CONF-791058--58

FAST-NEUTRON CAPTURE CROSS SECTIONS OF IMPORTANCE IN TECHNOLOGICAL APPLICATIONS

by

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MASTER

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Prepared for

International Conference

on

NUCLEAR CROSS SECTIONS FOR TECHNOLOGY

October 22-26, 1979

U of C-AUA-USDOE

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

Operated under Contract W-31-109-Eng-38 for the

U. S. DEPARTMENT OF ENERGY

FAST-NEUTRON CAPTURE CROSS SECTIONS OF IMPORTANCE IN TECHNOLOGICAL APPLICATIONS

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The importance of the capture cross section of the major fertile nuclei, ^{238}U and ^{232}Th, leads to the consideration of these data. The ^{238}U (n,γ) cross section is considered of priority as it is part of the ^{238}U - ^{239}Pu cycle. Experimental techniques used in the measurements of these data are considered. Data measured more recently are compared with provisions made for the possible explanations of differing results. It is concluded that the ^{238}U (n,γ) cross section is known with ~5% above 10 keV and fulfills the uncertainty limit for this cross section set to achieve design accuracy for k_{eff} and the breeding ratio above 500 keV. Below 500 keV, the present uncertainty falls short of the required 1.5 - 3.0% uncertainty. Specific recommendations are made to resolve existing discrepancies and data uncertainties.

[Fast Neutron Capture, ^{238}U, ^{232}Th, ^{240}Pu, Fission Product Nuclei]

INTRODUCTION

Measurement techniques used in the detection of neutron capture events were reviewed by Chrien[1] at the last conference on Nuclear Cross Sections and Technology held in Washington in 1975. There are few new developments, if any, in this area to justify a reconsideration of general measurement techniques. The standard capture cross section, $\sigma_{n,\gamma}$ (Au), was reviewed even more recently by Paulsen[2] and only one new measurement[3] was reported since then and another was revised.[4] However, these additions or changes will cause little, if any, changes of the evaluated standard capture cross section. Some considerations of the standard capture cross sections will be made by Wasson.[5]

Energy is on many peoples mind these days, and therefore it appears appropriate to consider the area more closely related to this topic. It is not generally realized, though well known among experts, that for many countries, specifically the U.S., the largest energy reserves based upon presently achieved levels of technology and at acceptable costs is in uranium and thorium deposits. The utilization of these energy reserves requires however, the breeding of nuclear fuel by some scheme; the LMFBR being the conventional approach, and more recently accelerator breeding and fusion - fission hybrids under serious discussions. It is obvious that nuclear data of uranium and thorium must play a predominant role in the design and evaluation of the economics of specific systems regardless of the breeding scheme being considered. Indeed, sensitivity studies show[6] that the capture cross section of ^{238}U is the most important, surpassed only by the neutron production cross sections (ν, $\sigma_{n,f}$) of the fertile materials. Historically, the most intensively investigated and currently dominating systems involve the ^{238}U - ^{239}Pu cycle. For a variety of reasons the ^{232}Th - ^{233}U cycle is being considered more recently.

A recent study of a large LMFBR benchmark model[7] shows the following distribution of total capture events in the inner core of such reactor:

^{238}U	70%	Fe	4%
^{239}Pu	13%	Ni	2%
^{240}Pu	4%	Cr	2%
^{241}Pu	2%	Rest	3%

The predominant role of the major fertile material is obvious from these numbers, however, a more important role may be expected of the capture in ^{235}U and ^{240}Pu if ^{235}U or reprocessed plutonium are considered as fuel for a first generation of breeder reactors. Capture in ^{240}Pu and fission products becomes more important with increased burn-up or considerations of other parts of the fuel-cyle.[10] Though a case can be made for the specific importance of many nuclei, it is preferable in view of the restricted time and space available to concentrate on the major fertile materials with the emphasis on the fast neutron capture cross section of ^{238}U.

The great importance of the ^{238}U neutron capture cross section would lead up to expect that this reaction process is well documented and uncertainties are low. However, a number of problems persisting for the last 10-15 years suggest substantial uncertainty and limited knowledge. Other problems are of more recent origin. Outstanding problems are:

1. The C/E (calculated vs. experimental) discrepancy for the central reaction rate ratio $^{28}c/^{49}f$ (^{238}U (n,f)/^{239}Pu(n,f)). The calculated ratio is usually found to be 3-9% higher than experimentally determined values[8], resulting in differential data adjustments, or requests for lower evaluated differential data.

2. The small sample central reactivity worth problem which exists for most major fertile and fissile materials.[9] The C/E discrepancy was in the order of ~20% for ^{238}U and adjustments of the capture cross section of ~12% were proposed[9] in order to resolve the discrepancy.

3. A C/E discrepancy of ~13% for $^{28}c/^{28}f$ for GODIVA with ENDF/B-V data resulted in requests for ^{238}U (n,γ) data adjustments.

Calculations with more recent nuclear data files resulted in substantially reduced ^{238}U central worth discrepancies, specifically for advanced fuels.[11] Facilities with harder neutron spectra[12] find agreement between calculated and experimental small sample central reactivity worth.

Commonly accepted goals for the design accuracy in k_{eff} and the breeding ratio are 0.5-1.0% and 2%, respectively.[13] The breeding ratio is directly related to $^{28}c/^{49}f$ and uncertainty levels required to achieve design accuracy were given in the 1.5-3% range by Usachev and Bodkov,[14] and Bohn et al.[6]

These requirements are reflected in nuclear data request lists[15,16] which contain requests for the accuracy of the ^{238}U (n,γ) cross section or its ratio to ^{235}U (n,f) in the range of 1.5-3% for the lower keV region and 3-7% for the MeV range. Weisbin et al.[17] gave a rather detailed breakdown of the uncertainties which are required to obtain design accuracy over the keV-MeV range. Figure 1 shows the requested accuracy which appears plausible since it reflects sensitivity calculations[5,18]. Weisbin et al., also estimated the present uncertainty of the ^{238}U (n,γ) cross section which is also shown in Fig. 1. The estimated uncertainty exceeds at all energies and by large margins, the requested level of uncertainty which appears an acceptable result. However, specific features and the general magnitude of the uncertainty estimated by Weisbin et al., cannot be as easily agreed upon: Measurement techniques or nuclear properties which would justify the suggestion that the cross section is less uncertain by a factor of 2 in the 41-67 keV range than in the adjacent regions are unknown. The size of the suggested uncertainty might reflect the dispersion of all existing data and ignore the improvements achieved in recent years.

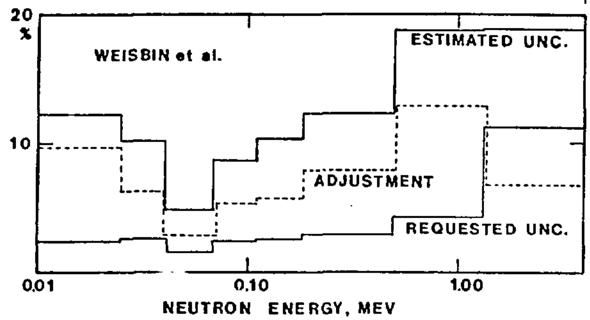

Fig. 1. Uncertainty limit for the ^{238}U (n,γ) cross section required in order to achieve design accuracy and the present uncertainty estimated by Weisbin et al.[23]. Also shown is the adjustment required to obtain agreement between differential and integral data.

In the following we will look in detail at measurements and data for ^{238}U (n,γ) in order to obtain a more realistic estimate of the present uncertainty of this important cross section. In doing so we will consider data which were presented after the conference on Neutron Cross Sections and Technology held at Knoxville in 1971.[20] Older data were discussed by Davey[19] and Poenitz.[20]

Some consideration will also be given to the capture cross sections of ^{232}Th and ^{240}Pu, and, as examples, the fission product nuclei of Rh and Pd.

