

**NUCLEAR DATA AND MEASUREMENTS SERIES**

**ANL/NDM-59**

**Review of the Neutron Capture Process in Fission Reactors**

by

W.P. Poenitz

July 1981

**ARGONNE NATIONAL LABORATORY,  
ARGONNE, ILLINOIS 60439, U.S.A.**

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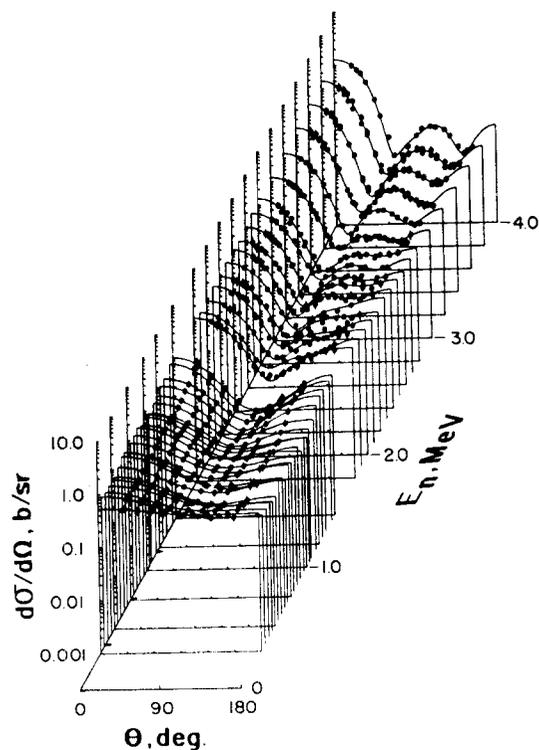
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USA

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ABSTRACT

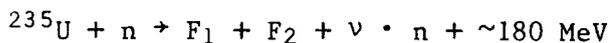
The importance of the neutron capture process and the status of the more important cross section data are reviewed. The capture in fertile and fissile nuclei is considered. For thermal reactors the thermal to epithermal capture ratio for  $^{238}\text{U}$  and  $^{232}\text{Th}$  remains a problem though some improvements were made with more recent measurements. The capture cross section of  $^{238}\text{U}$  in the fast energy range remains quite uncertain and a long standing discrepancy for the calculated versus experimental central reaction rate ratio C28/F49 persists. Capture in structural materials, fission product nuclei and the higher actinides is also considered.

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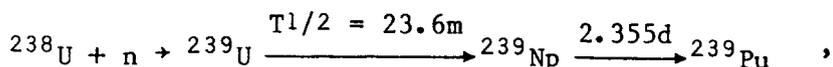
\*This work supported by the U.S. Department of Energy.

## I. INTRODUCTION

The neutron radiative capture process plays an important role in many aspects of nuclear power reactors and the associated fuel cycles. Presently operated thermal reactors burn fissile nuclei ( $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{233}\text{U}$ ) with the main objective of electrical power generation. However, the major thrust of the current reactor-research programs is oriented toward fast-breeder reactors which will not only produce electrical power but also convert, by neutron capture, abundant  $^{238}\text{U}$  to fissile fuel  $^{239}\text{Pu}$  (or  $^{232}\text{Th}$  to  $^{233}\text{U}$ ) which can in turn be used for further breeding or as fuel for thermal reactors in place of scarce  $^{235}\text{U}$ . The power production and the maintenance of the fission chain reaction in a reactor is based on the fission process, e.g. for  $^{235}\text{U}$ :



where  $F_1$  and  $F_2$  are two fission products and  $\nu$  is the average number of neutrons emitted per fission event. In a controlled nuclear chain reaction one of these neutrons must be available to initiate a new fission event. The remaining neutrons will be lost due to radiative capture in the fissile material, the fission product nuclei, the structural materials, and the coolant. They may also be lost due to absorption in the control rods or due to leakage from the reactor core and subsequent capture in the reactor shielding materials. In breeder reactors enough of the excess neutrons are captured in the fertile materials, e.g.  $^{238}\text{U}$ :



to produce more new fissile nuclei than are lost due to fission events. The above rough sketch clearly indicates the importance of the knowledge of capture cross sections for the calculation of the neutron economy in a reactor and for the evaluation of nuclear fuel cycles.

The fraction of neutrons lost due to radiative capture in fissile nuclei is usually expressed with the capture-to-fission ratio,  $\alpha = \sigma_{n,\gamma} / \sigma_{n,f}$ , which affects the number of neutrons produced per neutron absorbed,  $\eta = \nu \cdot \sigma_{n,f} / (\sigma_{n,f} + \sigma_{n,\gamma}) = \nu / (1 + \alpha)$ . For breeder reactors an important quantity is the "breeding ratio" which is the number of new fissile nuclei produced per fission event and given by

$$\text{BR} = \eta - 1 - A - L + (\nu' - 1)F$$

where  $A$  is the loss due to parasitic absorptions (mainly capture) in fission products, structural materials, coolant, and other materials,  $L$  is the leakage, and  $F$  are the fissions in fertile materials; thus a bonus ( $\nu'$  is the average number of fission neutrons in the fertile materials). The leakage term,  $L$  is dependent on the size of the reactor and usually about a factor of 3-5 smaller

than the absorption loss, A. The breeding ratio determines the economically important "doubling time", that is, the time needed to double the usable fission fuel via the "breeding gain" (BR-1).

Table I gives the fractions of capture events in various reactor materials for a typical 1000 MW sodium cooled fast breeder reactor (LMFBR) at start up time (LeSage et al.<sup>1</sup>).

Table I. Fractions of Capture Events in an LMFBR at Start-up

<sup>238</sup> U		.700
<sup>239</sup> Pu	} Fuel	.132
<sup>240</sup> Pu		.038
<sup>241</sup> Pu		.018
Structural Materials		.096
Coolant and Shield		.011
Others		.005

The reactor is assumed to be initially fueled by plutonium obtained from reprocessing of spent thermal reactor fuel elements. It is obvious that the capture in the fertile material, in this case <sup>238</sup>U, is the most important. As the burnup of the fuel proceeds fission products become an appreciable poison. Capture in the fission products decreases the reactivity of the reactor with increasing burn-up. This must be provided for with excess reactivity in the design. Neutron capture in fissile nuclei leads to a build-up of heavier nuclei, some with longer half lives which usually are  $\alpha$ -emitters. Though neutrons are lost due to capture in these transactinides, they have only a minor effect on the neutronics of a reactor because some are fissile nuclei which compensate for the neutron capture losses with gains via fission events. Although the production of higher actinides does not influence the neutronics of the reactor appreciably, they play an important aspect in other parts of the fuel-cycle, such as afterheat, waste-management and spent-fuel handling. The contributions of the different processes to the neutron economy can be seen in Table II which shows the number of neutrons contributing to the breeding gain averaged over the fuel cycle (Chang and Till<sup>2</sup>).

Table II. Breeding Ratio Components of a Metal Fueled Pu/U LMFBR

---

Neutron Gains	
η of Fissile Isotope	2.450
Fertile Fission Bonus	0.509
Neutron Losses	
Capture in Structural Materials	0.127
Capture in Fission Products	0.058
Capture in Coolant	0.008
Other Absorption Losses	0.025
Leakage Loss	0.082
<sup>241</sup> Pu Decay Loss	0.032
Net Neutrons for Breeding (Capture in <sup>238</sup> U)	1.627

---

An increase of the temperature in a reactor causes the Doppler-broadening of the resonances to increase and result in a reactivity change due to a decrease of the resonance-self-shielding effect. Capture in <sup>238</sup>U is the major source for the Doppler-effect and important for the control and safe operation of reactors.

Another aspect of the capture process is the γ-radiation against which the environment must be shielded. The appropriate calculation of the shielding requirements demands not only a knowledge of the frequency of the capture events but a knowledge of the associated γ-ray spectrum as well.

## II. CAPTURE IN FERTILE AND FISSILE NUCLEI

The dominance of neutron capture in the major fertile (<sup>238</sup>U, and <sup>232</sup>Th) and fissile (<sup>233</sup>U, <sup>235</sup>U, and <sup>239</sup>Pu) nuclei compared with capture in other materials follows from the relative abundance of these materials in a reactor and the size of their cross sections. The capture cross sections at thermal energies are substantially larger for fissile nuclei (≈100b) than for the fertile nuclei (2.7 and 7.4b for <sup>238</sup>U and <sup>232</sup>Th, respectively). Thus, a larger amount of neutrons is lost by capture in fissile nuclei in thermal than in fast reactors and α and η play a more significant role. The neutron spectra in fast reactors peak in the ~10 - ~100 keV range where the average capture cross sections of fertile and fissile nuclei are similar in size. As a consequence of the higher abundance of the fertile materials in fast reactors the neutron capture in these materials is of overwhelming importance as can be seen from Tables I and II.

