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FISSION-NEUTRON SPECTRA : BRIEF REVIEW AND STATUS OF THE LONG-STANDING INCONSISTENCIES BETWEEN DIFFERENTIAL AND INTEGRAL DATA

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The energy distribution $\chi(E)$ of neutrons emitted in the fission process of heavy nuclides plays a significant role in various areas of technological relevance. Most noticeably, in terms of fission nuclear reactors :

- it describes the source-term energy dependence of the integro-differential linear transport equation, that governs the space and energy distribution of the neutron angular density
- for dosimetry of the neutron-induced materials property changes (and in other technical branches like, for instance, activation analysis), the uranium-235 thermal fission neutron spectrum $\chi_{25}(E)$ has sustainedly been used as basic reference : microscopic integral cross-sections $\bar{\sigma}_r(i,\chi_{25})$ for nuclear reaction r in isotope i, averaged over $\chi_{25}(E)$, have been quoted during about two decades so far to characterize or normalize the response of neutron dosimeters. This is because a substantial part of radiation damage is caused by neutrons of energies at which reactor neutron spectra do not differ appreciably from $\chi_{25}(E)$.

The accuracy requirements for fundamental physical quantities in the first area above, e.g. reactor physics, are usually more severe than in the second one, especially as related to the development of LMFBR's [1] . Paradoxically enough, in triggering-out the renewal of international concern about fission neutron spectra (best exemplified by the 1972 IAEA specialists meeting [2]), it might be that the data inconsistencies as faced by dosimetry [3] have weighted more heavily than the ones identified on the reactor physics side. In less applied research, after the impetus to look for scission neutrons and their relationship to fundamental fission mechanisms went over, e.g. around 1966, semi-empirical evaporation models [4] prevailed, imposing a simple, one-parameter, maxwellian-type picture of fission neutron spectra, and of their link to a more vital quantity : $\bar{\nu}$, the mean number of neutrons emitted per fission. By that time, suggestions [5] from microscopic integral cross - section ratio data, $\bar{\sigma}_r(i,\chi_{25})/\bar{\sigma}_{r'}(i',\chi_{25})$, that the entirely empirical WATT fit [6] to the oldest $\chi_{25}(E)$ measurements could be reliable, were not considered to even deserve attention.

In 1968, an extensive and well-documented study [7] of fission-neutron spectra with eight activation detectors calibrated by exposure to mono-energetic neutrons from the Los Alamos Van de Graaff, revealed "intractable discrepancies between the observed and predicted fission spectrum responses" : the cases of the fission reactions ^{235}U(n,f), ^{237}Np(n,f) and ^{238}U(n,f) were emphasized.

A representation of fission neutron spectra in a few discrete-energy groups, unfolded from the integral reaction rate ratios, suggested an average energy $\bar{E}_{25}$ for $\chi_{25}(E)$ as high as 2.20 MeV, in contrast with the value of 1.935 MeV [4] accepted in such widely used nuclear data files as, by that time, ENDF/B-I.

Absolute integral cross-section measurements appeared to provide further support [8][9] to such hard spectrum and an independent analysis [3] of the combined integral cross-section data from the above experiments led to $\bar{E}_{25}$ = 2.27 MeV.

It is rather clear to-day that the implication of integral microscopic cross section data, insofar as $\bar{E}_{25}$ is concerned, somewhat differ, depending on the selection of the corresponding differential-energy cross-sections <u>and</u> their uncertainties; the choice of the integral data themselves, within their present accuracy limits [10], has a less sensitive, though non-negligible impact. The differences however remain within estimated uncertainties as shown by table I. Nevertheless, integral data tend to suggest a slightly harder $\chi_{25}(E)$ spectrum than according to differential measurements, which encompass $\overline{E}_{25}$ values ranging from (1.86 ± 0.06) MeV [18] up to (2.05 ± 0.05) MeV [19] with a weighted average of 1.97 ± 0.04 MeV[20]. The discrepancy is close to or within mutual uncertainties.

