $^{197}Au(n,\gamma)^{198}Au$ and $^{115}In(n,\gamma)^{116m}In$ fission spectrum average cross section measurements.
Cobalt foils are used to monitor the incident thermal neutron flux. The second assembly is similar, except that the fission source discs are now separated from the illustrated cadmium box by thin aluminium spacer rings, providing a source separation distance of 20.3 mm.
The two assemblies are fixed at their bottom to a thin stainless steel tube which is inserted vertically in the BR1 diam. 1 meter spherical cavity ; the mounting is such that the axis of the assemblies lie 10 cm above and 10 cm below the cavity center respectively, with the fission disc planes parallel to the vertical axis of the thermal column; these planes are kept more or less in the same angular orientation with respect to a reference direction (the main axis of the BR1 horizontal cylinder core loading) so as to maintain reasonable source strength invariance with respect to the angular dependence of the thermal neutron flux in the cavity. With such mounting, the neutron emission rates of the two fission sources in one assembly are closely identical and the fast neutron background in each assembly due to in-vacuum streaming from the other one is smaller than 1%;

this background is accurately derived from source strength determinations and simple geometrical considerations. Of course, the two assemblies need not to be exposed simultaneously, but they can be for the expensive high power runs, at almost no cost in accuracy. Total background responses can furthermore be determined reliably by removing the fission discs from one of the assemblies, as a result of

- the spatial and angular uniformity of cavity wall return neutron flux spectra for radii of the order of a fifth of the cavity radius (reaction rate backgrounds due to this component are negligible for threshold reactions) [19]
- the relatively weak spatial variation of scalar neutron and gamma ray fluxes at cavity center in absence of fission sources.

Advantage is taken also of these properties for fission spectrum average capture and fission cross section measurements [14][16].

The assembly illustrated on fig. 1, but without fission discs, was also used for the exposure of the activation foils at $\Sigma\Sigma$ center.

2.2. Spectral characterization and status of absolute integral cross section scales

The $\Sigma\Sigma$ neutron spectrum has been characterized by joint GfK (Karlsruhe), RCN (Petten) and CEN-SCK (Mol) spectrometry measurements [20] over the energy range 5 keV to 5.5 MeV with an estimated uncertainty of $\pm$ 5% between 20 keV and 1.5 MeV; above 1.5 MeV, the reliability of the measurements mostly depends on the accuracy of the $^{6}Li(n,\alpha)$ and $^{3}He(n,p)$ cross sections, which is at present not better than $\pm$ 10-15%; the reasonably well consistent ^{6}Li and ^{3}He spectrometry data for $\Sigma\Sigma$ are however supported further by new, preliminary proton recoil experiments extending up to 4 MeV [21]. One-dimensional discrete-ordinates transport theory computations based on ENDF/B-III [22] do not depart from the measurements by more than $\pm$ 10% and are used to extrapolate them outside of their range of coverage.

A number of integral reaction rate measurements have been [9] and are currently performed at the center of the ΣΣ facility on an interlaboratory basis.

The uranium-235 absolute fission rate at ΣΣ center is being defined jointly by four laboratories (GfK, RCN, NBS and MOL) with an accuracy goal of ± 1-2%, with respect to a redundant and precise monitoring scheme involving $^{197}Au(n,\gamma)^{198}Au$ and $^{115}In(n,n')^{115m}In$ activation foils and an adjacent fission chamber monitor. Present accuracy for this scale is ± 3.5% or better. All monitoring data are converted into best $^{115}In(n,n')^{115m}In$ central reaction rates for each irradiation in ΣΣ and these reaction rates are in turn linked to the available thermal neutron flux in the empty 50 cm cavity.

This monitoring procedure has been used also in the present work and the observed $^{115}In(n,n')^{115m}In$ reaction rates thus accurately associated to uranium-235 fission rates; the presently recommanded value for this reaction rate ratio is equal to

$$^{235}U(n,f)/^{115}In(n,n')^{115m}In = 27.1 \pm 0.9$$

and the ΣΣ average uranium-235 fission cross section is taken equal to

$$\bar{\sigma}_f(^{235}U,\ \Sigma\Sigma) = (1589 \pm 50)\ mb$$

which leads to a ΣΣ average $^{115}In(n,n')^{115m}In$ cross section of

$$\bar{\sigma}_{n,n'}(^{115}In,\ \Sigma\Sigma) = (58.6 \pm 3.0)\ mb \qquad (1)$$

The $^{115}In(n,n')^{115m}In$ reaction has been selected also as a transfer monitor to relate the measured average fission spectrum cross sections to $^{235}U(n,f)$ as well as to an absolute scale based on source strength determinations and straightforward geometry factors. Conversely to the case of ΣΣ experiments, such relationships have not yet been established to the required accuracy and are still considered preliminary; they will not be used at this stage, but it may tentatively be stated here that initial confirmation has been obtained

regarding the fact that the evaluated integral data in reference [23] are not overestimated, as recently suggested [24]. It is consequently assumed here that the $^{115}In(n,n')^{115m}In$ fission spectrum average cross section is equal to

$$\bar{\sigma}_{n,n'}(^{115}In, \chi_{25}) = (188 \pm 4) \text{ mb} \qquad (2)$$

Relations (1) and (2) may be considered as the presently recommended basis to express as absolute spectrum average cross sections the integral reaction rate ratios measured in this work.