The importance of the fertile and fissile nuclei with respect to neutron capture would lead us to expect that their cross sections are very well known. However, a number of problems have persisted for many years for which solutions are not yet in sight, though some improvements have been made. These problems may not be caused by a lack of knowledge of the cross section data

alone. Shortcomings of some models and approximations used to represent reactors or test facilities may contribute to these problems as well.

1. Fertile Nuclei

$^{238}\text{U}$  Capture

Measured values of the thermal neutron capture cross section of  $^{238}\text{U}$  obtained with different experimental techniques are generally in good agreement. The available data are shown in Fig. 1. The 2200 m/sec value of 2.70b from both, ENDF/B-IV and ENDF/B-V, is somewhat lower than the weighted average of the experimental data (2.711b) but agrees better with a value of 2.701b derived from the resonance parameters (Tomlinson et al.<sup>13</sup>). The resonance region presents a substantial problem. Resonance capture in thermal reactors accounts for about half of all  $^{238}\text{U}$  capture events and is strongly resonance self-shielded due to its concentration in the fuel elements. Calculations of the ratio of resonance capture vs. thermal capture with ENDF/B-IV data exceed the measured values by 3.6-5.8% (McCrosson and Hardy<sup>14</sup>). Recent experiments on the lower s-wave resonances by Olson et al.<sup>15</sup>, Lion and Chrien<sup>16</sup>, Poortmans et al.<sup>17</sup>, and Block et al.<sup>18</sup>, brought a reduction of  $\Gamma_Y$  by ~15% which was incorporated in ENDF/B-V and helped to improve the epithermal-to-thermal neutron capture ratio.

Stringent requirements are needed in the fast neutron energy range which is of interest for LMFBR's. Uncertainties of the  $^{238}\text{U}$  capture cross section as low as 1.5-3% are requested for the lower keV range and 3-10% in the higher keV and MeV range (Weisbin et al.<sup>19</sup>, Usachev and Bobkov<sup>20</sup>). The uncertainties achieved with more recent measurements of the  $^{238}\text{U}$  neutron capture cross section are at best  $\pm 5\%$  below 500 keV and  $\pm 10\%$  above (See review by Poenitz<sup>21</sup>). Major problems which exist in the high energy range are the calculated vs. experimental (CE) central reaction rate ratio discrepancy for  $^{238}\text{U}(n,\gamma)/^{239}\text{Pu}(n,\gamma)$  and the small sample central reactivity worth discrepancy for  $^{238}\text{U}$ . The central reaction rate ratio in reactor test facilities is usually calculated 3-9% higher than the experimentally determined value (LeSage and McKnight<sup>22</sup>). The CE-discrepancy for the small sample central reactivity worth is in the order of ~20% and adjustments of the capture cross section in the order of 12% would be required to resolve this problem (Bohn<sup>23</sup>).

Figure 2 show the more recent data measured for the  $^{238}\text{U}(n,\gamma)$  cross section in the fast neutron energy range. The cross section values are multiplied by the square root of the energy which allows these data to be displayed on a linear scale. Also shown in Fig. 2 is a cross section curve calculated in terms of the statistical and optical models and normalized with experimental values of  $\Gamma_Y/D$  obtained from resolved resonance parameters. Other parameters were selected to best represent the experimental data. The agreement of the experimental data with the calculated cross section curve appears to be general within a  $\pm 5\%$  range which is also shown in Fig. 2. The capture cross section of  $^{238}\text{U}$  fluctuates around the average in the unresolved resonance range. This can be seen in Fig. 3 where the capture cross section relative to its average is shown for different resolutions. These fluctuations cause some of the low-resolution data shown in Fig. 2 to scatter around the calculated curve, however, they have a negligible impact on reactor performance.

## $^{232}\text{Th}$ Capture

Although  $^{232}\text{Th}$  has been studied as a fertile material in experimental facilities and is in use in the Canadian reactor program (CANDU's), it was only more recently that interest in thorium increased substantially: Th not only presents an energy reserve which about equals that of  $^{238}\text{U}$  but also provides some options for alternative fuel cycles. Recent interest in  $^{232}\text{Th}$  has led to reviews which have shown the capture cross section data for this nucleus to be more sparse and uncertain than for  $^{238}\text{U}$ .

The thermal capture cross section appears reasonably well established. Two more recent measurements ( $7.33 \pm .17\text{b}$ , Poenitz and Smith<sup>31</sup>,  $7.41 \pm .08\text{b}$ , Chrien et al.<sup>32</sup>) agree well with the ENDF/B-IV value of  $7.40\text{b}$ .

Calculations of the epithermal vs. thermal capture ratio using ENDF/B-IV resulted in substantially lower values than experimentally observed (Ullo et al.<sup>33</sup>). New measurements of the  $^{232}\text{Th}$  resonances below 100 eV by Chrien et al.<sup>32</sup> resulted in resonance parameters ( $E_0, \Gamma_n, \Gamma_\gamma$ ) in good agreement with values recommended by Derrien<sup>34</sup>. However, the measured capture cross section was found to be substantially larger than ENDF/B-IV (up to a factor of 2) below the lowest energy resonance. Partial incorporation of these new measurements in ENDF/B-V should improve the CE-discrepancy for the epithermal-to-thermal capture ratio.

Recent measurements of the capture cross section in the unresolved and higher energy range (Macklin and Halperin<sup>35</sup>, Lindner et al.<sup>27</sup>, Poenitz and Smith<sup>31</sup>, Chrien et al.<sup>32</sup>, and Yamamuro et al.<sup>36</sup>) resulted in data about 30% lower than older measurements. After revision of the data by Macklin and Halperin<sup>35</sup> agreement between the newer data is within  $\sim \pm 5\%$ . The data are shown in Fig. 4.

## 2. Fissile Nuclei

$^{239}\text{Pu}$ ,  $^{235}\text{U}$  and  $^{233}\text{U}$

The neutron capture in fissile materials in thermal reactors appears to be sufficiently well known for neutronic calculations. Problems which exist in calculating parameters of test facilities seem to be related to other quantities, such as the fission spectra and resonance capture in the fertile materials. The thermal capture cross sections of the fissile nuclei  $^{233}\text{U}$ ,  $^{235}\text{U}$  and  $^{239}\text{Pu}$  were recently obtained in consistency fits of various experimental data ( $\sigma_{\text{tot}}$ ,  $\sigma_{\text{n,f}}$ ,  $\eta$ ,  $\sigma_{\text{n,n}}$ ,  $\sigma_{\text{abs}}$ ,  $\alpha$ ,  $\nu$ , and  $\nu$  of  $^{252}\text{Cf}$ ) which resulted in uncertainties of  $\sim 1-1\frac{1}{2}\%$  for the capture cross sections of  $^{235}\text{U}$  and  $^{239}\text{Pu}$  and  $\sim 5\%$  for  $^{233}\text{U}$  (Holden and Stehn<sup>37</sup>). The values are given in Table III and compared with ENDF/B-IV and -V values.

Table III. Thermal Neutron Capture Cross Sections of Fissile Nuclei (in barn)

	ENDF/B-IV	Consistency Fit	ENDF/B-V
$^{233}\text{U}$	46.20	$46.40 \pm 2.36$	45.76
$^{235}\text{U}$	97.22	$98.85 \pm 1.13$	98.38
$^{239}\text{Pu}$	269.71	$269.84 \pm 2.95$	270.20

Agreement is rather good between the differently evaluated values. The rather large uncertainty for  $^{233}\text{U}$  is not detrimental for neutronic calculations as the sensitivities of  $k_{\text{eff}}$  to cross section changes are low (Ullo et al.<sup>33</sup>).