The last statements above apply as well to measurements of the age to indium resonance energy in water, e.g. of the second moment of the spatial distribution of 1.44 eV energy neutrons slowing-down from a point fission source in infinite medium. The discrepancy between calculated and measured ages in water for ^{233}U and ^{235}U fission neutron sources is consistent with the discrepancy between calculated and measured eigenvalues for uranium-fueled, water moderated, homogeneous critical spheres [21].
The age discrepancy is known from long and is of such relevance that fast reactor experts even looked at the impact of $\overline{E}_{25}$ uncertainties on neutron spatial distributions, including within blanket zones [22][23].
These observations take the best of their weight when age data pertinent to ^{252}Cf sources are considered : age measurements here are the cleanest and most accurate ones [24] and recent differential data relative to the californium spontaneous fission neutron spectrum are of good quality. Table II shows similarly discrepant trends for $\overline{E}_{25}$ and for $\overline{E}_{52}$, the average energy for ^{252}Cf spontaneous fission. This again may indicate the standing of some differential-integral conflict regarding absolute spectral shapes, but not for the <u>ratio</u> of shapes; this last point is strongly supported by the most recent and fundamental integral cross-section data in χ_{25} and χ_{52} , as outlined below. This is a particularly gratifying outcome, which calls for reviewing briefly the status of another fundamental quantity, that persistently challenged measurers : the $\overline{E}_{29}/\overline{E}_{25}$ ratio.

The ratio of the plutonium-239 to uranium-235 fission spectrum average energies is particularly important in practice from the standpoint of LMFBR's core physics.
If the $\chi_{25}(E)$ and $\chi_{29}(E)$ are continuous functions depending on a few parameters it may be shown that precise measurements of suitably selected double integral

cross-section ratios $\frac{\bar{\sigma}_r(i,\chi_{29})/\bar{\sigma}_r(i,\chi_{25})}{\bar{\sigma}_{r'}(i',\chi_{29})/\bar{\sigma}_{r'}(i',\chi_{25})}$ allow to determine the shape parameters to almost an order of magnitude better than any known differential technique; this statement generally applies to any couple of continuous neutron spectra that do not differ too much from each other and is due to the fact that the uncertainties in the relative differential-energy cross-sections of detector reactions propagate little, as less as the two spectra are similar. It is seen from table III that the most recent differential ratio measurements tend to agree with integral data implications for $\bar{E}_{29}/\bar{E}_{25}$.

Another long-standing facet of the inconsistencies between differential and integral data in fission-neutron spectra lies in the general overprediction of the ^{235}U to ^{238}U fission rate ratio in thermal as well as in fast reactors. Inaccuracies in uranium-238 inelastic scattering data are responsible for part of this difficulty, to an extent that may be differently appreciated. It is most relevant to consider here the ratio $\bar{\sigma}_f(^{235}U,\chi_{25})/\bar{\sigma}_f(^{238}U,\chi_{25})$, which has been shown [10] to be a crucial milestone as well for the improvement and standardization of neutron dosimetry [35]. All published integral data for this quantity are summarized and commented in table IV.
As is the case for all fission-spectrum average cross-section ratio measurements, two considerations - only - dominate here the appraisal :

- what is the degree of spectrum purity
- what is the accuracy in determining the reaction rates.

The joint MOL-NBS measurements [37] have not yet been as extensively described as they deserve, but involve sustained and systematically coordinated efforts. The assessment of spectral purity remains a matter of current scrutinity insofar as the ~ 2.5% correction for neutron scattering and absorption within the source-detector assemblies is concerned. At present, this correction is the dominating factor of eventual bias related to spectrum distorsion. A similar remark applies to differential neutron spectrum measurements, a number of which, even among the most careful and recent ones, have not been corrected for such non-negligible [38], energy-dependent intrinsic experimental perturbations.(*)

Fission rate measurements by means of the NBS absolute fission chamber, which was the major, but not unique, instrument for the reaction rate determination, are well documented [39] and have been validated [40].

All these integral experiments lead to a weighted best value
$\bar{\sigma}_f(^{235}U,\chi_{25})/\bar{\sigma}_f(^{238}U,\chi_{25}) = 3.89 \pm 0.08$.

It may be regretted that this important quantity has been measured only by two groups programmatically connected since 1971, but this is balanced by the variety of experimental conditions used and the devoted amount of critical attention, sharpened by a sustained interest.

Predictions for the quantity above differ according to differential-energy cross-sections and spectral shapes accepted. Spectral uncertainties may induce differences of the order of ± 2.5%. A recent, simultaneous evaluation [41] of the fission cross-sections for ^{235}U, ^{239}Pu and ^{238}U gives a ratio of 4.25 for a Watt [6] representation ($\bar{\bar{E}}_{25}$ = 2.00 MeV) and of 4.44 for a Maxwellian ($\bar{E}$ = 1.935 MeV) [4] .