2.3. Spectral uniformity and integrity of the neutron fields

In the conditions of these experiments, corrections for neutron flux gradients, background and neutron scattering in the detector assemblies do not exceed 0.5% in $\Sigma\Sigma$ irradiations.

Table I gathers the corrections and associated uncertainties in fission spectrum experiments for the two adopted source separation distances.
The greatest corrections are related to photoactivation of rhodium and indium. They could be determined from background measurements as described in section 2.1., after substracting the computed cavity wall return [19] and source streaming components; they agree reasonably well with empirical predictions [25]. Reactor neutron backgrounds due to leakage from BR1 core are entirely negligible.
The greatest uncertainties originate in intrinsic perturbations due to neutron scattering and absorption in the source-detector assemblies. Such perturbations have been computed at NBS [18], in the approximation of a few energy groups and single scattering, for similar arrangments of sources and activation foils. No correction has been applied yet to the data of this work, due to the present lack of a computation for the exact, actual assemblies, but upper systematic error bounds have been assigned on basis of the mentioned calculations.

Axial flux gradients are weak over the spatial extent of the centered stacks of foils exposed in these measurements. They are essentially geometrical ones and are the steepest for the closiest source separation distance of 8.36 mm. Gradient measurements for this case are compared to calculations on fig. 2. Any irradiated rhodium or nickel foil was always surrounded by indium monitors so that gradient interpolations of the order of 0.1% could be done very accurately.

3. DETERMINATION OF ABSOLUTE ACTIVITES

3.1. Approach

All foils irradiated for these experiments have been measured at MOL by means of NaI(Tl) gamma-ray and X-ray spectrometers in a "principal" counting geometry. The X-ray spectrometer and its electronics were transported from the SEIBERSDORF laboratory and operated by an IAEA staff member.
The isotopic purity of the gamma ray spectra was checked with a high resolution Ge(Li) spectrometer.
The absolute activity scales for each reaction are however not based only on the calibration data for the principal counting geometry, but are composite ones encompassing independent calibrations of other arrangments in which a few foils were also measured.
For two of the investigated reactions, these scales could be defined as interlaboratory averages through transfer of reference samples between IAEA and MOL : cobalt-58 sources for the $^{58}Ni(n,p)^{58}Co$ reaction and chromium-51 sources for the $^{115}In(n,n')^{115m}In$ reaction, as the photopeak efficiencies of NaI(Tl) for ^{115m}In and ^{51}Cr are very close to each other. Similarly, ^{103}Pd sources may be used for interlaboratory transfer and/or comparison of calibration data for ^{103m}Rh [26]; such sources allowed a relevant control of the performances at MOL of the IAEA X-ray spectrometer.
All decay scheme data accepted for the evaluation of these measurements are gathered in table II.

3.2. Reaction $^{103}Rh(n,n')^{103m}Rh$

The X-ray spectrometer for rhodium counting consists of a NaI(Tl) crystal 2 mm thick and 38 mm diameter with a front window in beryllium, 0.125 mm thick; it is shielded by 10 cm of lead for background reduction. The pulse-height spectrum is displayed on a multichannel analyzer.

The efficiency of this spectrometer for 20 keV K-X rays was determined at a few sample-detector distances by means of calibrated sources of ^{103m}Rh electro-deposited on vyns films. The disintegration rate of these sources was defined accurately by the $4\pi e^- - X$ coincidence technique; a detailed description of the preparation and calibration of these sources is given elsewhere [8][27]. Through such approach, the derived absolute $^{103}Rh(n,n')^{103m}Rh$ reaction rates do not depend on conversion coefficients and fluorescence yields. Proper correction must be applied for X-ray selfabsorption within the diameter 10 mm, 0.1 mm thick activation foils; such corrections have been experimentally established as a function of the foil weight.

3.3. Reactions $^{115}In(n,n')^{115m}In$ and $^{58}Ni(n,p)^{58}Co$

The principal counting geometry involves pressing the activated foils flatly at contact and on top of the aluminium package of a NaI(Tl) 3" X 3" gamma ray spectrometer; the foils, centered coaxially to the sensitive volume, lie with their closiest face at a separation distance of 0.55 cm from the crystal.

The other arrangments for establishment of absolute activity scales are :

- foils at 5 and 10 cm from the NaI(Tl) spectrometer
- foils at 8 cm from a 68 cc. Ge(Li) diode : for the measurements, performed at SEIBERSDORF, ^{51}Cr was used as a substitute to ^{115m}In.

For all four counting conditions, the photopeak efficiency has been determined as a function of energy for centered, nearly-point sources and these calibrations were independent of each other in three cases.