The capture in fissile nuclei is also satisfactorily known for fast reactor applications. The capture cross sections are derived from capture-to-fission ratio ( $\alpha$ ) measurements for which reasonable agreement has been achieved in various experiments.  $\alpha$  of  $^{239}\text{Pu}$  stirred considerable interest when measurements by Schomberg et al.<sup>38</sup> in 1967 resulted in differences of up to a factor of 2 from the contemporary accepted evaluated data. Subsequent measurements by Schomberg and Sowerby<sup>39</sup>, Gwin et al.<sup>40</sup>, and Ryabov et al.<sup>41</sup> confirmed the deficiency of  $\alpha$  in the lower keV range though not to the extent originally suggested. Values differing by up to 15% from ENDF/B-V were recently found for the capture cross section of  $^{235}\text{U}$  between 1 and 100 keV.<sup>42</sup> Though this possible discrepancy has little effect on LMFBR-designs, which are expected to be Pu-fueled, it would affect calculations of U-fueled test-facilities or first-generation LMFBR's which might be  $^{235}\text{U}$ -fueled.

### III. CAPTURE IN STRUCTURAL, COOLANT AND SHIELDING MATERIALS

The thermal capture cross sections of structural materials are usually very small compared with cross sections for other materials involved in a reactor. This results in a reduced importance of these materials for the neutron economy in thermal reactors than is the case for fast reactors where cross section differences are less pronounced. Therefore, we will consider the capture in structural materials only in the context of fast reactors. The importance of the structural and coolant materials for fast reactor designs can be expected from the typical volume - percent distributions of ~60% for fuel (fissile and fertile), ~20% for structural materials, and ~20% for coolant. Sodium is considered in LMFBR designs as a coolant and also as a shield surrounding the core and reflector. Structural material appears in the form of fuel-cladding and supportive structures and consists mainly of stainless steel. Though the composition of stainless steel might vary widely, typical values which are used in design concepts are

- ~60-70% Fe
- ~18-20% Cr
- ~10-13% Ni
- ~2% Mo, Mn.

The capture cross sections of these elements averaged over a fast reactor spectrum differ such that the relative importance of Cr and Ni are inverted, the importance of Fe is decreased and the role for Mo and Mn is substantially increased.

Capture in structural materials is more important than capture in the fission products by a factor of 2-3 (see Table II). It influences the critical enrichment and the breeding gain. The commonly accepted goals for the design accuracy in  $k_{eff}$  and the BR of 0.5-1.0% and 2%, respectively, leads to requests for capture cross section uncertainties of 5-10% for Fe, Cr, and Ni and 10% for Mo and Mn.

The isotopic compositions of the elemental Fe and Cr are dominated by one major isotope each ( $^{56}\text{Fe}$ , 91.7%, and  $^{52}\text{Cr}$ , 83.8%) whereas for Ni two isotopes contribute larger amounts ( $^{58}\text{Ni}$ , 67.8%, and  $^{60}\text{Ni}$ , 26.2%). All of these isotopes are in a mass range where the cross sections in the main region of the fast reactor spectrum show distinct resonances. S-wave resonances in this range are large and broad and therefore strongly self-shielded, whereas p-wave resonances are narrow and small, and less self-shielded. Figure 5 shows as an example the capture cross sections of  $^{56}\text{Fe}$ ,  $^{58}\text{Ni}$ ,  $^{60}\text{Ni}$  and  $^{61}\text{Ni}$ . The dominance of one or two isotopes in the isotopic composition of Fe, Cr, or Ni does not imply that the minor isotopes play a correspondingly unimportant role. The level spacings vary greatly among the various isotopes (up to a factor of 10) and the differences of the resonance parameters increases the importance of some and decreases it for others. An example is  $^{53}\text{Cr}$  which occurs with only 9.5% in the isotopic composition of chromium but has an average capture section which is about a factor of 100 larger than that of the primary isotope  $^{52}\text{Cr}$ , and thus dominates the capture in elemental chromium.

The resonance nature of the cross sections of the structural materials results in their significant importance for the Doppler-effect in fast reactors. They contribute about as much as the fissile nuclei, but less than the fertile nuclei. The narrow p-wave resonances are strongly Doppler-broadened whereas the Doppler-broadening contributes little to the wide S-wave resonances. The major part (~50-80%) of the Doppler-effect caused by structural materials in fast reactors is due to the 1.5 eV p-wave resonance of  $^{56}\text{Fe}$  and the uncertainties of its resonance parameters contribute about 90% to the uncertainty in calculating the Doppler-effect of structural materials (Takano and Ishiguro<sup>43</sup>).

Measurements of the capture cross sections of structural materials encounter two major difficulties. The first problem is the spectrum of the  $\gamma$ -rays emitted in the capture process which is dominated by strong transitions to the ground state or low-lying excited states. This results in an appreciable spectral sensitivity of total-energy detectors (large-liquid scintillators) which is visible in changes of the measured pulse-height distribution (see Fig. 13, ANL/NDM-58). Some effect can also be expected from energy-proportional detectors because the transitions from the low-lying states fall into a range where the detector efficiency is not proportional to the  $\gamma$ -ray energy.

The second problem results from the much larger scattering than capture cross sections with  $\Gamma_n/\Gamma_\gamma$  typically in a range of  $10^3$  to  $10^4$ . This results in a large fraction of the interacting neutrons being scattered and subsequently causing secondary neutron capture events within the samples.

Required corrections for this effect are in the order of a factor of 2. The scattered neutrons cause an additional problem due to the neutron sensitivity of the capture  $\gamma$ -ray detectors. Several observations by Moxon et al.<sup>44</sup> may be explained in light of this problem: a)  $\Gamma_\gamma$  values for p-wave resonances (which have much smaller  $\Gamma_n/\Gamma_\gamma$  values) were reported to be smaller by factors of 2-3 than for s-wave resonances, b) capture cross section measurements with lead slowing-down spectrometers which have a very low neutron sensitivity usually yield lower values than those obtained with other techniques, and c) activation measurements often result in considerably lower cross section values than obtained with prompt detection techniques. The latter could also be due to the smaller samples used in activation measurements which reduces the capture probability for secondary neutrons.

Very few capture cross section measurements on structural materials were carried out prior to 1965. Some average cross section data were obtained with lead slowing-down spectrometers (Kapchigashev et al.<sup>45</sup>) at low energies and with large liquid scintillator detectors (Diven et al.<sup>46</sup>) at higher energies. The analysis of resonance parameters requires in addition to capture yield data, the knowledge of the total cross section. As a result of this data need, more recent experimental efforts were oriented toward the simultaneous provision of transmission and capture data. Such comprehensive total and capture cross section data sets were obtained for  $^{50,52,53,54}\text{Cr}$  and  $^{60}\text{Ni}$  at Rensselaer Polytechnic Institute (Stieglitz et al.<sup>47</sup>) and for  $^{50,52,53}\text{Cr}$ ,  $^{54,57,58}\text{Fe}$ , and  $^{61,62,64}\text{Ni}$  at Karlsruhe (Beer and Spencer,<sup>48</sup>). Many p-wave resonances were found in the capture measurements but were missing in the transmission measurements due to a lack of resolution. The capture measurements yield only values of  $g\Gamma_n\Gamma_\gamma/\Gamma$  and might be incorrect if the neutron capture data cannot be corrected for multiple scattering effects because of missing total cross section information. Thus, the analysis of these data was in part incomplete.

Measurements with substantially improved resolution were carried out at Oak Ridge National Laboratory for the same and some additional isotopes (Allen et al.<sup>49</sup>). Such measurements were also made at the EURATOM laboratories at Geel for the isotopes  $^{54,56,57}\text{Fe}$ , and  $^{50,52,53}\text{Cr}$  (Brusegan et al.<sup>50</sup>) and at Harwell for elemental iron, nickel and chromium (Gayther et al.<sup>51</sup>) and various isotopes (James and Syme<sup>52</sup>).

Some of the problems in determining resonance parameters can be seen in Table IV which lists the experimental results for  $\Gamma_\gamma$  of two important resonances of  $^{56}\text{Fe}$ . The 1.15 keV resonance is a p-wave resonance and agreement for  $\Gamma_\gamma$  is rather good. However, discrepancies are found even between newer measurements for the s-wave resonance at 27.7 keV.

Table IV. Experimental  $\Gamma_\gamma$  - Values for two Resonances of  $^{56}\text{Fe}$ .