II. EXPERIMENTAL DATA ANALYSIS

A. Experimental Data Interpretation

Before considering experimental data we should achieve some understanding about their interpretation. Cross section values are at best given with the following information:

E,	the energy of the measurement
ΔE,	the uncertainty of this energy
R,	the resolution
σ,	the average cross section
Δσ,	the estimated total uncertainty of σ
$\Delta\sigma_{stat}$,	the statistical uncertainty of σ
σ_{ST},	the standard cross section or reference cross section used in the measurements.

As pointed out in another paper of these proceedings[21,22], the true uncertainty is most likely larger than the quoted uncertainty because only some errors are accounted for by the experimentor. Insufficient accounting or recognition of uncertainties is only one reason why data are often barely agreeing within their quoted error bars. Another reason is that the energy uncertainties cause uncertainties of the cross section due to the energy dependence of the cross section and of the standard. Data sets measured with different resolutions might differ substantially if the measured cross section is not smoothly varying with energy.

B. Experimental Techniques used in the Measurements of ^{238}U and ^{232}Th Capture Cross Sections

Diffences found between various experimental data sets of fast neutron capture cross sections of ^{238}U and ^{232}Th exceed, by far, the levels of uncertainty found in other important cross section measurements, e.g., ^{235}U (n,f). Thus we may suspect that these differences and discrepancies are caused by the detection of the capture events. Techniques which are used in the detection of capture events are:

Absorption

The capture cross section is only a small fraction of the total cross section and therefore difficult to determine with this technique. Spherical shell transmission measurements were used[24,25] but require extensive Monte Carlo interpretations[26] which depend on many parameters. The structure of the cross sections found in the energy range below 100 keV imply local values of the average level spacings and strength functions which might be different from those determined in the lower-energy range and used in the Monte Carlo calculations.[23]

Activation

There are several reasons why the activation technique should be used in the measurement of ^{238}U and ^{232}Th capture cross sections. One reason is that reactor reaction rate measurements use this technique, thus compatibility can be checked if differential data also use activation. Another reason is that very specific calibration techniques exist which should, in principle, permit accurate determination of capture rates in ^{238}U and ^{232}Th. The alpha-emitters ^{243}Am and ^{237}Np decay to the daughter nuclei of the capture process, ^{239}Np and ^{233}Pa. Samples of ^{243}Am and ^{237}Np which were α-counted in low geometry detectors and which are in equilibruim with their daughters can then be used for the calibration of the γ-counting equipment which detects the decay of ^{239}Np or ^{233}Pa. The uncertainty for this calibration is not expected to exceed 1%.

Prompt γ-Detection Techniques

Several different prompt γ-detection techniques are in use. Large liquid scintillators[27] (LLS) or 4π-NaI-detectors[28] were used to absorb the total γ-energy emitted in the decay of the compound nucleus. The major problem in this technique, if applied to ^{238}U (n,γ) and ^{232}Th (n,γ), is the low neutron binding energy for these reactions. A large background for low energies requires the threshold for the detection of capture events to be set at ~2-3 MeV which results in an efficiency or only ~60-70%. Figure 2 shows the pulse-height spectra obtained with the 1100 litre RPI-LLS[29] (on the left side) and the 1300 litre

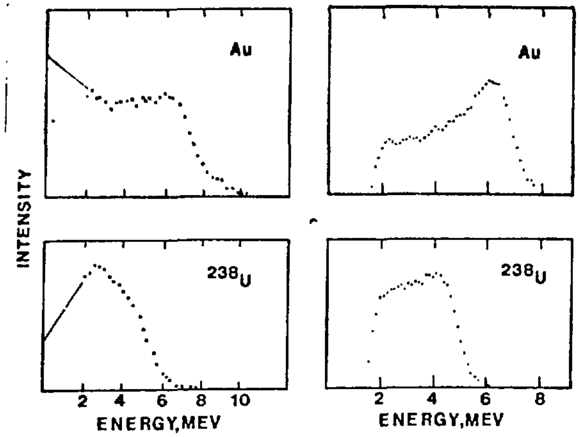

Fig. 2. Pulse-height spectra obtained with a large liquid scintillator. The spectra on the right were measured with a 1100 litre tank at RPI and the spectra on the left were obtained with a 1300 litre tank at ANL.

ANL-LLS[30] (right side) for gold and ^{238}U capture events. It is apprent that a large uncertainty exists due to the extrapolation to zero pulse-height. The figure shows the extrapolation chosen by RPI. The larger LLS from ANL shows improved spectra but the uncertainty for ^{238}U is still large and was estimated to be ~7%. One might suspect that the efficiency of these detectors is sensitive to changes in the primary γ-spectrum. Moxon-Rae detectors[31] were designed to have an efficiency which is proportional to the energy of the detected photon. This results in a proportionability to the total γ-energy released in the decay of the compound state. In other detectors with higher efficiencies, the proportionability to the photon energy is achieved by introducing spectrum weighting techniques.[32] Moxon-Rae detectors and their derivatives are expected to be sensitive to the absorption of ays in the sample and to losses due to electron conversion of transitions which occur on the deexcitation cascades.

Measurements of the ^{238}U (n,γ) cross section were made with all three prompt γ-detection techniques[31,33,34,35] with LINAC accelerators. In all cases the black resonance technique was used for the normalization of the data. It is of interest to compare these data for which the relative differences from the data by Moxon are shown in Fig. 3. The first noticable feature in these differences concerns the several large fluctuations of the values obtained by DeSaussure et al., vs. those reported by Moxon. These fluctuations cannot come from cross section fluctuations because both data sets were integrated over the same bin-widths. The neutron flux spectrum obtained with a LINAC accelerator shows considerable structure which is associated with resonances of light mass nuclei which are present in the structural materials of these facilities. Figure 3 indicates the energies at which resonances in aluminum occur. It appears that the fluctuations in the ratio of the cross section data by DeSaussure et al., and Moxon are coincident with resonances in Aℓ. A check against a theoretically calculated neutron capture cross section suggests that the fluctuations are due to the data by Moxon[31] and not due to the measurements by DeSaussure et al.[33].

The near constant difference between the LLS-tank data and the Moxon-Rae-detector results at low energies suggest a different efficiency for one or both detectors for the 6.6 eV resonance vs. the average over many S-wave resonances. Chrien[36] points out the anomalously high direct transition of 4.059 MeV between thermal energies and the 6.67 eV resonance. The difference between the LLS data and the Moxon-Rae detector result increases to ~14% at higher energies. This increase might be associated with spectra-changes due to an increasing amount of p-wave capture which is shown in Fig. 3 (right scale). The data by Quan and Block[35] were also obtained with a LLS and normalized with the black-resonance technique, however; an iron-filtered beam was used. Quan and Block measured the ratio of the detection efficiency for 24.3 keV neutron capture and capture in the 6.67 eV resonance. An ~11% difference (±3%) was found and attributed to p-wave contribution to the neutron capture at 24 keV. Without applying this correction the 24 keV value by Quan and Block would have agreed with the result by Moxon. However, if one were to conclude that a similar, though smaller correction should be applied for the LLS measurements by DeSaussure et al., the difference of those data relative to Moxon's values would increase. The ratio of data obtained with the pulse-height spectrum weighting technique relative to the data by Moxon is shown in Fig. 3 in a qualitative manner based upon figures given in Ref. 34. It appears that the pulse-height spectrum weighting technique results in the same normalization difference relative to LLS data as Moxon, however, it yields an even larger change in the range where p-wave neutron capture contributes substantially.