The calibration sources for the principal counting arrangment were essentially[(a)] prepared by the CEN-SCK Absolute Radioactivity Section and were

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(a) Some intercomparisons have also been performed with CBNM, EURATOM, Geel.

assayed by 4 $\pi\beta$-γ and 4 $\pi\beta$ techniques; these sources were first counted at contact of the NaI(Tl) spectrometer on their vyns supports immediately after their standardization; they were then transferred on an aluminium backing diam. 10 mm, 0.1 mm thick, encapsulated in a thin aluminium wrapping, and counted again. Geometrical corrections necessary to apply the derived photopeak efficiencies to the actual activation foils are small, despite of the short separation distance from the crystal : typically $\sim$ 0.3% axial geometry correction for 0.127 mm thick foils and $\sim$ 0.2% radial geometry correction for a foil diameter of 10 mm. In this geometry however, corrections for gamma ray self absorption within the foils are important, e.g. 1.8% for indium. This work was performed in 1965 [28]. When ^{137}Cs sources prepared at this time are remeasured, no change in their disintegration rate can be observed after decay correction, as expected for normal operation performances.

At the separation distances of 5 and 10 cm from this same NaI(Tl) spectrometer, the photopeak efficiencies have been measured recently, essentially on the basis of standard sources supplied by the "Laboratoire de Métrologie des Radio-Isotopes" (LMRI, SACLAY, FRANCE); part of these sources are disintegration rate standards (activities given in microcuries) and part are emission rate standards (gamma/min. 4 π).

The Ge(Li) spectrometer at SEIBERSDORF, IAEA, was calibrated by means of in-home standard sources.

3.4. Intercomparison of absolute activity scales

A series of ^{51}Cr reference sources were prepared by deposition and evaporation of an active drop in an aluminium encapsulation simulating approximately a dosimetry foil diam. 10 x 0.13 mm. Such sources are appropriate for interlaboratory transfer of ^{115m}In calibration data for NaI(Tl) gamma ray spectrometers because the difference in photopeak efficiencies at 320 and 336 keV is small - around 3%, depending on geometry - and can be determined accurately, even on a straightforward computational basis, e.g. variation of total efficiency times the photofraction variation. In table III are documented the intercomparative measurements performed for such a reference ^{51}Cr source.

Similarly, an activated nickel dosimetry foil has been exchanged for intercomparison of ^{58}Co absolute activity measurements and the corresponding comparison data are gathered in table IV.

It is seen that on the average, the two laboratories are in agreement within uncertainties for the chromium-51 absolute activity scale and differ by 2% for cobalt-58. Further work is needed to resolve this discrepancy.

The absolute reaction rate data for indium and nickel reported below have been adjusted so as to be consistent with an interlaboratory activity scale defined as straight average between the IAEA and mean MOL scales illustrated by tables III and IV.

4. IRRADIATIONS AND RESULTS

Table V summarizes the effective irradiation times and nominal BR1 reactor power levels for the seven main runs performed in order to obtain the raw reaction rate data gathered in table VI.

Note that, except in the case of rhodium, the quoted statistical errors are external ones resulting from the combination of counting data for two (or more) foils exposed together in a sandwich stack and gradient corrected; the statistical weight for each foil is defined from the standard deviation of the mean of four to five individual counts.

In three of the runs, measurements were simultaneously achieved in $\Sigma\Sigma$ and in two fission-spectrum source-detector assemblies; all relevant physical and geometrical details are given in section 2.

Additional runs provided the already mentioned background and gradient reaction rate data for the fission spectrum assemblies.

The effective irradiation time $\bar{T}(\lambda)$ for activation of a radioisotope with half life $T_{1/2} = \ln 2/\lambda$, exposed to a time-dependent neutron flux specified by the power level profile function f(t), is deduced from the relationship

$$1 - e^{-\lambda \bar{T}(\lambda)} = \frac{\lambda e^{-\lambda T} \int_0^T f(t)\, e^{\lambda t}\, dt}{\bar{f}}$$

in which • T is the true, total irradiation duration

• $\overline{f}$ is some appropriately selected time-averaging of f(t), e.g.

$$\overline{f} = \int_{\Delta t} f(t)\, dt$$

where Δt is taken here as the true exposure duration at the nominal reactor power.

In the actual conditions of these experiments, the dependence of $\overline{T}$ on λ is negligibly weak for all runs, except the first two ones. These are just the high power, relatively long irradiations which are required to generate proper activation rates of the nickel foils, but are inadequate for rhodium measurements.

The cobalt monitoring foils in the fission spectrum assemblies are intended at checking against possible systematic bias related to

a) the significant differences in flux profiles throughout these irradiations

b) the large, e.g. up to two decades, fluence span to be encompassed by the indium foils.

When compared to indium in the runs performed at a similar reactor power level and effective irradiation time, they furthermore allow to judge in a sensitive way the neutronic reproducibility of these assemblies; the central fast flux level for a same total fission neutron source strength heavily depends on the separation distance between the fission discs : the shortest, the most. For the assembly sketched on fig. 1, the cobalt foils closely detect the 2π-average thermal neutron flux impinging on the fission foils, which is proportional to the source strength per unit fission foil weight. The data in table VII suggest an excellent reproducibility of the source-detector assemblies in these fission-spectrum experiments, as well as the existence of some

systematic error in the reaction rate ratios displayed in table VI, due to causes a) and b) outlined above. This is supported by gold foils exposed in monitoring reference locations for ΣΣ. This bias has been accounted for only by linear propagation of a combined, ± 0.6% (2 σ) systematic error on the final reaction rate ratios because the application of a correction based on gold and cobalt monitors was felt somewhat controversial : the evidence exists for the need of some correction within the quoted uncertainty, but in views of the critically bound sampling population, it is not statistically meaningful (counting statistics, limited reproducibility versus angular fluxes, and possible geometry-induced spreads as implicated from detailed mechanical controls).