$E_0/\text{KeV}$	Year	$\Gamma_\gamma$	Reference <sup>53</sup>
1.15	1964	0.60 $\pm$ 0.06	Block
	1965	0.56	Moxon
	1969	0.57 $\pm$ 0.06	Julien
	1977	0.785 $\pm$ 0.100	Poortmans
	1977	0.60 $\pm$ 0.06	Perey <sup>a</sup>
	1979	0.615 $\pm$ 0.026	Gayther
	1979	0.610 $\pm$ 0.060	Brusegan
	27.7	1968	1.44 $\pm$ 0.14
1970		1.40 $\pm$ 0.02	Ernst
1977		1.25 $\pm$ 0.2	Froehner <sup>b</sup>
1977		1.60 $\pm$ 0.2	Allen
1977		1.4 $\pm$ 0.1	Perey <sup>a</sup>
1979		0.75 $\pm$ 0.20	Moxon
1979		0.99 $\pm$ 0.04	Wisshae
1979		0.85 $\pm$ 0.05	Gayther
1979		0.80 $\pm$ 0.20	Brusegan

<sup>a</sup>Analysis of various ORNL measurements.

<sup>b</sup>Reanalysis of the measurements by Ernst.

Although substantial improvements have been made for the capture cross sections of structural materials, uncertainties still remain exceeding the requested uncertainty levels of 5-10%. Changes of the one-group cross section for several fast criticals<sup>54</sup> (ZPR-6-6A, ZPR-6-7, and ZPR-9-31) between ENDF/B-IV and -V are indicative of the unsettled nature of these data (see Table V).

Table V. Percent Changes of One-Group Cross Sections Between ENDF/B-V and /B-IV (Pennington<sup>54</sup>)

Material	$\frac{\bar{\sigma}_{n,\gamma} \text{ V} / \bar{\sigma}_{n,\gamma} \text{ IV}}$
Fe	+6.2%
Ni	-3.1%
Cr	+8.9%
Mn	-19%
Mo	-6.8%
Na	-36%

The capture cross section of the coolant and shield Na is rather small and thus less important for the neutronics of a reactor (see Table II). Cross section changes were substantial between Version V and IV of ENDF/B (see Table V), mainly due to changes of the resonance parameters with  $\Gamma_\gamma$  values differing by factors of 2-4. The uncertainty of  $\Gamma_\gamma$  of the 2.85 keV resonance contributes to the capture component of the sodium-void effect in LMFBR's, however, probably no more than ~0.1% which is not considered very significant.

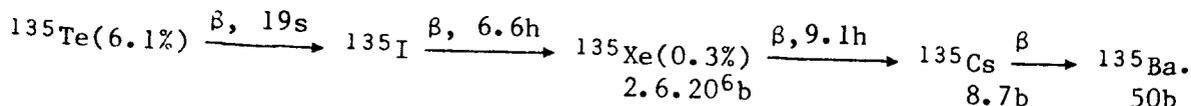
#### IV. CAPTURE IN FISSION PRODUCT NUCLEI

The neutron fission process results in fission product nuclei with a mass distribution showing a well known double mass peak. The lighter masses are in a range of  $A \sim 80-100$  and the heavier masses are in a range of  $A \sim 125-160$ . Many of the fission product nuclei decay with a short half-life to stable nuclei or nuclei with longer half-lives which accumulate in the fuel rods. The relative importance of these fission products as neutron capture poisons is given by the frequency of their occurrence, their half-lives, and the size of their capture cross sections averaged over the reactor spectrum. The absorption of neutrons in the fission products causes a negative reactivity change which must be anticipated with a sufficient reactivity excess in the design of a reactor.

##### 1. Thermal Reactors

The thermal cross sections and the resonance integral determine the neutron capture in fission product nuclei in thermal reactors. These quantities can vary by several orders of magnitude due to the statistical nature of resonance energies and neutron widths. This results in a distinct selectivity for the importance of some fission products as poisons in thermal reactors which is in contrast to fast reactors where the absorption losses are governed by the more systematic nature of the average cross sections at higher energies. The dominance of neutron capture in a few fission products in thermal reactors introduces dynamical effects which influence the operation of the reactor within the time span of the involved half-lives.

Of primary importance is the fission product  $^{135}\text{Xe}$  which accounts for more than 50% of the fission product neutron absorption of a freshly started thermal reactor. It has a resonance at 0.084 eV, a  $\Gamma_\gamma$  of 90.7 meV and a reduced neutron width of  $2g\Gamma_n^0 = 88.7$  meV, resulting in a thermal capture cross section of  $\sim 2.65 \cdot 10^6$  b (see Fig. 6). Although  $^{135}\text{Xe}$  constitutes only 0.3% of the fission products in the  $^{235}\text{U}$  fission process, it is also enhanced from the decay of  $^{135}\text{Te}$  which constitutes 6.1%:



The reactivity change is therefore determined mainly by three factors:

- a) the decay of  $^{135}\text{I}$  with a half-life of 6.6h which causes the  $^{135}\text{Xe}$  build-up,
- b) the decay of  $^{135}\text{Xe}$  with a half-life of 9.1h which removes some of the poison, and
- c) the burn-up of  $^{135}\text{Xe}$  during reactor operation which also removes some of this poison.

The first and the second factors are of importance for reactor shut-downs.  $^{135}\text{Xe}$  builds-up due to  $^{135}\text{I}$  decay above the equilibrium value which it reaches due to the burn-up during reactor operation. Excess reactivity is required for restarting the reactor in order to overcome the  $^{135}\text{Xe}$  poison, or sufficient time must elapse in order to permit the decay of  $^{135}\text{I}$  and subsequently  $^{135}\text{Xe}$ . The build-up of  $^{135}\text{Xe}$  after reactor shut-down will be proportionally larger for higher neutron flux reactors. The excess reactivity required in a high-flux reactor because of the  $^{135}\text{Xe}$  poison is in the order of several percent.

The neutron flux in the reactor core is much higher than at the reactor edge and  $^{135}\text{Xe}$  decay and burn-up play a different role at different positions in the reactor. Spatial and time oscillations of the reactor power are the result of the neutron capture in  $^{135}\text{Xe}$ : A change of the neutron flux at one point will alter the  $^{135}\text{Xe}$  absorption which in turn will change the reactivity and amplify the original flux change. This will continue until the poison production catches up and reverses the trend.

Another fission product with a very large thermal cross section ( $\sim 4.1 \cdot 10^4 \text{ b}$ ) is  $^{149}\text{Sm}$  which is stable and occurs in the fission process with 1.1% probability.  $^{149}\text{Sm}$  is also produced by the decay of  $^{149}\text{Pm}$  with a half-life of 53.1h. This long half-life requires the reactor to be built with sufficient excess reactivity in order to compensate for the  $^{149}\text{Sm}$  build-up after shut-down as waiting for its decay would be impractical. However, the capture cross section is much smaller than for  $^{135}\text{Xe}$  (see Fig. 6) and therefore the required excess reactivity is about one order of magnitude less.

As a reactor is operated over a longer time span other fission products accumulate and the relative importance of neutron capture in  $^{135}\text{Xe}$  and  $^{149}\text{Sm}$  decreases. Table VI compares the total neutron captures in some of the major fission products after 4000h and after 26400h of reactor operation (Schenter and England<sup>55</sup>).

Table VI. Accumulated Neutron Capture Events in Individual Fission Products Relative to Total Captures in Fission Products after 4000h and 26400h of Reactor Operation

<u>Nuclide</u>	<u>4000h</u>	<u>26400h</u>
$^{135}\text{Xe}$	51.7	15.1
$^{149}\text{Sm}$	13.6	5.7
$^{151}\text{Sm}$	4.9	3.2
$^{143}\text{Nd}$	4.7	9.9
$^{147}\text{Pm}$	3.8	4.6
$^{103}\text{Rh}$	2.9	9.8
$^{131}\text{Xe}$	2.6	6.0
$^{133}\text{Cs}$	2.4	6.2
$^{99}\text{Tc}$	1.9	4.9
$^{152}\text{Sm}$	1.6	3.9

## 2. Fast Reactors

The neutron capture cross sections of the fission product nuclei averaged over a fast-reactor spectrum are much smaller than some of their thermal cross sections and show some systematic behavior. They typically differ from each other by less than a factor of 10. As a result, the relative importance of neutron capture is more evenly distributed over a larger number of fission products. Table VII lists the contributions of the 25 most important fission products to the total neutron absorption by fission products in a typical fast reactor. The more even distribution of capture in various fission products makes the problems of transients and restart for fast reactors negligible, and the main effect is the long term reactivity change caused by stable fission products or those with a long half-life.

The capture cross sections of the more important nuclei must be known with an uncertainty of ~10% in order to calculate the bulk reactivity effect of the fission products to a required accuracy of 5-10% (Rowlands<sup>56</sup>). More stringent requirements (~5-7% uncertainties of cross sections) were stated recently (Hammer<sup>57</sup>). Some of the fission products (e.g. Nd) are used as burn-up monitors for the fuel and their cross sections are required to be known more accurately.