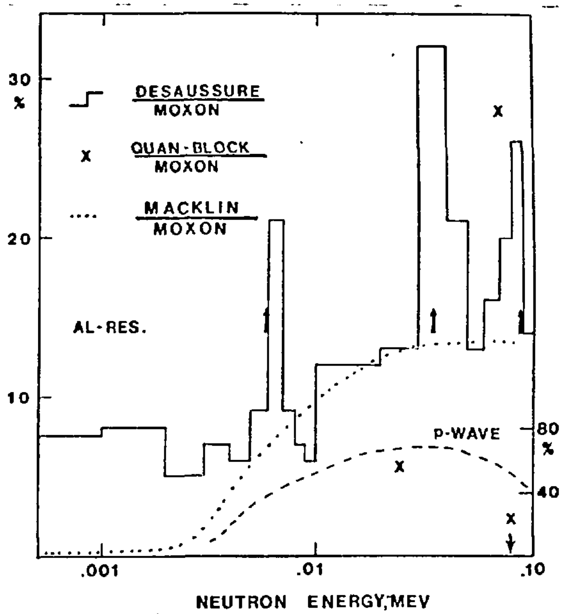

Fig. 3. Differences obtained for the γ^{38}U (n,γ) cross section measured with prompt-γ-detection techniques and the black-resonance normalization.

The values by Quan and Block[35] vs. the Moxon data differ at 69.8 keV and 80.3 keV from the 24.3 keV value by ~23% (higher) and ~12% (lower), respectively. The resolution is different for both measurements, thus this difference might indicate fluctuations of the cross section which will be considered next.

C. Cross Section Fluctuations

Fluctuations were found for many cross sections which once were considered to vary smoothly with

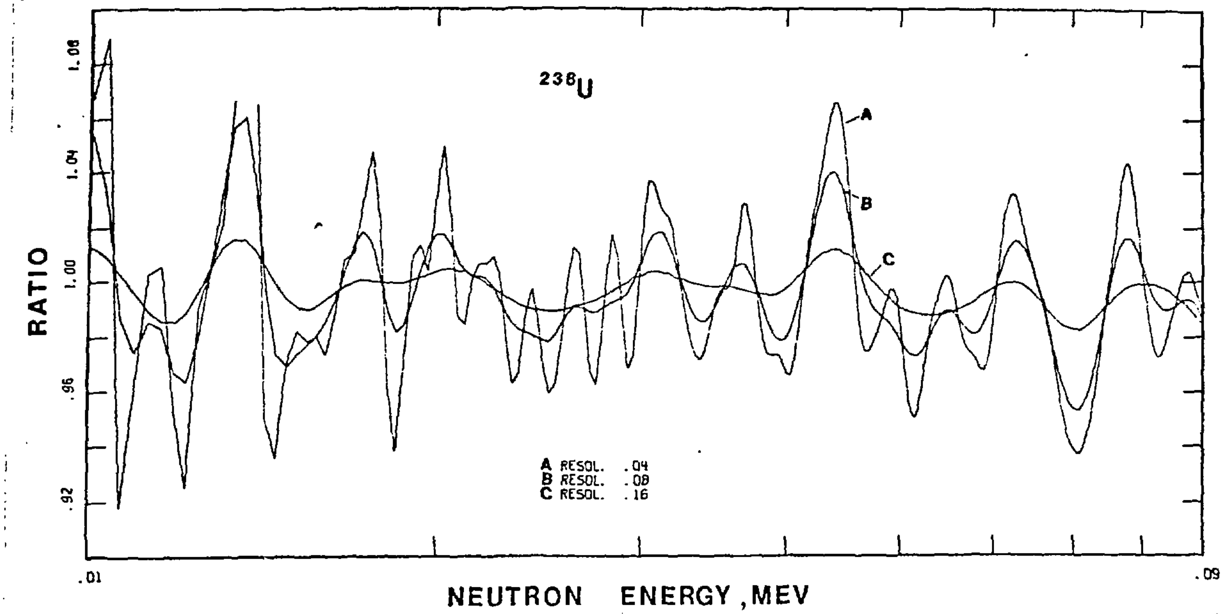

Fig. 4. Fluctuations of the ^{238}U (n,γ) cross section for different resolutions.

energy as long as the resolution was large compared with the compound resonance level spacing. These fluctuations are of importance in the interpresentation of differential cross section data though they should have little impact on the technological applications. Fluctuations of the ^{238}U (n,γ) cross sections were found by DeSaussure et al.,[33] and Spencer and Kaeppeler.[37] A physical interpretation and analysis of these fluctuations was given by Perez et al.[34] The present question is how much must a cross section value measured with the resolution $\alpha = \Delta E/E$ (FWHM) be corrected in order to correspond to the average cross section which varies smoothly with energy (large α). The present analysis of ^{238}U (n,γ) cross section fluctuations is based upon the data by DeSaussure[33] and is similar to the analysis of ^{235}U (n,f) cross section fluctuations by Bowman et al.,[38] but differs with respect to the quoted quantity. In the present analysis $\sigma(\alpha)/\sigma$ ($\alpha = .30$) was derived which is independent of the cross section normalization and can be directly used in order to apply a correction to measured data. The averaging was carried out with a Gaussian:

$$\frac{\sigma(\alpha,E)}{\sigma(E,\alpha=.3)} = \frac{1}{a\sqrt{2\pi}} \; \frac{\sigma(E')}{\sigma(E,\alpha=.3)} e^{-\frac{(E-E')^2}{2a^2}} dE'$$

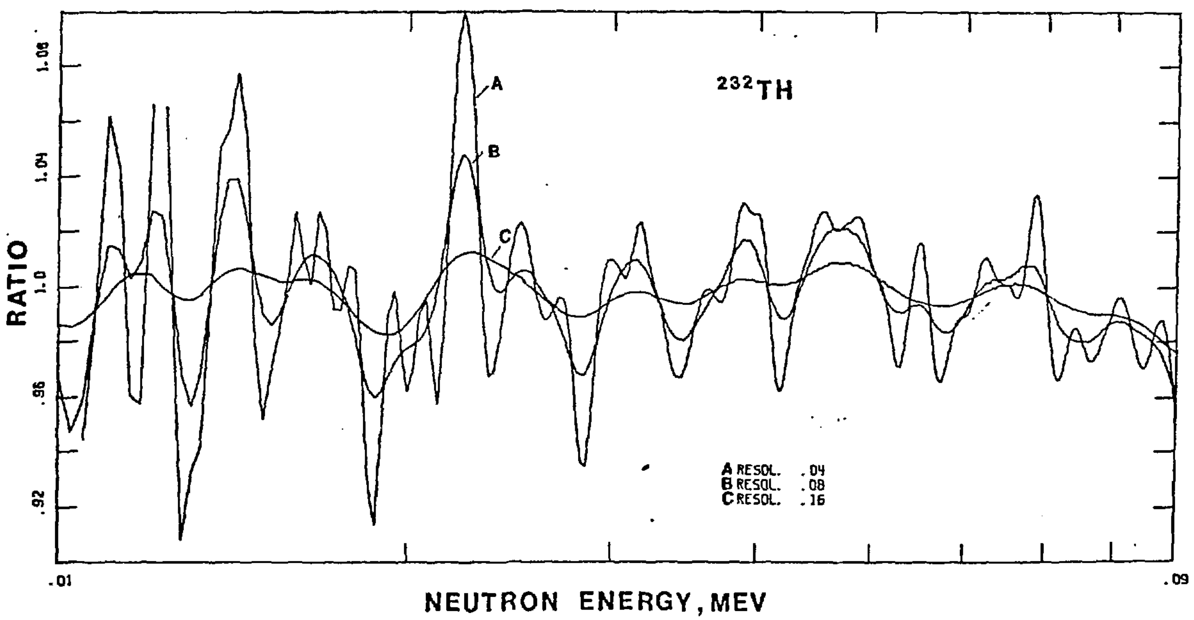

Fig. 5. Fluctuations of the ^{232}Th (n,γ) cross section for different resolutions.

with $a = \alpha E/2\sqrt{2\ln 2}$. Figure 4 shows that substantial corrections may need to be applied in the 10 - 100 keV range if average cross sections are to be derived from individual data points. However, corrections for measurements with a Sb-Be Photo-neutron source or an iron-filtered beam should not exceed 1-2%. A measurement at 30 keV at the ^{7}Li (p,n) ^{7}Be threshold should require only a minor correction. We can conclude from Fig. 4 that fluctuations of the ^{238}U (n,γ) cross section cannot explain the Fe-filter beam data at 69.8 keV and 80.3 keV by Quan and Block.[35] The discrepancy after applying the correction is actually larger at 69.8 keV than previously concluded.