In table VIII is presented a thorough evaluation of all uncertainties involved in this work. This table is self-explanatory.

Table IX displays the final absolute reaction rate ratios established by these measurements, as well as their estimated accuracy.

5. DATA APPRAISAL

Absolute spectral average cross sections implicated by this work are tentatively forwarded in table X on the basis of the normalizations proposed in section 2.2., relations (1) and (2). They are also compared to the predictions of selected differential data. Within respective uncertainties, satisfactory agreement is observed.

A brief discussion is relevant.

The present integral reaction rate ratio $^{58}Ni(n,p)^{58}Co/^{115}In(n,n')^{115m}In$ in the ^{235}U fission spectrum is equal within errors to the one previously measured at MOL in a somewhat heavier source-detector assembly, when this earlier result - obtained in one of the presently used counting geometries - is rescaled to the improved absolute activity scales of this work; this provides increased confidence into the assessment of uncertainties for the domina-

ting, neutron-field related correction in fission spectrum integral measurements for threshold reactions, e.g. the perturbation by neutron scattering and absorption within the source-detector assemblies.

Reasonable agreement is also observed on this nickel-to-indium ratio with the most recently reported integral measurements [24]; however, the large discrepancy on absolute scales remains, as a result of the normalization adopted here, and this discrepancy may even be underestimated if the current results of the new systematical measurements under progress at MOL stand firm.

The present integral results in $\Sigma\Sigma$ and in the ^{235}U thermal fission neutron spectrum decisively support the differential energy cross section determinations of BUTLER and SANTRY [29] for the $^{103}Rh(n,n')^{103m}Rh$ reaction below a few MeV and rule out both the conflicting experimental data and/or suggestions of some recent investigations [30][31]; this is particularly gratifying in views of the strong need for the selection of a rhodium differential-energy cross section measurement that provides consistency with integral data in dosimetry benchmark neutron fields.

6. CONCLUSIONS

The present experiments are likely to contribute noticeably to the standardization of nuclear data for reactor neutron dosimetry.

The circulation by the IAEA of calibrated sources of ^{103}Pd, ^{51}Cr and ^{58}Co is expected to provide the direct and precise channel needed to relate with each other and with this work the $^{103}Rh(n,n')^{103}Rh$, $^{115}In(n,n')^{115m}In$ and $^{58}Ni(n,p)^{58}Co$ reaction rate measurements performed in dosimetry benchmark neutron fields and, more generally, in all dosimetry applications.

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TABLE I. CORRECTIONS[a] AND ESTIMATED UNCERTAINTIES RELATED TO THE NEUTRON FIELD PURITY AND UNIFORMITY IN THE FISSION SPECTRUM EXPERIMENTS

CORRECTION SOURCE	CORRECTION FACTOR FOR SOURCE SEPARATION DISTANCE OF	
	(0.836 ± 0.01) cm	(2.03 ± 0.01) cm
Neutron scattering and absorption in source-detector assembly	1.00 ± 0.01 [b]	1.000 ± 0.005 [b]
Cavity wall return background	0.9997 ± 0.0001 [c]	0.9992 ± 0.0001 [c]
Reactor neutron background	1.000 ± 0.000	1.000 ± 0.000
Photoactivation RHODIUM	0.9891 ± 0.0015	0.9811 ± 0.0040
Photoactivation INDIUM	0.9968 ± 0.0010	0.9944 ± 0.0015
Photoactivation NICKEL	1.000 ± 0.000	1.000 ± 0.000
Streaming from other source-detector assembly (if any) [d]	0.9987 ± 0.0002	0.9962 ± 0.0002
Flux gradients[e] (typical)	0.9990 ± 0.0005	0.9995 ± 0.0005
Max. rms sum of uncertainties	± 1.01%	± 0.65%
Max. sum of uncertainties	± 1.21%	± 0.96%

(a) Given as factors by which to multiply the individual raw reaction rate data.

(b) The actual corrections for each of the investigated reactions are not expected to exceed the quoted uncertainties.

(c) Depends weakly on the considered reaction.

(d) This streaming component, when present, affects all reactions to identically the same extent and the error is thus not propagated.

(e) Applied to one reaction rate only when forming ratios.