Measurements of capture cross sections of fission products in the fast energy range encounter various difficulties. Suitable samples are not always available, the radioactivity of even longer-living nuclei restricts the applicability of most measurement techniques. As a result experimental data for some nuclei are sparse, discrepant for others, or not available at all. Figure 7 shows experimental data for the capture cross section of

Table VII. The Twenty-five Most Important Fission Product Absorbers in Fast Reactor Cores (Schenter and England<sup>55</sup>)

Nuclide	Contribution to Total Absorption %	Nuclide	Contribution to Total Absorption %
Pd 105	9.9	Ru 106	2.3
Tc 99	8.6	Nd 143	2.3
Ru 101	7.7	Xe 131	1.9
Pd 107	6.2	Sm 151	1.9
Rh 103	5.5	Mo 95	1.5
Cs 133	4.9	Ru 104	1.3
Pm 147	3.5	Eu 153	1.3
Sm 149	3.4	Mo 98	1.2
Nd 145	3.4	Ce 144	1.1
Ru 102	3.3	I 129	1.0
Cs 135	3.0	Mo 100	0.9
Mo 97	2.9	Pr 141	0.9
Ag 109	2.7	Total	82.6

<sup>109</sup>Ag which scatter by a factor of ~2. The agreement between experimental data for other nuclei may be better, with spreads of ±25% and even ±15% for more recent data. An example for this is given with Fig. 8 which shows the capture cross section of <sup>103</sup>Rh. The complete lack of data for some nuclei and the existing discrepancies for others have encouraged the use of theoretical calculations of these cross sections in terms of the statistical model and the optical model (for example: Benzi and Reffo<sup>58</sup>, Schmittroth and Schenter<sup>59</sup>). Such calculated cross sections utilize experimental data for the  $\gamma$ -width,  $\Gamma_\gamma$ , the average level spacing,  $D$ , and the neutron strength function where available, or, again, systematics and calculated values. The number of parameters on which these calculations depend is substantial and good predictions can only be expected if experimental information is available to guide the normalization. This is specifically the case at higher energies (> 1 MeV) where large differences must be expected between various model calculations which are based upon different parameter choices and approximations. Table VIII compares the calculated neutron capture cross sections at 2 MeV for several fission product nuclei. Differences between the theoretically calculated cross sections exceed a factor of two for most nuclei and even factors of 5 and 10 occur.

The nonexistence or deficiencies of differential data and the uncertainties of theoretical calculations have led to yet another approach. Cross sections averaged over a fast reactor neutron spectrum can be measured with small samples because of the available high neutron flux. Such average cross sections can then be used to normalize theoretically calculated cross section shapes. Additional information about the cross section shape can be obtained if measurements in several reactors or test facilities were made which have substantially different neutron spectra.

The ENDF/B-V evaluation is based upon all three sources of information. Differential and integral (averaged) experimental data were used together with calculated cross sections in a minimization procedure (Schenter and England<sup>55</sup>).

The large number of fission product nuclei contributing to the poisoning of fast reactors complicates design calculations. Therefore, a lump fission product cross section is used in some calculations as an approximation. This cross section is derived by summing the individual cross sections according to the frequency of the occurrence of specific nuclei. A somewhat more sophisticated approach is to take into account cross section systematics. Odd-even and even-odd nuclei have much larger cross sections than even-even nuclei. Capture in one of the former may lead to the latter and thus reduce the poisoning. Thus two lump fission product cross sections are being used in some calculations.

Table VIII. Comparison Between Different Calculated Fission Product Capture Cross Sections (in mbarn) at 2 MeV (Iijima et al.<sup>60</sup>).

<u>Nuclide</u>	<u>Cook<sup>61</sup></u>	<u>Benzi<sup>58</sup></u>	<u>Lauterbach<sup>62</sup></u>	<u>Iijima<sup>60</sup></u>
Zr-93	1.43	---	11.7	55.6
Mo-95	12.7	13	22.1	32.7
Mo-97	5.47	9.37	27.1	24.0
Tc-99	18.2	---	66.9	82.2
Ru-101	96.1	26.24	80.3	67.1
Ru-101	58.3	90.82	26.1	73.5
Rh-103	40.2	44.2	63.6	63.3
Ru-104	15.2	25.0	26.8	37.5
Pd-105	17.8	35.6	94.3	80.8
Ru-106	8.11	---	24.3	27.0
Pd-107	43.6	---	91.4	78.1
Ag-109	65.9	96.1	117.2	112.2
I-129	21.3	---	39.6	56.7
Xe-131	17.2	16.1	31.5	28.7
Cs-133	10.7	19.2	45.4	30.7
Nd-143	10.8	13.1	17.6	120.5
Nd-144	30.8	28.3	65.8	34.2
Sm-147	24.0	36.9	71.1	82.7
Sm-149	31.1	47.7	135.0	238.1
Sm-151	137.1	50.7	194.2	249.1

## V. THE PRODUCTION OF HIGHER ACTINIDES

Neutron capture in heavy fissile and fertile elements plays a predominant role in the build-up of transactinides in reactors. Other factors are radioactive decay,  $(n,2n)$ , and fission reactions. A typical build-up chain is shown in Fig. 9. Nuclear data and specifically neutron capture cross sections are required to evaluate the various effects of these higher transactinides for different aspects of the nuclear technology which become increasingly important. The overwhelming concern is the disposal of the transactinides as waste, however, other aspects as for example the influence of the actinides on the reactor reactivity and breeding gain are of interest. A positive element of the creation of the higher actinides is the usefulness of several nuclei for a variety of applications, e.g. in medical, industrial and agricultural areas.

Problems relating to the higher actinides can be divided into two major groups:

1. The changes of the neutronics of the reactor core which result from the changes of the isotopic composition of the fertile and fissile materials, and affect the reactivity, reactor power, and breeding gain.
2. Problems created by the higher actinides for other parts of the nuclear fuel cycle. These include fuel handling, transport and reprocessing, waste management, subcritical reactivity during refueling, and fuel fabrication from recycled fissile/fertile materials.

A distinct difference exists between the higher plutonium isotopes  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ , and  $^{242}\text{Pu}$  which contribute a reactivity loss of  $\sim 2\%$  and the other higher transactinides. The former are the inevitable result of LWR reprocessed fuel. Concentrations of  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$  exceed other higher transactinides by at least one or two orders of magnitude and their nuclear cross section data are usually matching the primary fuel nuclei in importance. The concentrations of  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$  in recycled fuel from LWR's and LMFBR's are somewhat different (see Table IX), and thus, so is the importance of their nuclear data. For both types of reactors the higher plutonium, americium, curium, and californium isotopes have an insignificant overall impact on the neutronics, although effects for individual isotopes are none - zero. The effects on the reactivity, internal breeding gain, and total power of a fast reactor due to the higher transactinides are given in Table X. For comparison the more frequent Pu - isotopes are also listed.

Table IX. Comparison of the Yearly Production of Transactinides in LWR's and LMFBR's (Kuesters and Lalovic<sup>63</sup>).

Isotope	kg/year	
	LWR (U - fueled)	LMFBR
Pu 236	0.02	0.002
Pu 238	4.5	15
Pu 239	145	1318
Pu 240	59	441
Pu 241	27	120
Pu 242	9	75
Am 241	2	11
Am 242M	0.01	0.2
Am 243	2.5	5.6
Cm 242	0.09	0.3
Cm 243	0.02	0.02
Cm 244	0.83	0.3

Table X. Effect of the Transactinides on Fast Breeder Parameters (Barre and Bouchard<sup>64</sup>).

Nuclide	Internal Breeding Gain	Total Power %	Total Absorption %
U-232	0	0	0
Np-237	0.005	0	0.1
Np-239	0	0	0.1
Pu-236	0	0	0
Pu-238	-0.003	0.5	0.3
Am-241	0.023	0.2	0.6
Am-242M	-0.007	0.3	0.1
Am-243	0.004	0.1	0.5
Cm-242	-0.001	0.1	0
Cm-243	0	0	0
Cm-244	0	0.1	0
Total	+0.021	1.3	1.7
Pu-240	0.115	6.3	6.4
Pu-241	-0.143	7.8	4.0
Pu-242	-0.001	0.7	1.1

Measurements of capture cross sections of the higher actinides are troubled by two major problems. The first is the scarcity of suitable samples. Substantially larger amounts are needed for capture measurements than for fission or total cross section experiments. Exceptions are the thermal cross sections which are usually large and thus require smaller sample masses, and integral values which can be carried out at higher neutron flux levels than available for differential cross section experiments. The second problem is the limited applicability of available measurement techniques. High spontaneous fission decay and  $\alpha$ -decay rates, or fission competition make measurements very difficult.