A brief look at the major differential ratio measurements from 1967 till March 1975 [42][43][44][45][46] shows that the $\sigma_{f_5}(E)/\sigma_{f_8}(E)$ in this evaluation may be high by up to 3 - 4% above 1.5 MeV - which is within the uncertainties quoted by the evaluators. If this is real, a energy scale calibration bias of ~ 50 ke in the sharp threshold rise below 2 MeV would suffice to remove the differential-integral discrepancy. The possibility of such energy calibration error has been pointed out [12] from systematics in integral-versus-differential microscopic cross-section ratios. At this time, the detailed, ~ 50 keV resolution measurements of LAMPHERE [47], were usually considered to provide the

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(*) The importance to carefully consider such small, "intimate" corrections may best be illustrated by revealing here that a ~ 1.2% error affects the recently published [37] MOL-NBS neptunium data : indeed, energy - dependent effects were believed small enough to justify application of a same, 1.6% perturbation correction to the ^{237}Np and ^{238}U fission rates, but the correction is only 0.4% for ^{237}Np, as now evidenced.

best $\sigma_{f8}(E)$ shape below the first plateau. The attention of integral measurers included a wider array of nuclear reactions, such as the ones of relevance to reactor neutron dosimetry. It is valuable to provide here a similar, though brief coverage.

In the present context, it is first interesting to examine, table V, the departures between fundamental fission rate ratios in the ^{235}U and ^{252}Cf fission-neutron spectra as recently computed and measured [37][48].
The consistency of discrepancies is remarkable and again points out at a gratifying agreement between differential and integral data regarding ratios of fission spectrum shapes. It is clear on another hand that either the absolute spectral shapes and/or differential-energy fission cross-section ratios involve significant errors.
This is confirmed by the results in table VI, which is an interim revision of a previous evaluation [10]; this current reevaluation accounts for recent integral measurements not covered before [48][49][50][51][52][53]. Table VI clearly places the remaining burdain (if any) of integral-versus-differential inconsistencies on differential-energy and/or integral cross-section data. The listed nuclear reactions have integral responses that span a large range of neutron energies and are variably sensitive to fission-spectrum shape inadequacies. For example, the ^{27}Al(n,α) differential-energy cross section is deemed accurate to ± 5% [54], but the derived integral cross section would differ by as much as ~ 30% for a 5% change in $\bar{E}_{25}$.

In conclusion, the main points in this brief review are :

- Integral implications of the ratios of average energies of the ^{252}Cf to ^{235}U and ^{239}Pu to ^{235}U fission-neutron spectra agree within errors with the most recent differential measurements in which same or similar approaches were adopted for investigating each couple of nuclides; integral data inherently provide specifications more accurate by about an order of magnitude, to the extent that reportedly possible structure [64] in differential spectrum data remains of negligible practical concern.

- Integral measurements tend to implicate slightly harder fission-neutron spectra than the bulk of differential experiments, but this trend is believed to lie within the present mutual uncertainties of differential and integral cross-section data and of differential neutron spectrometry.

- A more satisfactory analysis of remaining inconsistencies in the area discussed here requires the implementation of appropriate differential-energy cross-section error files as well as closier consideration of neutron scattering and absorption corrections within samples in most differential fission spectrum measurements.

- Fundamental differential-energy cross-sections like $^{235}U(n,f)$, $^{238}U(n,f)$, $^{239}Pu(n,f)$ and their ratios are not ascertained to the accuracies required for fast reactor design and operation; they do not match well enough accurate integral data in standard neutron fields and clean macroscopic fast neutron assemblies.

TABLE I : URANIUM-235 THERMAL NEUTRON-INDUCED FISSION NEUTRON SPECTRUM : AVERAGE ENERGY IMPLICATED BY MICROSCOPIC INTEGRAL CROSS SECTION MEASUREMENTS

REFERENCES	SOURCE OF INTEGRAL DATA	INPUT DIFFERENTIAL DATA	IMPLICATION FOR $\bar{E}_{25}$ (a)
GRUNDL 68 [7]	Own	Own and literature	2.20 ± 0.18
McELROY 69 [3]	[7] [8] [9]	SAND II [17]	2.27(± 0.06) (b)
McELROY 72 [11]	[7] [8] [9]	SAND II [17]	2.14 ± 0.07
FABRY et al 70 [12]	Own and literature [7] [15] [16]	Own evaluation	2.00 ± 0.05 (c)
KIMURA et al 73 [13]	Own	Own evaluation and measurements	2.00 (d)
McELROY 75 [14]	Literature evaluation [10]	ENDF/BIV	2.05 ± 0.03

(a) Except when otherwise indicated, $\chi_{25}(E)$ is represented as a multigroup flux spectrum to be unfolded from the integral data

(b) The error only quantifies the uniqueness of the unfolded spectrum, irrespectivily of the uncertainties in the integral and differential input data

(c) $\chi_{25}(E)$ represented by a simplified, light-heavy fragment groups evaporation model with a ∿ 18 % admixture of central neutrons. Three main parameters adjusted. Error deemed to be dominated by shape uncertainties in the ^{235}U energy-dependent fission cross section.