TABLE II. DECAY SCHEME DATA ACCEPTED FOR THE EVALUATION OF THE MEASUREMENTS

NUCLIDE	DETECTED RADIATION AND ENERGY	RADIATION INTENSITY (% per decay)	HALF LIFE	COMMENTS
^{103m}Rh	20.2 keV $\bar{K}_{\alpha}$ + 23 keV $\bar{K}_{\beta}$ X-rays	6.76 ± 0.02 [a]	56.116 ± 0.009 min. [b]	
^{103}Pd	"	-	16.96 ± 0.06 d [c]	Used only as reference for control of equipment stability and transfer of calibration between laboratories [a]
^{115m}In	336.2 ± 0.1 keV [d] gamma ray	45.9 ± 0.1 [e]	4.50 ± 0.02 h [d]	
^{51}Cr [f]	320.07 ± 0.01 keV gamma ray	9.83 ± 0.14	27.70 ± 0.04 d	
^{58}Co [f]	810.75 ± 0.015 keV gamma ray	99.45 ± 0.01	70.78 ± 0.10 d	Coincidence summing corrections (if any) based on BAMBYNEK and LEGRAND evaluated decay scheme (f)

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(b) GÜNTHER, E.R., KNAUF, K., WALZ, K.F., JARI 24, (1973) 87

(c) CZOCK, K.H., HASELBERGER, N., REICHEL, F., POPA, S., Report IAEA/RL-23-1, 1974

(d) HELMER, R.G., GREENWOOD, R.C., "Evaluated Decay Scheme Data", Trans. ANS vol. 17, p. 517 (1973)

(e) HANSEN, H.H., DE ROOST, E., VAN DER EIJK, W., VANINBROUKX, R.,- Z. Physik 269 (1974) p.155

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TABLE III. INTERCOMPARISON OF ACTIVITY MEASUREMENTS FOR A REFERENCE ^{51}Cr SOURCE[a]

DETECTOR	SOURCE-TO-DETECTOR DISTANCE (cm)	RESULT, dps[b]	CALIBRATION METHOD
NaI(Tl) 3" x 3" MOL	0.55[c]	41720[d]	Photopeak efficiency[e] derived from 4 π β-γ sources[f]
	5	41436[d]	" [g]
	10	41570[d]	" [g]
	5	40674[e]	Photopeak efficiency[d] derived from LMRI standard[g] (γ/min. 4 π)
	10	41108[e]	"
	AVERAGE RESULT	41233(± 1.7%)	
Ge(Li) 68 cc. IAEA	8	41247(± 1%)[d]	Photopeak efficiency derived from 4 π β-γ sources

[a] Drop deposited in a aluminium encapsulation simulating a diam. 10 x 0.13 mm dosimetry foil. [b] On 6-6-74 12h00 GMT time.

[c] Source placed at contact and on top of crystal package; geometry as for ^{115m}In measurements.

[d] Independent on radiation intensity per decay.

[e] For a radiation intensity per decay of 0.0983 ± 0.0014.

[f] 1965 work [28] (actually measured - and not interpolated - efficiency). [g] 1974 work.

TABLE IV. INTERCOMPARISON OF ACTIVITY MEASUREMENTS FOR A REFERENCE ^{58}Co SOURCE[a]

DETECTOR	SOURCE-TO-DETECTOR DISTANCE (cm)	RESULT, dps[b]	COMMENTS ON CALIBRATION METHOD
NaI(Tl) 3" x 3" MOL	0.55[c]	11195 (± 0.2%)[d]	Photopeak efficiency based on 1965 work[28][e][g]
	5	11093 (± 0.3%)[d]	Photopeak efficiency interpolated[f][g], 1974
	10	11203 (± 0.3%)[d]	"
	AVERAGE RESULT	11164 (± 1%)	
Ge(Li) 68 cc. IAEA	8	11375 (± 1%)	Photopeak efficiency interpolated[g]

(a) Activated nickel dosimetry foil diam. 1/2" x 0.1 mm.

(b) On 24.1.74 8h00 GMT time. Corrected for γ ray self absorption.

(c) Source placed at contact and on top of crystal package; geometry for nickel cross section measurements.

(d) Systematic error not included.

(e) 810.75 keV efficiency taken as (1.030 ± 0.002) times 834.83 keV efficiency obtained from a CBNM ^{54}Mn standard [28].

(f) Efficiency curve mostly based on LMRI standard sources.

(g) Corrections for coincidence summing are respectively 9.8%, 1.8% and 0.69% at 0.55, 5 and 10 cm from NaI(Tl) and negligible (< 0.1%) for Ge(Li).

TABLE V. SOME DATA RELATIVE TO THE JOINT IRRADIATIONS IN ΣΣ AND THE FISSION SPECTRUM

Run n°	Effective irradiation time (sec)	Reactor power level (nominal)
1	14765[(a)]	3.2 MW
2	45246[(a)]	3.2 MW
3	3805	400 kW
4	3600	1 MW
5	3620	400 kW
6	3620	400 kW
7	3621	400 kW

(a) For the $^{58}Ni(n,p)^{58}Co$ reaction.

TABLE VI. ABSOLUTE RAW REACTION RATES PER NUCLIDE (x 10^{17}) AND RAW REACTION RATE RATIOS MEASURED IN ΣΣ AND IN THE ^{235}U THERMAL FISSION NEUTRON SPECTRUM[a]