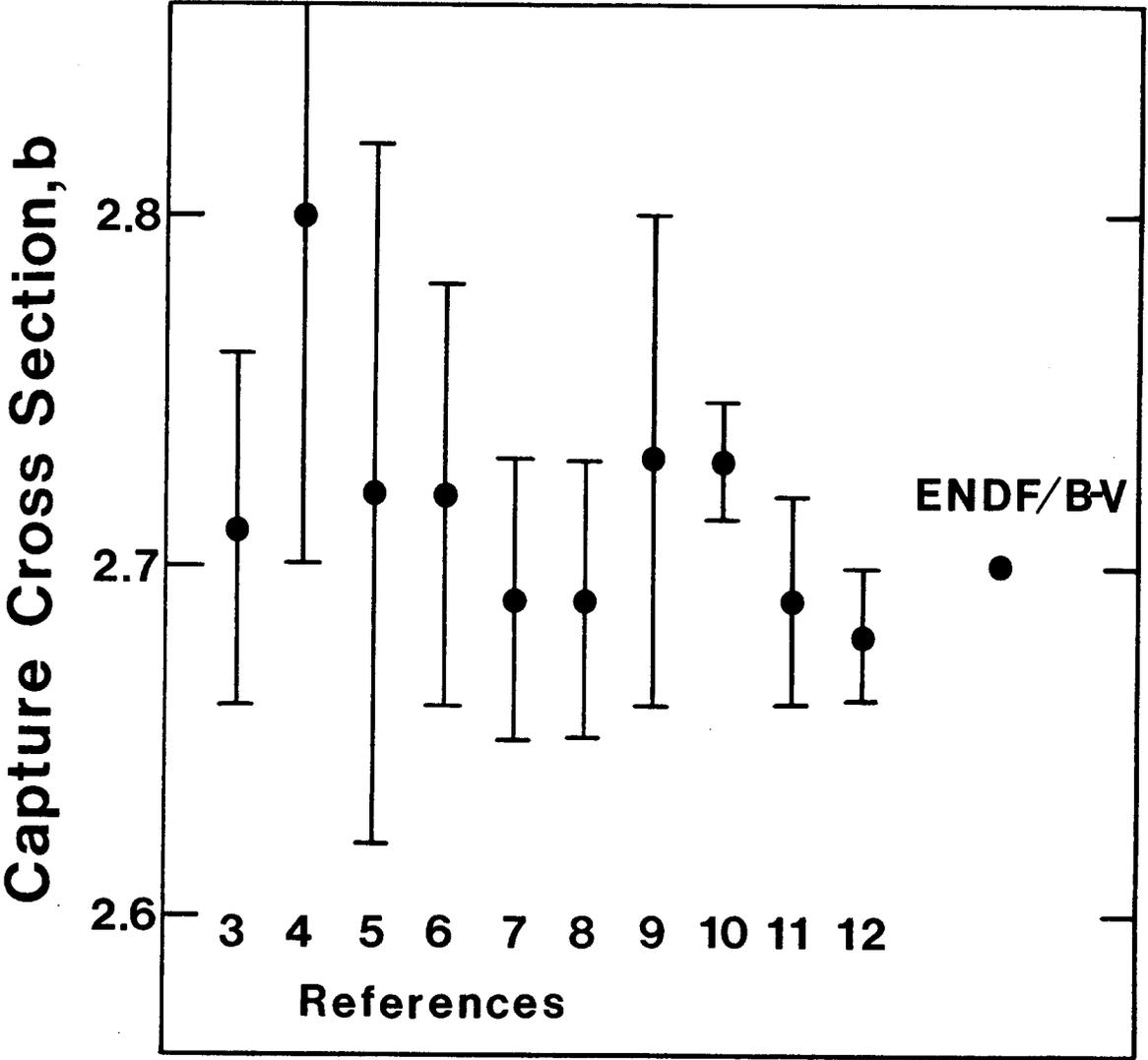
The thermal capture cross sections and the infinite dilute resonance integrals are usually known at about the requested uncertainty levels. A surprising exception is the resonance integral of  $^{240}\text{Pu}$  which is sought to be known to 3% but is uncertain by  $\sim 17\%$  (Benjamin<sup>65</sup>). The low energy cross section of  $^{240}\text{Pu}$  is dominated by a  $10^5\text{b}$  resonance at 1 eV which contributes  $\sim 99\%$  of the thermal cross section. This resonance was excluded in more recent measurements (because it would require special samples) and its resonance parameters ( $\Gamma_\gamma, \Gamma_n$ ) are known from older measurements with  $\sim 10\%$  uncertainty (Weston<sup>66</sup>). The well known thermal cross section may be used as a constraint for a resonance parameters fit and reduce the uncertainty in the resonance range (Thompson and Leonard<sup>67</sup>). Data in the unresolved resonance and higher keV energy range, important for fast reactors, agree within typically  $\sim \pm 15\%$ . The measurements by Weston and Todd<sup>68</sup> are in best agreement with the average resonance parameters.

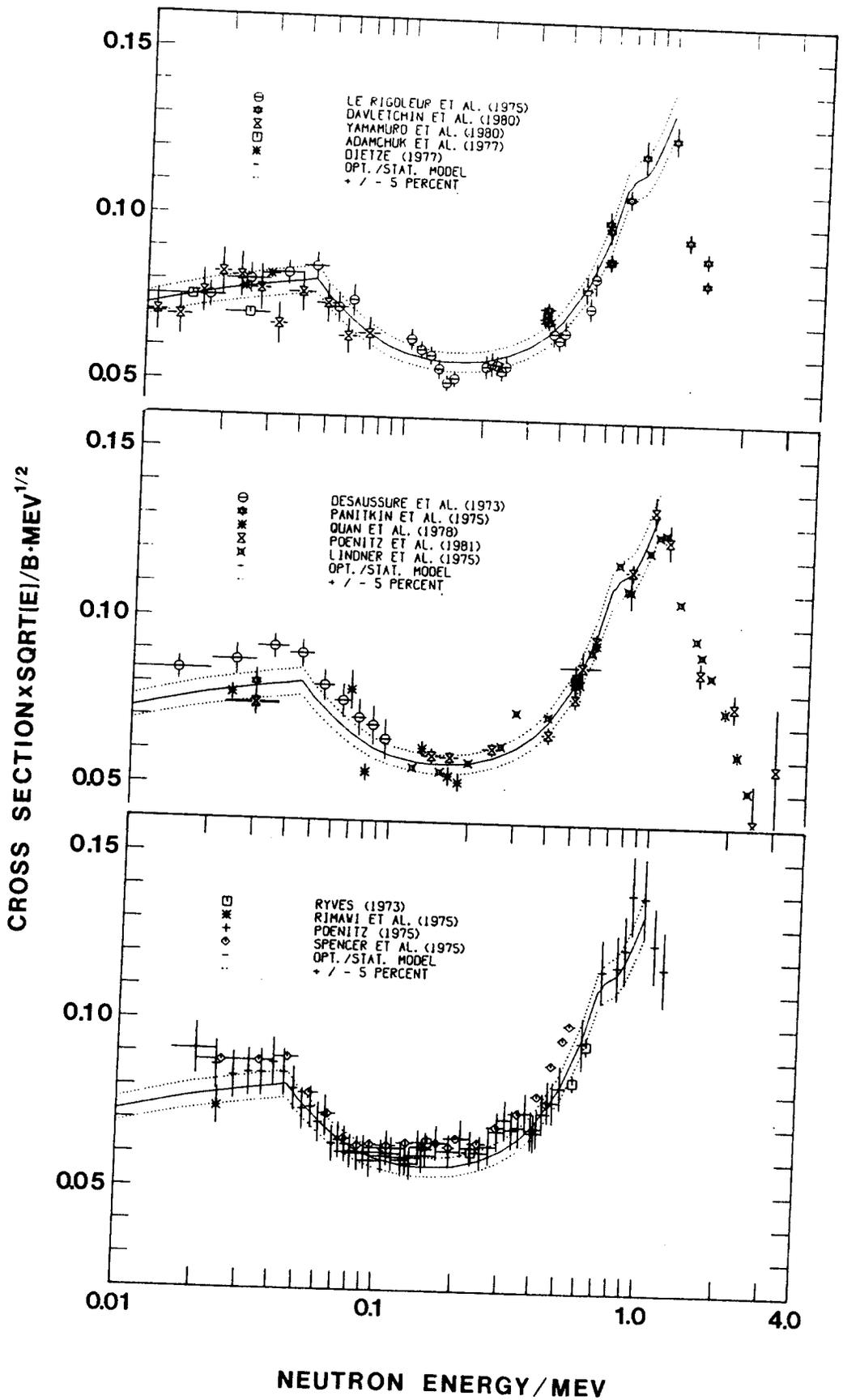
Capture in  $^{240}\text{Pu}$  leads to  $^{241}\text{Pu}$  which is a good reactor fuel and its fission cross section and decay to  $^{241}\text{Am}$  are more important for reactor neutronics than neutron capture. However, neutron capture in  $^{241}\text{Pu}$  and in  $^{241}\text{Am}$  lead to nuclei which create fuel handling ( $^{242}\text{Cm}$ ,  $^{244}\text{Cm}$  and  $^{252}\text{Cf}$ , which are neutron emitters) and waste management problems.