Equivalent information on the fluctuations of the ^{232}Th (n,γ) cross section was derived from the data by Macklin and Halperin[39] and is given in Fig. 5. The fluctuations in ^{238}U (n,γ) and ^{232}Th (n,γ) are compared in Fig. 6 for α = 0.04. It is surprising to find nearly a one-to-one correspondence of maxima and minima between the two cross sections. This appears to hold up even for finer resolutions (α = 0.02 and 0.01) though worse statistics complicates the comparison. It also appears that the areas between major minima labeled "a" through "k" in Fig. 6 are very similar for the two nuclei. It can be concluded that the structure in these data was not caused by the experimental equipment because both data sets were measured with different detector systems[33,39] and the structure in the ^{238}U cross section was confirmed by a measurement at another facility.[37]

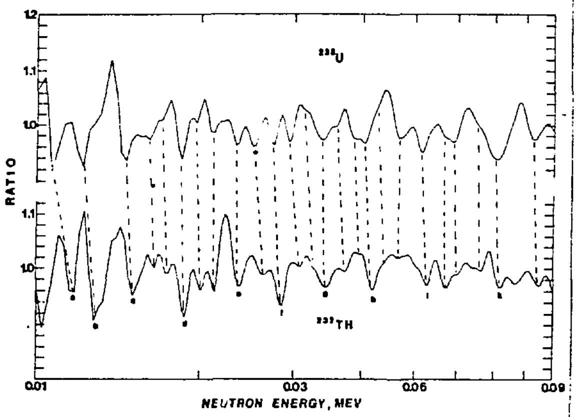

Fig. 6. Comparison of the fluctuations of the neutron capture cross sections of ^{238}U and ^{232}Th.

D. Resonance Self-Shielding and Multiple Scattering Corrections

At higher neutron energies, the cross sections vary rather smoothly with energy. The major effect which may cause erroneous measurement results besides the uncertainty of the detection efficiency are capture events which were caused by neutrons scattered within the sample. Inelastically scattered neutrons have a much higher capture probability due to the higher capture cross section at lower energies. The average flight path length within the sample increases for all scattered neutrons and thus increases the capture probability. Such events cannot be separated by the TOF technique and the effect is most obvious at 14 MeV where measured cross section values were too large by a factor of 4-5 due to these scattered neutrons.

At lower neutron energies the cross section has well separated compound nuclear resonances and resonance self-shielding reduces the measured effective cross section. The effects caused by neutron scattering and resonance self-shielding were treated by Schmitt,[40] Dresner,[41] and Macklin[42] with analytical approximative methods. Bogard,[43] Miller and Poenitz,[26] and Froehner[44] calculated these effects with Monte Carlo techniques. The most frequently used solutions for correcting resonance self-shielding and multiple scattering are those given by Schmitt, Dresner and Macklin. Figure 7 shows the correction calculated by DeSaussure et al.,[33] for a sample with a density of 0.0028 at /b. between 15 and 100 keV. Calculations were carried out for the same sample with the Monte Carlo code SESH by Froehner at several energies. These reults are also shown in Fig. 7. Additional calculations were carried out with a Monte Carlo code which was developed for the higer energy region and thus does not treat resonance self-shielding. However, first order resonance self-shielding effects could be estimated with another code and the combined effect is also shown in Fig. 7. The comparisons of the results from the Monte Carlo techniques and the analytical treatment of the resonance self-shielding and multiple neutron scattering effects shows a difference which exceeds the required uncertainty limit for the ^{238}U (n,γ) cross section in this energy range. The computer code SESH developed by Froehner will be shortly available from the National Computer Code Center at Argonne National Laboratory.

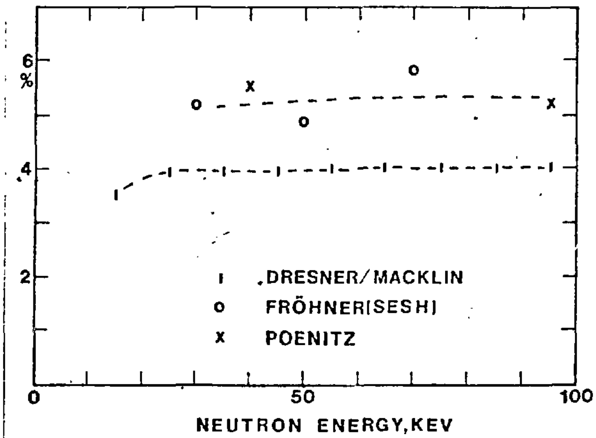

Fig. 7. Comparison of resonance self-shielding and multiple scattering corrections calculated with Monte Carlo techniques and analytical methods.

III. DISCUSSION OF THE EXPERIMENTAL DATA BASE

It is common to use double logarithmic scales for the graphical display of capture cross section data. This choise best accommodates the range of the data which change by ~2 orders of magnitude between 10 keV and 4 MeV. However, data differences which exceed by far the required uncertainty limit for the ^{238}U (n,γ) cross section are suppressed in such displays and are near indistinguishable, therefore in this presentation we are using instead, displays of $\sqrt{E} \cdot \sigma(E)$ which removes about one order of magnitude from the range of the data and permits the display with a linear scale.

A. The Utilization of Independent Experimental Data to Derive Capture Cross Sections with Theoretical Model Calculations

There are substantial discrepancies between some of the experimental data as was discussed before.

Therefore, it should be worthwhile to consider whether other experimental data can be utilized in order to derive additional information which might be helpful in deciding which capture cross section data are realistic and which are not. The capture cross section can be calculated in terms of the statistical model with neutron transmission coefficients derived from the optical model. The gamma strength function, (Γ_γ/D), can be calculated with a giant-dipole resonance for the γ-transition probability and a Fermi-gas model for the level density. In order to follow this outline a substantial number of parameters must be determined with other experimental data. For this presentation we used measurements of the total neutron cross section[44] and of elastic and inelastic scattering cross sections[45] in a fitting procedure in order to determine the optical model parameters for the calculation of the neutron transmission coefficients.[54] The parameters of the giant resonance γ-transition probability can be determined from (γ,n)-cross sections, however, it is common to utilize Γ_γ and ⟨D⟩ values determined with resonances observed in measurements at low energies (1-2 keV above the neutron binding energy) for normalization of the S-wave gamma strength function. Table I shows some of the more recent values and indicates the substantial uncertainty which appears to be mainly due to uncertainties of the average level spacing, ⟨D⟩. Figure 8 shows the resulting uncertainty for $\sqrt{E}\cdot\sigma$ of ^{238}U (n,γ). Additional parameters which must be considered are the spin-cut-off parameter, σ, and the level density formula parameters, a, which is related to the nuclear temperature.[53] Using the observed average level spacing, ⟨D⟩, as a constraint, we can use different choices of (a,σ) in order to obtain agreement with experimental capture cross sections in the higher keV range. This is shown in the middle section of Fig. 8. The degrees of freedom of the neutron channel which determine the width fluctuation correction is another parameter which determines the capture cross section below ~200 keV. Figure 8 shows calculations with and without the width fluctuation correction. For all the present calculations $1.0 + T^{0.6}$ was used for the neutron width fluctuation degrees of freedom,[54] where T is the transmission coefficient.