	ΣΣ			Fission spectrum (s = 0.836 cm)[b]			Fission spectrum (s = 2.03 cm)[b]		
Run Number	Nickel	Indium	Ratio	Nickel	Indium	Ratio	Nickel	Indium	Ratio
1	6.034(±0.3%)	12.66 (±0.6%)	0.4766(±0.7%)	5.041(±0.2%)	8.855(±0.3%)	0.5693(±0.4%)	3.156(±0.3%)	5.487(±0.5%)	0.5752(±0.6%)
2	6.081(±0.3%)	12.88 (±0.3%)	0.4721(±0.4%)	5.491(±0.5%)	9.641(±0.3%)	0.5695(±0.6%)	3.161(±0.4%)	5.514(±0.2%)	0.5733(±0.5%)
Run Number	Rhodium	Indium	Ratio	Rhodium	Indium	Ratio	Rhodium	Indium	Ratio
3	7.303(±0.9%)	1.462(±0.5%)	4.995 (±1.0%)	-	-	-	-	-	-
4	-	-	-	-	-	-	6.727(±1.5%)	1.687(± 1%)	3.988 (±1.8%)
5	7.607(±0.7%)	1.507(± 1%)	5.048 (±1.2%)	5.004(±0.8%)	1.279(± 2%)	3.912(±2.1%)	-	-	-
6	-	-	-	4.499(± 1%)	1.162(±2.5%)	3.872(±2.7%)	-	-	-
7	7.342(±1.4%)	1.477(± 1%)	4.971 (±1.7%)	4.534(±0.5%)	1.168(± 1%)	3.882(±1.1%)	2.623(±0.2%)	0.667(± 1%)	3.932 (± 1%)

(a) Quoted uncertainties statistical only

(b) s = separation distance between the enriched uranium metal disc sources driving the fission spectrum

TABLE VII. RELATION BETWEEN THE $^{115}In(n,n')^{115m}In$ REACTION RATE PER NUCLIDE AND THE INCIDENT THERMAL NEUTRON FLUX IN FISSION SPECTRUM EXPERIMENTS[a]

Run number	Central indium reaction rate	Thermal neutron flux[b]	Ratio [c]
1	8.855, -17	1.767, +9	54.52, -27[d]
2	9.641, -17	1.772, +9	54.41, -27
5	1.279, -17	2.321, +8	55.11, -27
6	1.162, -17	2.124, +8	54.71, -27
7	1.168, -17	2.113, +8	55.28, -27

(a) Source-detector assembly with a source separation distance of 8.36 mm.

(b) 2π integral of the angular flux impinging on the source discs.

(c) The random uncertainty on the individual ratios is of the order of $\pm$ 0.5% for runs 1 and 2, of $\pm$ 1 - 2.5% for runs 5 to 7.

(d) A scaling by 8.8% has been applied to make this figure comparable to the other ones : the uranium-235 sources used for this measurement had a weight 8.8% lower than for the other runs, thus producing a correspondingly weaker fast flux for the same incident thermal flux.

TABLE VIII. ESTIMATED UNCERTAINTIES OF ABSOLUTE REACTION RATES

ERROR SOURCE	ESTIMATED UNCERTAINTIES (%)		
	$^{103}Rh(n,n')^{103m}Rh$	$^{115}In(n,n')^{115m}In$	$^{58}Ni(n,p)^{58}Co$
Neutron field integrity[a] FISSION SPECTRUM	± 1.0 to ± 1.2	± 0.7 to ± 1.1	± 0.6 to ± 1.1
ΣΣ	± 0.5	± 0.3	± 0.3
Run-to-run reproducibility[b]	- (c)	- (c)	- (c)
Counting efficiency	± 0.4	± 1.0	± 1.0
Radiation intensity	included in efficiency	± 0.2	± 0.01
Half life	negligible	± 0.3	± 0.2
Radiation self absorption	± 0.5	± 0.2	negligible
Other counting corrections[d]	negligible	negligible	included in efficiency
Total uncertainty[e] FISSION SPECTRUM	± 2.0	± 2.5	± 2.0
ΣΣ	± 1.5	± 2.0	± 1.5

(a) See table I for fission spectrum. The uncertainty quoted for ΣΣ is related to the neutron field perturbation by the detectors.

(b) Includes counting statistics and reproducibility, weighting errors, irradiation flux profile corrections, neutron field stability.

(c) Directly assessed for reaction rate ratios. Not propagated here.

(d) Dead time losses, background, coincidence summing (if any), random summing.

(e) For a 95% confidence interval.

TABLE IX. FINAL ABSOLUTE REACTION RATE RATIOS[a]

REACTIONS	ΣΣ	FISSION SPECTRUM S = 0.836 cm[b]	FISSION SPECTRUM S = 2.03 cm[b]
$\frac{^{103}Rh(n,n')^{103m}Rh}{^{115}In(n,n')^{115m}In}$			
Average raw ratio	5.009	3.887	3.944
Corrected ratio	5.009	3.862	3.892
FINAL RESULT	5.01	3.88	
Systematic error	± 4%	± 5%	
Statistical error	± 1%	± 1%	
$\frac{^{58}Ni(n,p)^{58}Co}{^{115}In(n,n')^{115m}In}$			
Average raw ratio	0.4745	0.5694	0.5742
Corrected ratio	0.4745	0.5712	0.5774
FINAL RESULT	0.474	0.574	
Systematic error	± 4%	± 5%	
Statistical error	± 0.5%	± 0.5%	

(a) All quoted uncertainties estimated at the 2 σ confidence level

(b) S : separation distance between fission source discs in the fission spectrum assemblies.