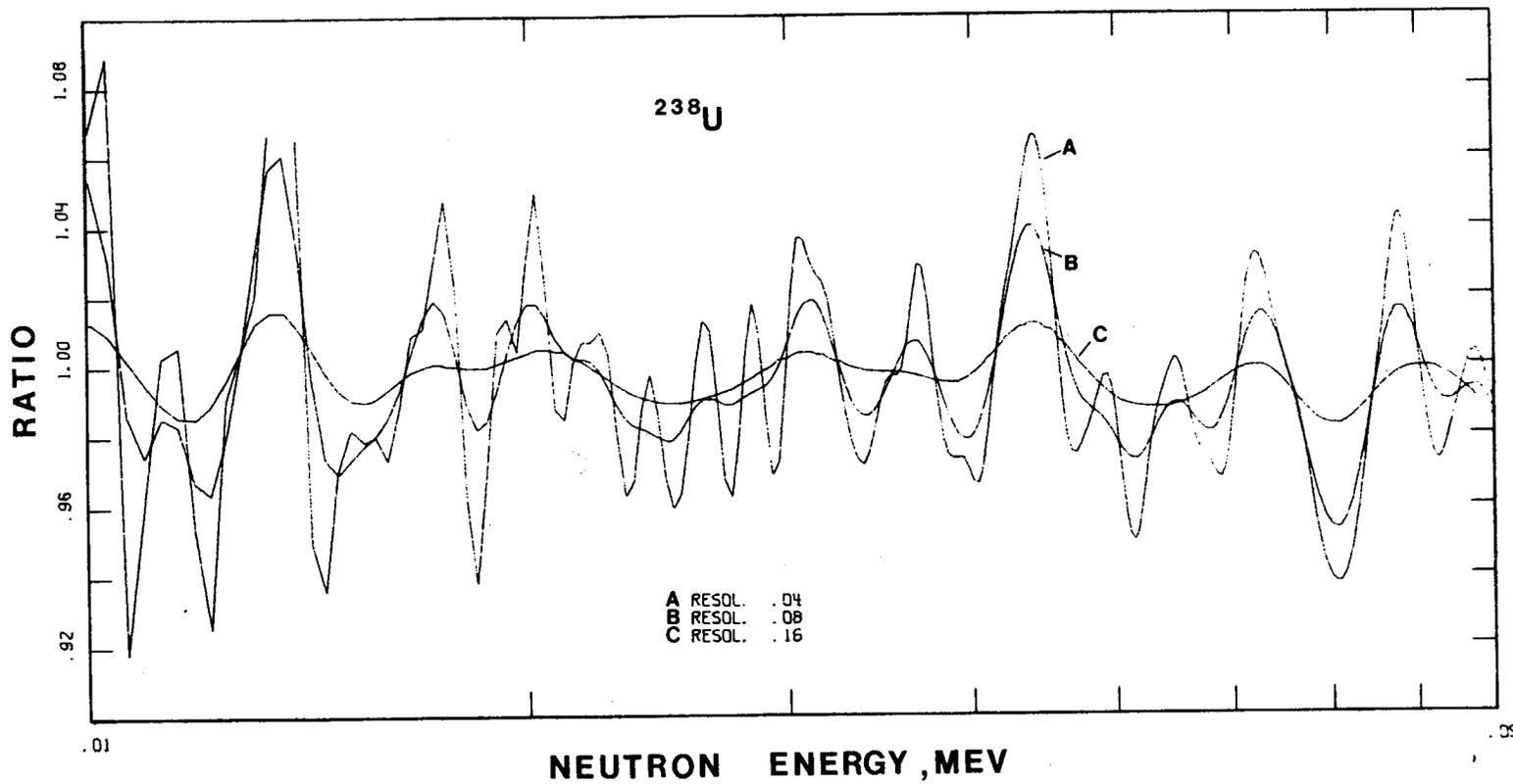
With the exception of  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$  and  $^{241}\text{Am}$ , few differential data have been measured except at low energies in the resolved resonance energy range. Therefore, evaluated data must rely heavily on theoretical calculations to provide capture cross sections in the higher keV energy range. Experimental resonance parameters and integral values are used for normalization; and nuclei for which experimental values exist provide test cases for such calculations. Figure 10 shows, for example a theoretically calculated capture cross section for  $^{240}\text{Pu}$ . Agreement with experimental data is within the  $\pm 15\%$  data scatter.