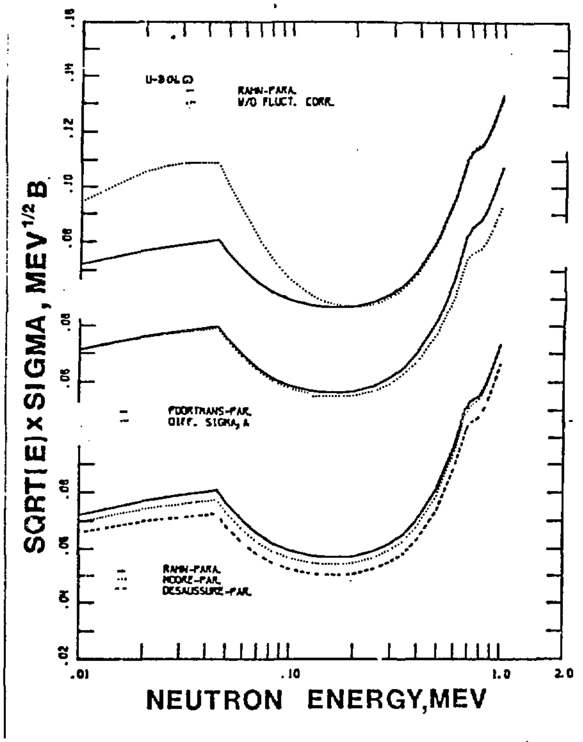

Fig. 8. Changes obtained for capture cross section calculated with the statistical and optical model for the variations of some parameters.

A spherical optical model was used in the present calculations. Some improvements might be expected with a deformed optical model, however, changes will be minor.[55] Γ_γ values for p-wave neutrons were estimated by Moore[49] to be ~5% larger than for S-wave capture, thus expected changes for the capture cross section would be <5%.

TABLE I. Γ_γ and ⟨D⟩ Values Used for Theoretical Model Calculations of the Capture Cross Sections of ^{238}U, ^{232}Th and ^{240}Pu

Nuclei	Author	Reference	$\overline{\Gamma}_\gamma$,meV	⟨D⟩,eV
^{238}U	Rahn et al.	46	22.9 ± 1.1	20.8
	DeSaussure et al.	47,48	23.1 ± 0.8	24.8
	Moore, Keyworth	49,50	21.5 ± 1.4	20.9
	Poortmans et al.	51	23.6	22.0
	ENDF/B-V	52	23.5	20.0
^{232}Th	Rahn et al.	46	21.2 ± 0.9	16.7
^{240}Pu	Lynn	53	33	14.0

B. Recent Fast Neutron Capture Cross Section Data of ^{238}U

Figure 9 shows experimental data which were measured absolutely (Ryves et al.,[56] and Panitkin et al.[57]), relative to the H(n,n) cross section (Davletchin et al.[58]), or relative to the ^{235}U (n,γ) cross section (Lindner et al.,[59] and "Present Results," which will be reported at this session. The activation technique was used for the detection of the capture events for all data shown in Fig. 9. A statistical model calculation was carried out as discussed above and the result is shown together with a ±5% range in Fig. 9. The $\overline{\Gamma}_\gamma$ and ⟨D⟩ values used for the normalization are those reported by Rahn et al.[46]. The level density parameters, a and σ, were adjusted

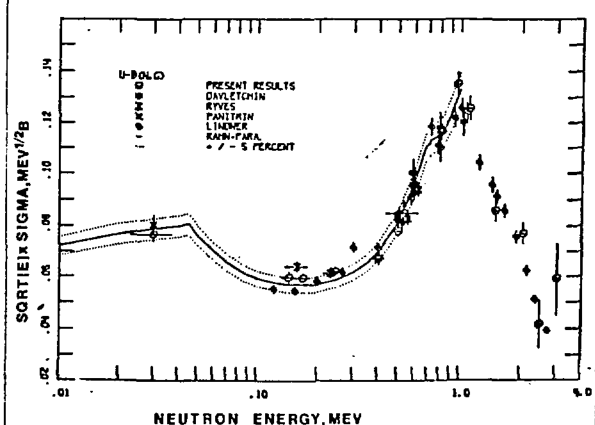

Fig. 9. Comparison of experimental data for ^{238}U (n,γ). Data in this figure were obtained with the activation technique.

in order to obtain a reasonable agreement between the calculated cross section and the experimental data in the higher keV-range. Inspection of Fig. 9 shows that the data fall into a ±5% band with the exception of two points which we can ignore. The larger spread around 150 keV should be noted. The experimental data shown in Fig. 9 suggest somewhat lower values above 500 keV and somewhat higher values around 200 keV. The latter is also supported by measurements relative to the standard capture cross section of gold (Poenitz,[60] and Spencer and Kaeppeler[37]) which are shown in Fig. 10. These data were measured with a large liquid scintillator and have subsequently larger uncertainties. The data by Poenitz are within the ±5% band except around 200 keV as discussed above. The data by Spencer and Kaeppler are generally higher and exclude with their uncertainty limit the ±5% band, though these data overlap with their uncertainties, the uncertainties of the measurements by Poenitz. Also shown in Fig. 10 are measurements by Panitkin et al.[61] relative to the ^{235}U (n,f) cross section. These data were the result of shape measurements and were renormalized to the data by Lindner et al.[59].

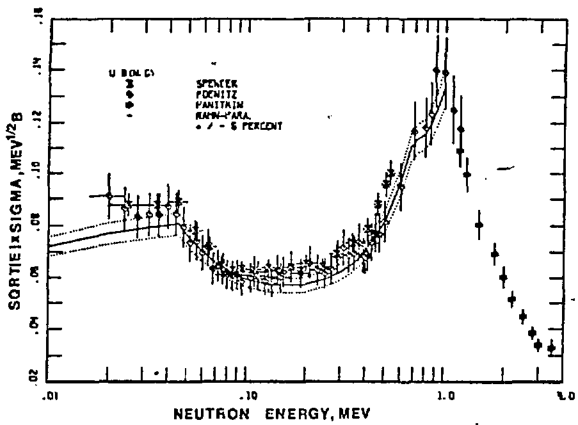

Fig. 10. Comparison of experimental data for the ^{238}U (n,γ) cross section.

Figure 11 repeats some of the data shown in Fig. 9 and shows the recent spherical shell transmission data by Dietze [25] and the results from measurements by LeRigoleur et al.[62]. The pulse-height spectrum weighting technique was used in the latter measurement. The measured values at 45 keV and at 62.5 keV are on or close to a maximum of the fluctuating cross section; thus, values for the average cross section below 100 keV are again well within the ±5% band. However, the values between 100 and 200 kev appear to suggest a decrease of the cross section which could be explained with inelastic scattering to the 4$^+$ level at 148 keV. The calculated cross section does not confirm such a sudden decrease of the capture cross section and other experimental data differ in this energy-range. It is helpful in this context to consider other cross sections measured by LeRigoleur et al.,[62] specifically the capture cross section of ^{103}Rh shown in Fig.17 below, where it is discussed as a fission product. The first three excited levels of ^{103}Rh are at 40, 93 and 297 keV with spins and parities of 7/2$^+$, 9/2$^+$ and 3/2$^-$, respectively. The spin and parity of the ground state are 1/2$^-$, thus only the 3/2$^-$ state represents appreciable competition to neutron radiative capture and elastic scattering and a capture cross section varying smoothly with energy over the 100 - 200 keV range must be expected. The data by LeRigoleur et al., however, show a structure in this range for ^{103}Rh similar to their data for ^{238}U. This suggests that this structure is caused by experimental effects which were not corrected.