(d) Involves essentially a recommandation among conflicting literaturé representations

TABLE II : FISSION NEUTRON SPECTRA : AVERAGE ENERGIES IMPLICATED BY AGE TO INDIUM RESONANCE IN WATER

QUANTITY	NUCLIDE	
	^{235}U	^{252}Cf
Observation Measured age to indium resonance energy in water	26.6 ± 0.3 [25] 26.24 ± 0.33 [26]	28.69 ± 0.39 [24]
Average energy $\bar{E}_\chi$: implicated by age data	2.01 ± 0.05	2.21 ± 0.05
. from evaluated [20] differential spectrum measurements	1.97 ± 0.04 (a)	2.13 ± 0.025 (a)

(a) Uncertainties (1 σ) estimated by propagation of errors quoted on recommanded group fluxes [20]

TABLE III : FISSION NEUTRON SPECTRA : RATIO OF AVERAGE ENERGIES FOR PLUTONIUM-239 AND URANIUM-235

AUTHORS	$\bar{E}_{29}/\bar{E}_{25}$
Integral experiments	
KOVALEV 1957 [27] (a)	1.036 ± 0.005 (b)
BONNER 1961 [28] (c)	1.040
GRUNDL 1968 [7] (a)	1.039 ± 0.002
FABRY 1972 [29]	[1.039 ± 0.005] (d)
Differential measurements	
BARNARD 1965 [30]	1.085 ± 0.03
CONDE 1966 [18]	1.081 ± 0.05
BELOV 1969 [31]	1.048 ± 0.03
SMITH 1971 [32] (e)	1.075 ± 0.02
KNITTER 1975 [33]	1.035 ± 0.017
JOHANSSON 1975 [34]	1.051 ± 0.02

(a) Unfolding from activation measurements

(b) Reinterpretation [7] of the original experimental data (whose result is quoted [27] as 1.026)

(c) Unfolding from response ratios of sphere moderated neutron detectors

(d) Activation measurements mostly conceived as test of selection among previously conflicting data ; no unfolding originally presented

(e) Experiment especially aimed at checking the spectral shape ratio

TABLE IV : SUMMARY OF THE PUBLISHED MEASUREMENTS OF THE ^{235}U TO ^{238}U INTEGRAL FISSION CROSS SECTION RATIO IN THE ^{235}U THERMAL FISSION NEUTRON SPECTRUM[a]

Type of information	GRUNDL 1968[7]	FABRY et al 1968, 1970[9][12]	GRUNDL 1972[36]	MOL-NBS 1975[37]
Result	$3.85 \pm 0.23^{(b)}$	3.78 ± 0.18	3.71 ± 0.17	3.94 ± 0.08
Fission source	2 coaxial enriched metal discs Ø 10 x 0.13 mm	1 or 2 coaxial enriched oxyde discs Ø 19.5 x 2.8 mm and Ø 9.68 x 2.8 mm	1 enriched metal disc Ø 15.9 x 0.1 mm	0.1 mm thick enriched metal cylindrical shell Ø 33 x 77 mm or Ø 10 x 63 mm
Cavity type	10 cm diam. spherical in heavy water(d)	50 cm diam. spherical in carbon	30 cm side cubic in carbon	100 cm diam. spherical in carbon
Wall return correction for ^{235}U (c)	~ 8%(d) computed	~ 2% measured	~ 6% computed	~ 5% to ~ 40% (intentionnally varied) measured and computed agree to ≤ 0.5%
Fission detection	activation fission foils calibrated at Van de Graaf(b)	track recorders	fission ionization chambers	fission ionization chambers (three different types)

[a] All measurements have been performed in cavities hollowed out of reactor thermal columns.

[b] For a ratio $\sigma_{f5}(E)/\sigma_{f8}(E)$ = 1185 mb (± 2%)/505 mb at 2.75 ± 0.18 MeV.

[c] This is usually the highest and most questioned one in such experiments, but by far not the most critical (see text).