TABLE X. IMPLICATED SPECTRAL AVERAGE CROSS SECTIONS AND PREDICTIONS FROM DIFFERENTIAL DATA

REACTION	SPECTRAL AVERAGE CROSS SECTIONS (mb) [a]				SELECTED REFERENCE FOR DIFFERENTIAL CROSS SECTION
	^{235}U FISSION SPECTRUM [b]		MOL-ΣΣ [c]		
	INTEGRAL DATA (this work)	DIFFERENTIAL DATA	INTEGRAL DATA (this work)	DIFFERENTIAL DATA	
^{103}Rh(n,n')^{103m}Rh	729 ± 20 ± 50	720 ± 40	294 ± 7 ± 22	295 ± 16	BUTLER, SANTRY [29]
^{115}In(n,n')^{115m}In	188 [d] ± 10 [e]	187.5 ± 15	58.6 [d] ± 3.0	57.1 ± 4.8	FABRY et al. [14]
^{58}Ni(n,p)^{58}Co	108.0 ± 2.7 [f] ± 8	111 ± 8.5	27.8 ± 0.6 [g] ± 1.9	25.6 ± 2.2	FABRY et al. [14]

(a) All uncertainties are 1 σ estimates. Both absolute and relative errors are quoted for the integral data.

(b) The WATT representation, average energy 2 MeV, is used to weight the differential-energy cross sections.

(c) Spectral shape as evaluated in [9].

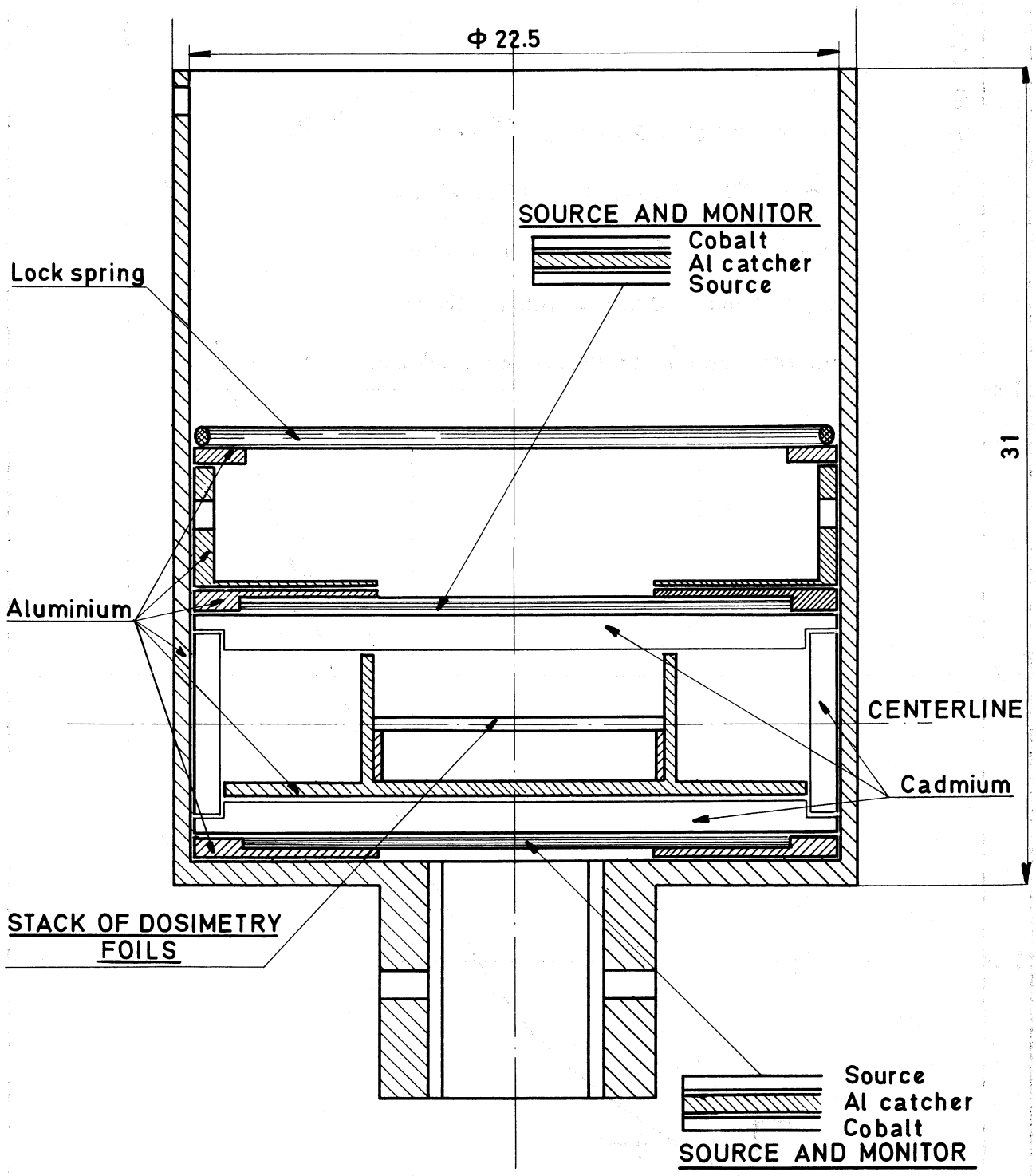
(d) Accepted for normalization of the ratio measurements.