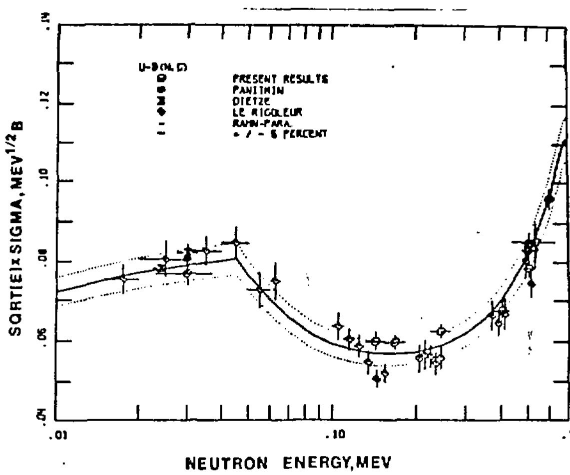

Fig. 11. Comparison of experimental data for the ^{238}U (n,γ) cross section. Data normalized in the keV-MeV range are shown.

A correction of ~1% for cross section fluctuations will decrease the value at 30 keV, and a correction of ~2% will increase the value at 24 keV measured by Dietze.[25]

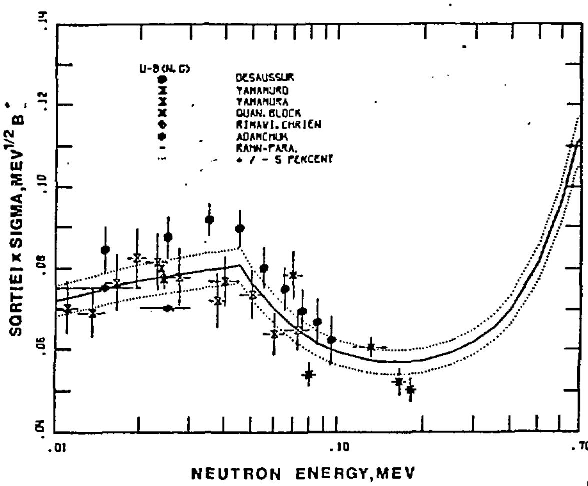

Fig. 12. Comparison of experimental data for the ^{238}U (n,γ) cross section. Data normalized at low energies (eV) are shown.

Figure 12 shows data obtained with white-spectra time-of-flight techniques at LINACs or with Fe-filter beams. These data appear to contradict the result of the model calculation and the ±5% range shown in all previous figures. The exceptions are the data by Rimawi and Chrien,[64] and by Yamamuro et al.[65]. Rimawi and Chrien used an Fe-filtered neutron beam and the activation technique. Yamamuro et al., used the pulse-height spectrum weighting technique and normalized the data to a value obtained previously[66] with an Fe-filtered neutron beam. The agreement between the data by Yamamuro et al., and the ±5% range will improve for several values if corrections for the cross section fluctuations are applied, however, not for the value at 13.5 keV. The data by DeSaussure et al.,[33] are systematically higher than the calculated cross section over the total energy range by

5-15%. The values shown in Fig. 12 are 10 keV-bin averages; therefore, fluctuation corrections do not apply. The data by DeSaussure et al., are the result of an extensive measurement program with detailed reports on the measurements and available data, thus, these data cannot be easily dismissed. Some corrections were proposed[67] but would result in changes well within the uncertainty limits of the reported data. It appears a curious coincident that the measurement, by DeSaussure et al., result in high cross section values compared with other recent data (within the last 8 years) whereas the parameters for $\langle\Gamma_\gamma\rangle$ and $\langle D\rangle$ measured or evaluated by DeSaussure result in the lowest calculated cross section (see Fig. 38).

The two values at 69.8 and 80.3 keV by Quan and Block[35] cannot be improved with corrections for cross section fluctuations, as pointed out above. The values above 100 keV are inconclusive and the implied cross section shape is not supported by the model calculation. It appears that a substantially better foreground to background ratio was obtained in this experiment at 24.3 keV than at all other energies.

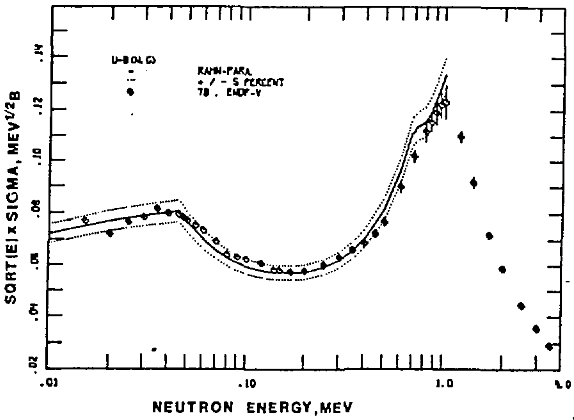

Fig. 13. Comparison of a cross section calculated in terms of the statistical and optical model and the evaluated cross section of ENDF/B-V.

The result of the statistical model calculation which was shown in the previous figures is compared in Fig. 13 with ENDF/B-V.[45,52] The evaluation of ENDF/B-V included all available data up to 1977. Several data sets considered here were not available for this evaluation and one data set used in the ENDF/B-V evaluation (Pearlstein and Moxon[68]) was not considered here because this set has remained preliminary for now more than 6 years. ENDF/B-V cross section values are also found well within the ±5% band considered above. The scattering of values at lower energies appears to come from matching different evaluations. Some differences relative to the calculated cross section are supported by the more recently measured data which were discussed above: Figures 9 and 11 indicate support for somewhat lower values above 400 keV. The optical model fit to the total and inelastic cross section data resulted in an understimation of the inelastic cross section of the 45 keV level between 400 and 800 keV. Somewhat higher cross section values below 200 keV are supported with the data by LeRigoleur et al.,[62] Ryves et al.[56] and recent measurements at ANL. The resulting cross section shape would imply a slower rising inelastic scattering cross section for the 45 keV level than presently obtained with the optical model fit.

C. <u>Recent Fast Neutron Capture Cross Section Data of ^{238}Th</u>

Uncertainty requirements for the capture cross section of ^{232}Th can be expected to be rather similar to those for ^{238}U if a ^{232}Th-^{233}U cycle is considered. The interest in the ^{232}Th - ^{233}U cycle is more recent and only a few newer measurements are available. Some of this more recent data are shown in Fig. 14. Older data are without exception higher than the more recent values, usually by 10-20%. As can be seen in Fig. 14, the values from the more recent fast neutron capture cross section measurements of ^{232}Th are found at best in a ±10% range, suggesting a factor of 2 larger uncertainty for ^{232}Th (n,γ) than for ^{238}U (n,γ). A discrepancy appears to exist between the data by Macklin and Halperin[69] on the one hand and those by Lindner et al.[59] and Poenitz and Smith[70] on the other hand. This discrepancy is in the order of 10-15%. The data by Macklin and Halperin were obtained with the pulse-height spectra weighting technique and recently revised values are shown in Fig. 14. The data by Lindner et al., and Poenitz and Smith were measured relative to the ^{235}U (n,f) cross section and the activation technique was used for the determination of the capture events. Some of the values by Poenitz and Smith were measured with a large liquid scintillator, but normalized to the activation data. Measurements by Chrien et al.[71] and Yamamuro et al.,[66] both taken with an Fe-filtered beam, appear to differ similarly from the data by Macklin and Halperin. DeSaussure and Macklin[72] pointed out that cross section values of the data by Macklin and Halperin between resonances indicate that too much background may have been subtracted from the measured events spectra. It appears that the pulse-height spectrum weighting technique measurements by Macklin lend support for lower ^{232}Th values than those measured by others, whereas they lend support for the higher ^{238}U values measured by DeSaussure et al., at least above 20 keV. The cross section calculated with the statistical model and shown in Fig. 14 was normalized with $\langle\Gamma_\gamma\rangle$ and $\langle D\rangle$ by Rahn et al.[46]. The Columbia University parameters resulted in good agreement for ^{238}U (n,γ) cross sections in the keV-range which suggested the use of ^{232}Th parameters from the same source. Figure 15 shows a comparison with the ENDF/B-V evaluated data file. Agreement is within the +/- 10% range of the experimental data. The difference in shape below the first inelastic scattering level appears to be caused by matching different evaluations in this range.