[d] Also control runs in 21 cm side cubic cavity in carbon, ~ 2% ^{235}U wall return.

TABLE V : INTEGRAL FISSION CROSS SECTION RATIOS IN STANDARD FISSION NEUTRON SPECTRA

FISSION NEUTRON SPECTRUM → NUCLIDES ↓	^{235}U		^{252}Cf	
	Measured[37] cross section ratio	Discrepancy measured/predicted(a)	Measured[48] cross section ratio	Discrepancy measured/predicted(a)
^{239}Pu/^{235}U(b)	1.505 (± 2.2 %)	1.050	1.500 (± 1.6 %)	1.042
^{238}U/^{235}U	0.254 (± 2.0 %)	1.071	0.266 (± 1.7 %)	1.060
^{237}Np/^{235}U	1.091 (± 3.0 %)(c)	1.028	1.105 (± 2.2 %)	1.023

(a) Convolution of ENDF/BIV cross sections with evaluated[20] differential neutron spectrum measurements

(b) Ratio largely insensitive to shape differences in fission neutron spectra

(c) The originally published result[37] is updated here by improved correction for neutron scattering perturbation within the source-detector assembly (see footnote p 5)

TABLE VI : ABSOLUTE MICROSCOPIC INTEGRAL CROSS SECTIONS[(a)] IN THE THERMAL-NEUTRON INDUCED URANIUM-235 FISSION NEUTRON SPECTRUM

REACTIONS[(b)]	RECOMMENDED[(c)] INTEGRAL DATA	PREDICTED[(d)(e)]	RATIO $\frac{\text{MEASURED}}{\text{PREDICTED}}$	COMPILATION OF ALL KNOWN ABSOLUTE INTEGRAL MEASUREMENTS *
$^{235}U(n,f)$	1220 (± 2.5 %)	1243 (± 3.5 %)	0.981	1202 ± 30 [49](f), 1220 ± 80 [55], 1330 ± 50 [56] , 1340 ± 40 [12]
$^{239}Pu(n,f)$	1830 (± 3.4 %)	1782 (± 5 %)	1.027	1796 ± 45 [48](f), 1792 ± 60 [57](f)
$^{237}Np(n,f)$	1331 (± 3.9 %)	1319 (± 6 %)	1.009	-
$^{115}In(n,n')^{115m}In$	193 (± 4.4 %)	181 (± 7 %)[(g)]	1.066	177 ± 9.5 [13], 204 ± 12 [58] (h)
$^{238}U(n,f)$	313.5 (± 3.2 %)	295 (± 6 %)	1.063	312 ± 4.5 [59], 310 ± 10 [60] (i)
$^{58}Ni(n,p)^{58}Co$	111 (± 4.7 %)	101.3 (± 7%)	1.096	102 ± 5.5 [13]
$^{32}S(n,p)^{32}P$	67.5 (± 5.6 %)	63.7 (± 7%)	1.060	65 ± 3 [61] , 74 ± 3 [8]
$^{27}Al(n,\alpha)^{24}Na$	0.708(± 5.6 %)	0.725(± 8 %)	1.024	0.644 ± 0.036 [13] , 0.63 ± 0.03 [61] 0.78 ± 0.03 [8]

(a) All uncertainties are for a 68.3 % confidence interval (1σ). Only are considered here the reactions with the best known $\sigma(E)$ data.

(b) Arranged in order of increasing energy response.

(c) Evaluation in progress, along an approach previously documented [10].

(d) Convolution of ENDF/BIV cross sections with evaluated differential neutron spectrum measurements

(e) Errors include $X_{25}(E)$ uncertainties assigned in [20] and rough appraisal of $\sigma(E)$ uncertainties.

(f) Original data obtained with ^{252}Cf sources and transferred to $X_{25}(E)$ case (0.2 % and 0.5 % adjustment for ^{235}U and ^{239}Pu fission respectively).

(g) ENDF/BIV data rescaled to be consistent with a branching ratio of 45.9 % [62] (radiation intensity per decay) for the ^{115m}In 336.2 keV gamma ray, as preferred in interpreting the integral measurements.

(h) A 1967 MOL result [8] of 200 ± 8 mb is not tabulated because it is now known [53] to be biased by ~ 2 - 3 %, insofar as the activation rate determination alone is concerned.

(i) A third, apparently consistent, measurement [63] is not quoted for reasons outlined elsewhere [10].

* Excluding the numerous and more accurate ratio data considered for evaluation purposes, column 2.

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