(e) The error has been increased with respect to reference [23].

(f) The earlier MOL result [14], corrected to match the presently improved absolute activity scales and normalized to 188 mb for ^{115}In(n,n')^{115m}In, is equal to (110 ± 3.5) mb.

(g) The earlier MOL result [9], corrected to match the presently improved absolute activity scales and normalized to 58.6 mb for ^{115}In(n,n')^{115m}In, is equal to (28.0 ± 0.7) mb.

Fig.1. TYPICAL SOURCE-DETECTOR ASSEMBLY FOR ACTIVATION MEASUREMENTS IN THERMAL-NEUTRON INDUCED FISSION NEUTRON SPECTRA



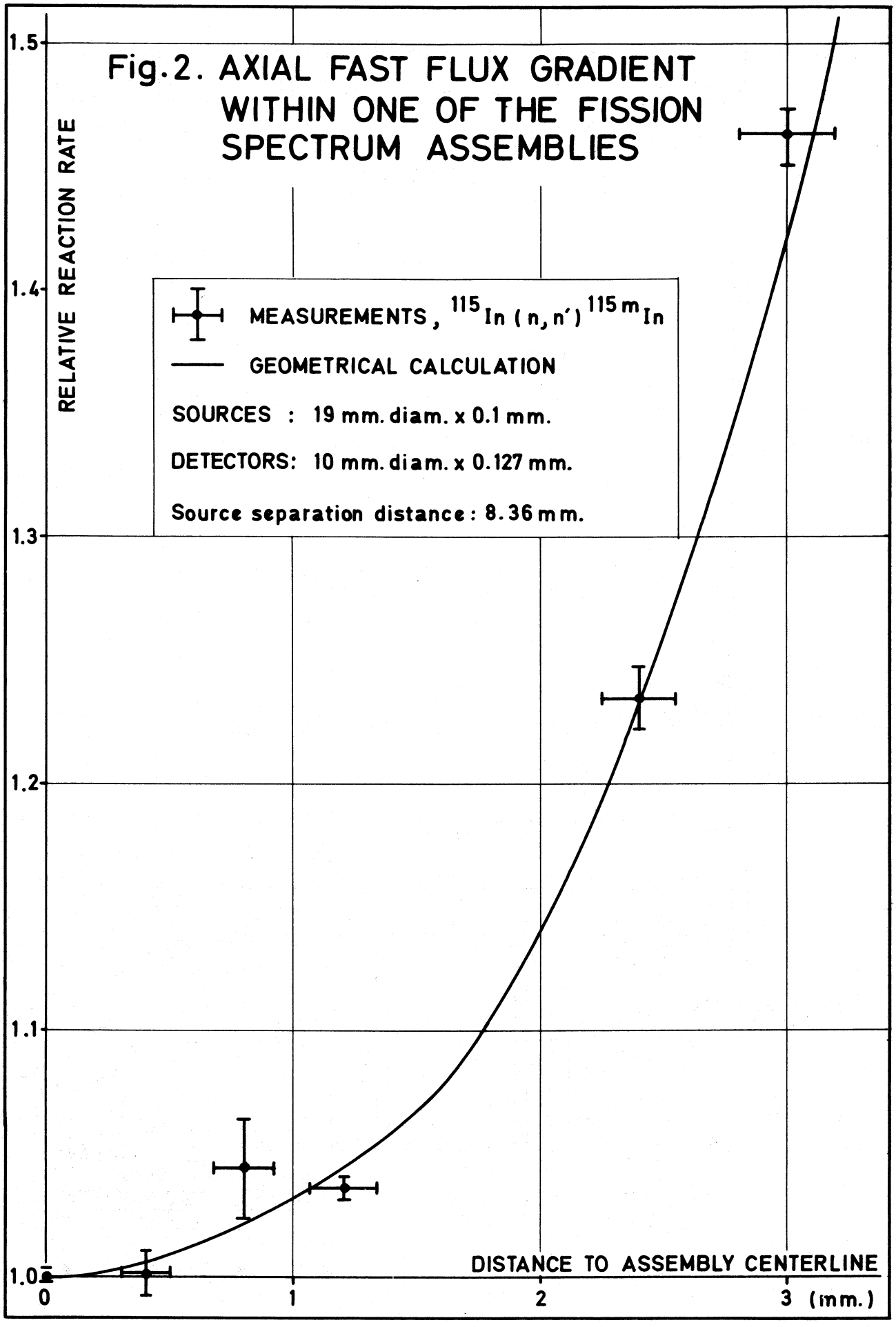


Fig.2. AXIAL FAST FLUX GRADIENT WITHIN ONE OF THE FISSION SPECTRUM ASSEMBLIES
RELATIVE REACTION RATE
1.5
1.4
1.3
1.2
1.1
1.0
MEASUREMENTS, $^{115}In(n,n')^{115m}In$
GEOMETRICAL CALCULATION
SOURCES : 19 mm. diam. x 0.1 mm.
DETECTORS: 10 mm. diam. x 0.127 mm.
Source separation distance: 8.36 mm.
DISTANCE TO ASSEMBLY CENTERLINE
0
1
2
3 (mm.)