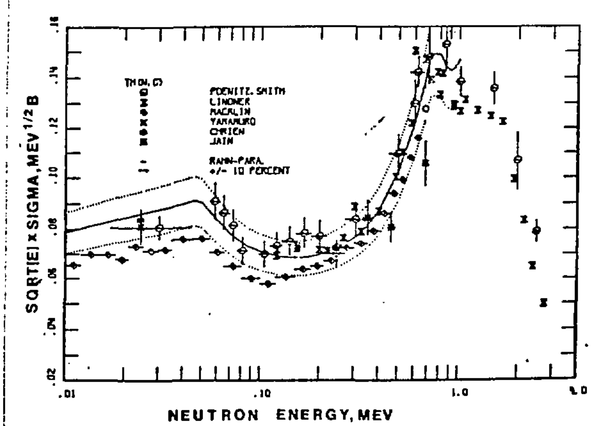

Fig. 14. Comparison of recent data for the capture cross section of ^{232}Th.

Kaeppeler considered the $\sigma_{n,\gamma}$ (^{238}U)/$\sigma_{n,\gamma}$ (Au) ratios which can be derived from their data. However, they apparently obtained different results than can be derived from the tables in which they present their ^{240}Pu (n,γ) and ^{242}Pu (n,γ) data.

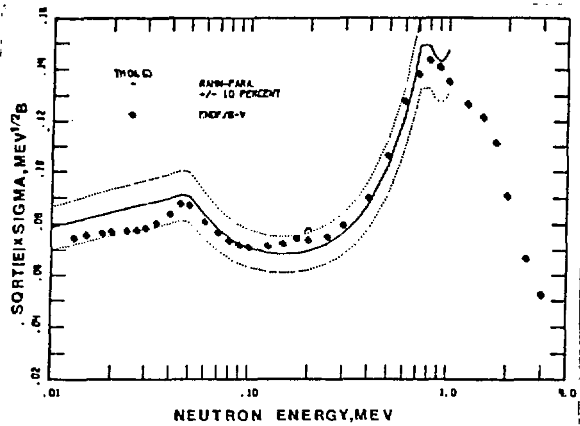

Fig. 15. Comparison of the ^{232}Th (n,γ) cross section and the calculated curve shown in Fig. 14.

D. ^{240}Pu Fast Neutron Capture Cross Section Data

The ^{240}Pu (n,γ) cross section is the major "doorway" for the buildup of higher actinides. It is of importance for burn-up calculations and uncertainty requests [73] are in the order of ~4%. Figure 15 shows the available data. It is obvious that the requested uncertainty has not yet been reached: The spread of all data appears to be over a 30% range. There is a normalization difference of ~25% between the data by Hockenbury et al.[74] and the values obtained by Weston et al.[75]. The recent data by Wisshak and Kaeppeler[76] were measured with very short flight paths of 6.8 and 13.3 cm. These data have a different shape than the data by Weston et al.[75]. The present optical model calculation supports the shape of the data by Weston et al. Wisshak and Kaeppeler point out

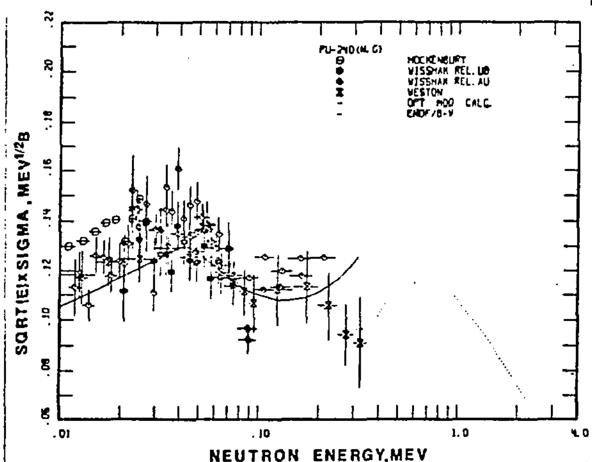

Fig. 16. Comparison of experimental data for the ^{240}Pu (n,γ) cross section and ENDF/B-V.

that their data differs from the results by Weston et al., around 30-40 keV. They measured the ^{240}Pu (n,γ) and the ^{242}Pu (n,γ) cross sections relative to the Au (n,γ) and the ^{238}U (n,γ) cross sections and reported the measured ratios. Therefore, ratios of $\sigma_{n,\gamma}$ (^{238}U)/$\sigma_{n,\gamma}$ (Au) can be derived from both, their ^{240}Pu (n,γ) and the ^{242}Pu (n,γ) measurements. These ratios differ by 20-25% around 30 keV which exceeds the estimated systematic uncertainty by about a factor of 4. A curious point appears to be that Wisshak and

E. Fission Product Capture Cross Sections

Capture cross sections of fission product nuclei are the "picture book example" for the proposition that cross sections which are difficult to measure can be calculated with nuclear models for which the parameters were determined with other, reliable experimental data. A survey of fast neutron capture cross sections of fission product nuclei which were calculated with nuclear model codes[77] shows that difference between various calculations may be as large as a factor of 4 if no experimental data exist at all. Values of

$$\delta = \frac{1}{n}\sqrt{\sum \frac{(\sigma_i - \bar{\sigma})^2}{\bar{\sigma}^2}}$$

were considered at 100 keV and 1 MeV, with σ_i the calculated cross section (i = 1...n) and $\bar{\sigma}$ the average of all evaluated cross sections. As an average over all fission product nuclei, δ was 0.31 and 0.32 if no experimental data were available and 0.15 and 0.20 at 100 keV and 1 MeV, respectively, if at least one or two experimental values were reported.

Substantial improvement can be expected if in the absence of differential data, integral values measured in facilities like CFRMF or STEK can be used in the evaluation of fission product nuclei. This approach was used extensively in the evaluation of fission product capture cross sections for ENDF/B-V by Schenter et al.[78]. Requests for fission product capture cross sections usually state requested uncertainties of ~10%.

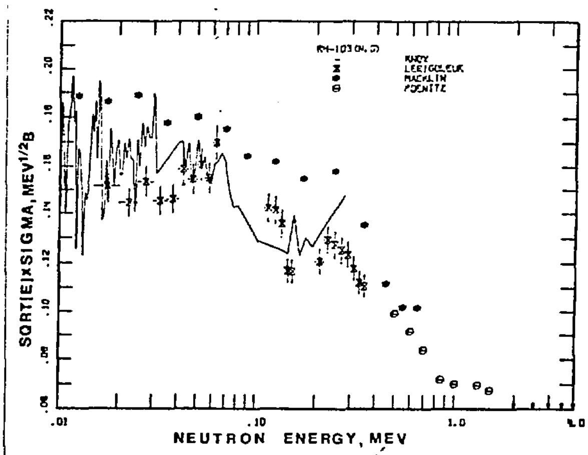

Fig. 17. Comparison of recent experimental data for the capture cross section of the fission product nuclei ^{103}Rh.

Figure 17 shows recent data for the capture cross section of ^{103}Rh. This cross section can be measured by the activation technique and prompt γ-detection techiques. All more recent values shown in Fig. 17 were obtained with the prompt γ-detection technique. Knox et al.[79] and Poenitz[80] used a large liquid scintillator and LeRigoleur et al.[62] and Macklin [81] used the pulse-height spectrum weighting technique. Large fluctuations of the cross section can be seen at low energies as should be expected. The cross section in the 100-300 keV range measured by LeRigoleur et al. shows an unexpected shape which was discussed in context of the ^{238}U (n,γ) cross section above. There appears to be a difference in normalization of ~10-20% between the recent data by Macklin and all other values shown in Fig. 17.

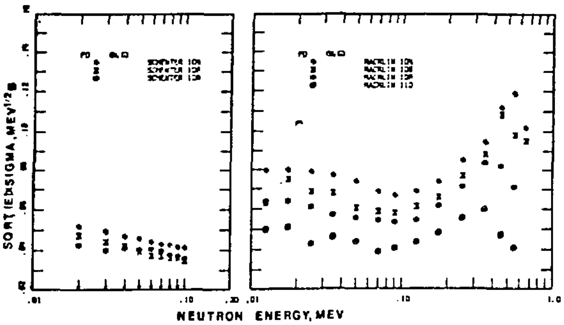

Fig. 18. Comparison of evaluated and experimental data for the even isotopes of Pd.

Figure 18 compares evaluated data by Schenter[78] and experimental values by Macklin[81] for the even isotopes of Pd. The evaluated cross sections are lower for all isotopes and the cross section for ^{110}Pd (n,γ) adjusted with integral values is lower by a factor of 5 than the experimental values for this and the other even isotopes. This cannot be understood from nuclear systematics. The evaluated cross section for the odd isotope ^{105}Pd is larger than experimental differential data (~10-15%). Because the ^{105}Pd (n,q) cross section is much larger than the capture cross sections of the even isotopes, a large part of the substantial difference for the even isotopes is compensated in the elemental cross section shown in Fig. 19. As for ^{103}Rh, recent ANL measurements support lower cross sections than measured by Macklin.

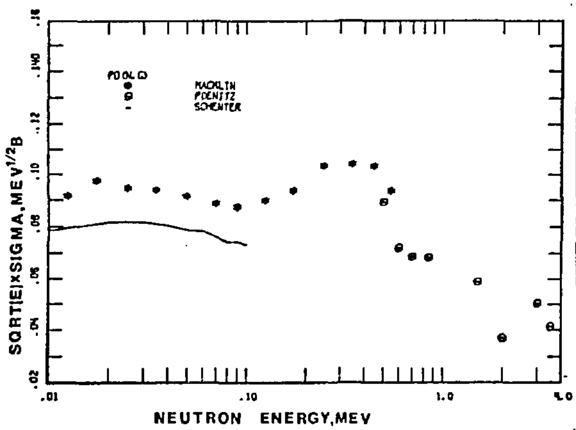

Fig. 19. Capture cross section of Pd.

IV. DISCUSSION, CONCLUSIONS AND RECOMMENDATIONS

^{238}U (n,γ)

Recent fast neutron capture cross section data measured for ^{238}U fall in a ±5% uncertainty band. This suggests that the present data uncertainty above 500 keV is less than one third of the uncertainty estimated by Weisbin et al.,[17] and falls below the required uncertainty limit above this energy (see Fig. 1). The ±5% range is in question below 500 keV where two data sets exceed this range with their estimated uncertainties. The data by DeSaussure et al.,[33] suggest 10 - 12% higher values between 10 and 45 keV than the evaluated data file ENDF/B-V. The data by Spencer and Kaeppeler[37] suggest 8 - 10% higher values between 25 and 45 keV and ~10% higher values between 100 keV and 500 keV. However, the majority of the data apparently does not follow such increases.

- *The present uncertainty of the ^{238}U (n,γ) cross section is believed to be ±5% above 10 keV and ±10% above 1 MeV. This satisfies the uncertainty limit required to achieve design accuracy for k_{eff} and the breeding ratio above 500 keV but does not fulfill the 1.5 - 3.0% uncertainty requirement below 500 keV.*

The present ±5% uncertainty limit of the ^{238}U (n,γ) cross section can not accommodate data adjustments which Weisbin et al.[17] showed are needed in order to obtain agreement between differential cross section data and integral measurement results. Considerations of C/E - ratios require the investigation of all possible sources for existing discrepancies. The C/E discrepancy for c^{28}/f^{49} could be logically caused by:

1. Errors of the differential ^{238}U (n,γ) data.
2. Errors of the experimental integral value.
3. Errors of other differential data which result in a miscalculation of c^{28}/f^{49}.
4. Errors in the computational process.
5. Errors of the differential ^{239}Pu (n,f) and/or ^{235}U (n,f) data.

The C/E - discrepancy of the central reaction rate ratios for GODIVA which was mentioned in the introduction can most likely be explained with errors of other evaluated data which cause a miscalculation of the neutron spectrum[21,82] (above point 3.). Weisbin et al.[17] showed that lowering the experimental integral value for c^{28}/f^{49} by 3% leads to substantially reduced requirements for lowering the differential data. The adjustments required in order to obtain agreement between experimental integral and evaluated differential data would be less than 5% for all but one energy interval; therefore such adjustment would fall below the present uncertainty estimate. This suggests a coordination of measurement techniques between integral and differential measurement groups in order to assure consistency. An extensive effort in this direction is presently underway at Argonne National Laboratory and substantial improvements appear to emerge. Intercomparisons of ^{238}U (n,γ) reaction rate measurements were also made by dosimetry groups for the Big Ten and CFRMF,[83] however, results were reported with uncertainties of 2.2 - 4.9% and differences were 2.0 - 4.5%. This is obviously insufficient considering the requirements shown in Fig. 1.

- *^{238}U capture rate measurements should be compared between groups involved in measuring c^{28}/f^{25} reaction rate ratios for benchmark facilities. Participation by differential data measurement groups is desirable.*

Investigations at Argonne show that different techniques yield a consistency within ± 1 - 1.5%. There is some indication that the thermal capture cross section might be too high. It appears that an uncertainty of less than 1% should be achievable.

The question to be considered next is how to improve on the present ^{238}U (n,γ) differential cross section uncertainty. Present uncertainty levels and differences between data sets measured with prompt γ-detection techniques are such that a resolution cannot be expected without expensive efforts and long, time-consuming programs. Consideration of the (in)sensitivty of the present model calculations - parameter variations leads to the following recommendation.

- *Absolute values should be measured in the energy range where the lowest uncertainties are required and nuclear model calculations with the constraints given by low energy Γ_γ and D and high-energy $\sigma_{n,\gamma}$ data should be used to provide the shape of the cross section.*

Opportunities for measuring such absolute values exist with iron-filtered beams at 24.3 keV, Sb-Be-sources at 22.8 keV and the forward neutron cone at the threshold of the ^{7}Li (p,n) ^{7}Be-reaction with keV. Presently available absolute measurements in this range are given in Table II. These values were corrected for cross section fluctuations and referenced to 30 keV. The weighted average is in very good agreement with iron-filtered beam measurements by Rimawi and Chrien[64] and Quan and Block[35] relative to the ^{10}B (n,α) cross section.

TABLE II. Absolute Values of ^{238}U (n,γ) at 30 KeV

Technique	Reference	Value, mb
^{7}Be Associated Activity	Menlove and Poenitz[84]	473 ± 14
	Panitkin et al.[57]	465 ± 23
Spherical Shell Transmission	Belanova et al.[24] and Miller and Poenitz[26]	442 ± 35
	Dietze [25]	470 ± 30
Weighted Average		468 ± 5

^{232}Th (n,γ)

- *Presently available data are inconsistent and uncertain by ±10%. Additional experimental values are required over the total energy range. ^{238}U (n,γ) should serve as an example for finding a solution to the problem.*

^{240}Pu (n,γ)

- *Present data differ in a ± 15% uncertainty range.*

^{240}Pu differs substantially from ^{238}U and ^{232}Th in regard to the available choices for the detection of the capture events. The activation technique might not be applicable and spherical shell transmission experiments might not be possible due to the lack of sufficient material or the complications caused by its high radioactivity. However, the level structure of ^{240}Pu is rather similar to ^{238}U.

- *Measurements relative to the ^{238}U (n,γ) cross section with prompt γ-detection techniques are recommended.*

ACKNOWLEDGEMENTS

The author appreciated discussions with Dr. P. A. Moldauer. This work is supported by the U. S. Department of Energy.

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