## Figure Captions

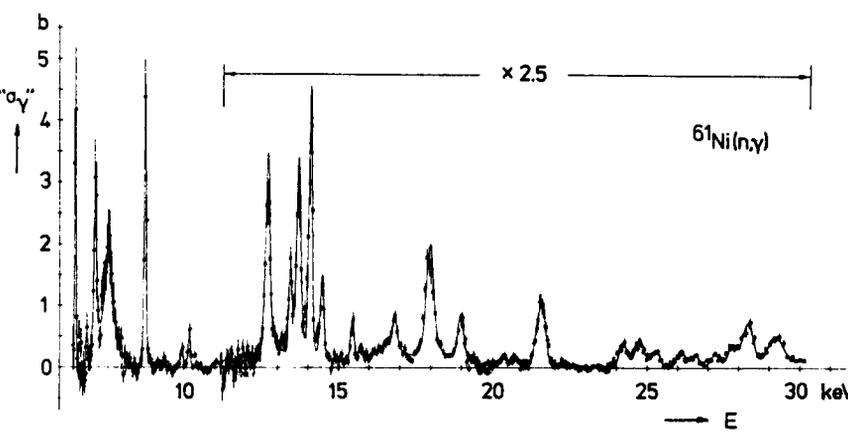
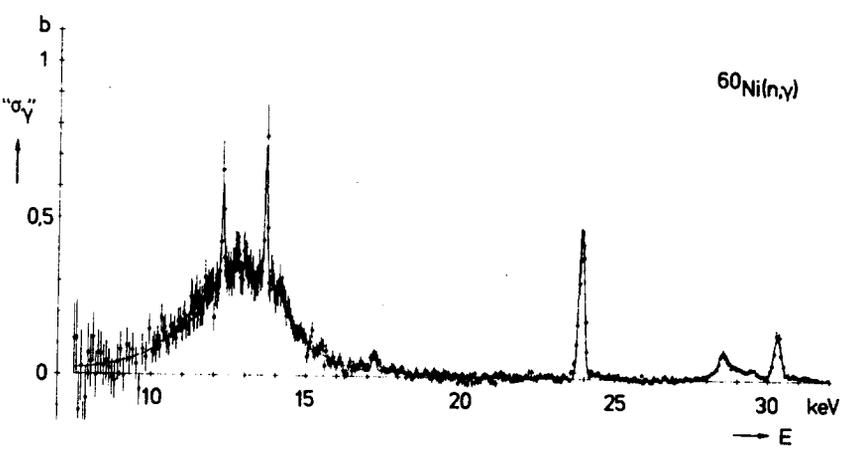
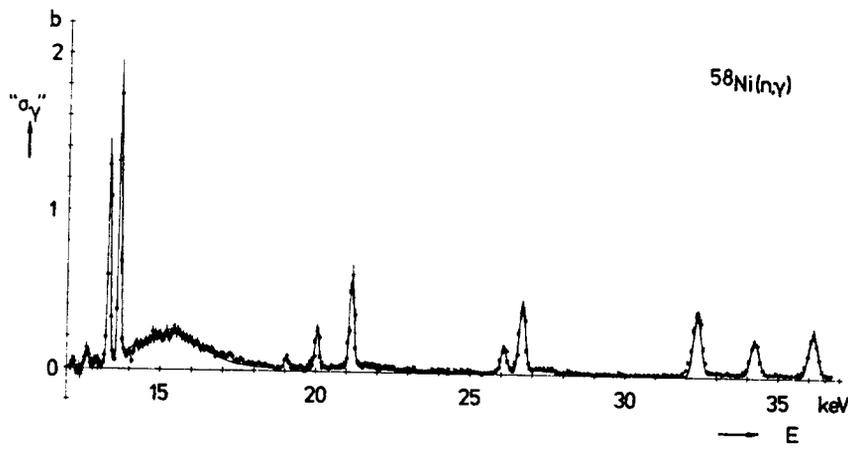
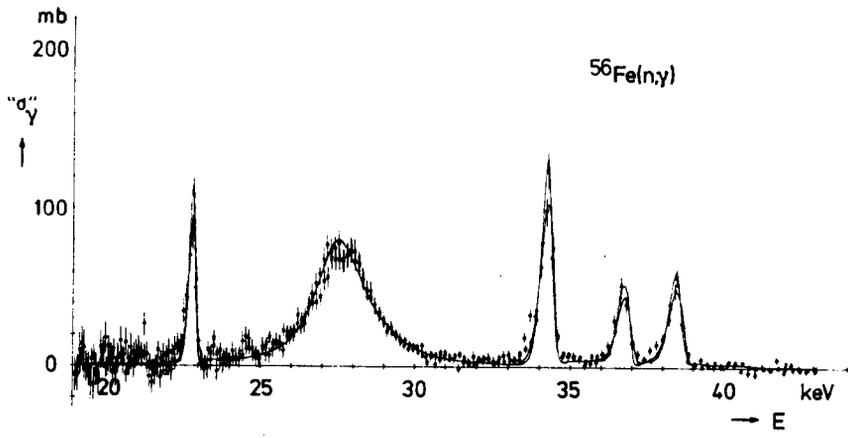
- Fig. 1. Experimental Results for the Thermal Neutron Capture Cross Section of  $^{238}\text{U}$ . The ENDF/B-V Value is also shown.
- Fig. 2. Comparison of Experimental Data with a Theoretically Calculated Cross Section of  $^{238}\text{U}(u, \gamma)$ .
- Fig. 3. Fluctuations of the  $^{238}\text{U}$  Neutron Capture Cross Section in the 10 - 90 keV Region. The Data by DeSaussure et al.<sup>30</sup> Averaged with Various Resolutions Relative to a Smoothly Averaged Cross Section are Shown.
- Fig. 4. Comparison of Recent Experimental Data with a Theoretically Calculated Cross Section of  $^{232}\text{Th}(n, \gamma)$ .
- Fig. 5. The Neutron Capture Cross Sections of  $^{56}\text{Fe}$ ,  $^{58}\text{Ni}$ ,  $^{60}\text{Ni}$  and  $^{61}\text{Ni}$ . Narrow p-Wave Resonances with Large Peak Cross Sections and Wide S-Wave Resonances are Evident. The Odd-Mass Nuclei  $^{61}\text{Ni}$  has a Substantially Larger Cross Section than the Even-Mass Isotopes.
- Fig. 6. The Neutron Capture Cross Sections of the Fission Product Poisons  $^{135}\text{Xe}$  and  $^{149}\text{Sm}$  in the Thermal Energy Range.
- Fig. 7. Experimental Data, a Theoretical Calculation, and two Evaluations of the Neutron Capture Cross Section of  $^{109}\text{Ag}$ . Discrepancies Exceeding a Factor of Two are Obvious in Both, Experimental Results and Evaluations.
- Fig. 8. More Recent Experimental Results for the Fast Neutron Capture Cross Section of  $^{103}\text{Rh}$ . The Data are in a  $\pm 15$  Percent Range, thus Falling Short of the  $\pm 7$  Percent Required for Technological Applications. Cross Section Values, Multiplied by the Squareroot of the Energy, are Shown.
- Fig. 9. A Schematic of the Build-up of the Higher Actinides. Neutron Capture is Indicated by Horizontal Double-Solid Arrows.
- Fig. 10. The Neutron Capture Cross Section of  $^{240}\text{Pu}$ . Experimental Data are Shown Along with a Theoretical Calculation (Solid Line) and the Evaluation of ENDF/B-V (Dotted Line).

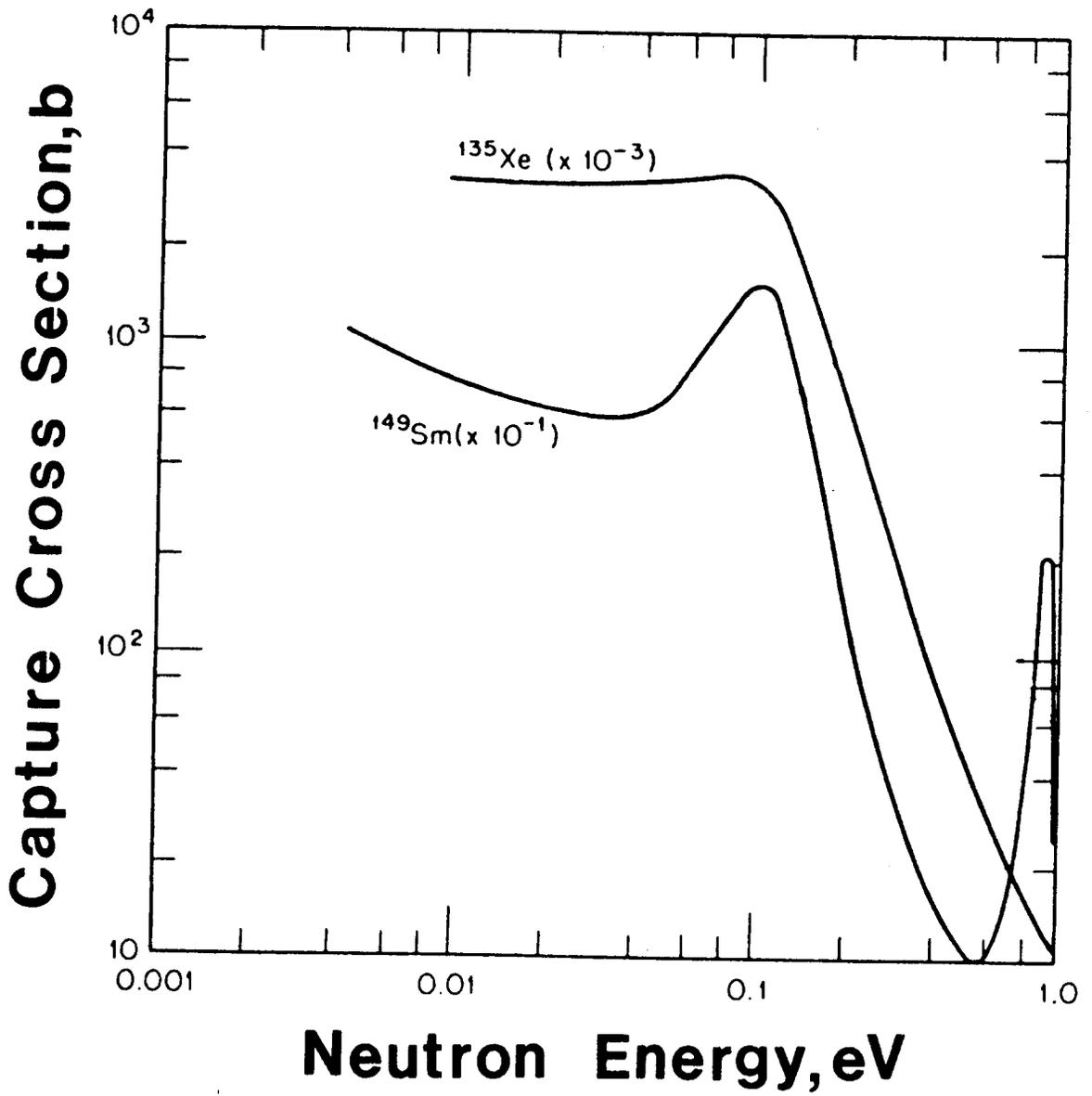


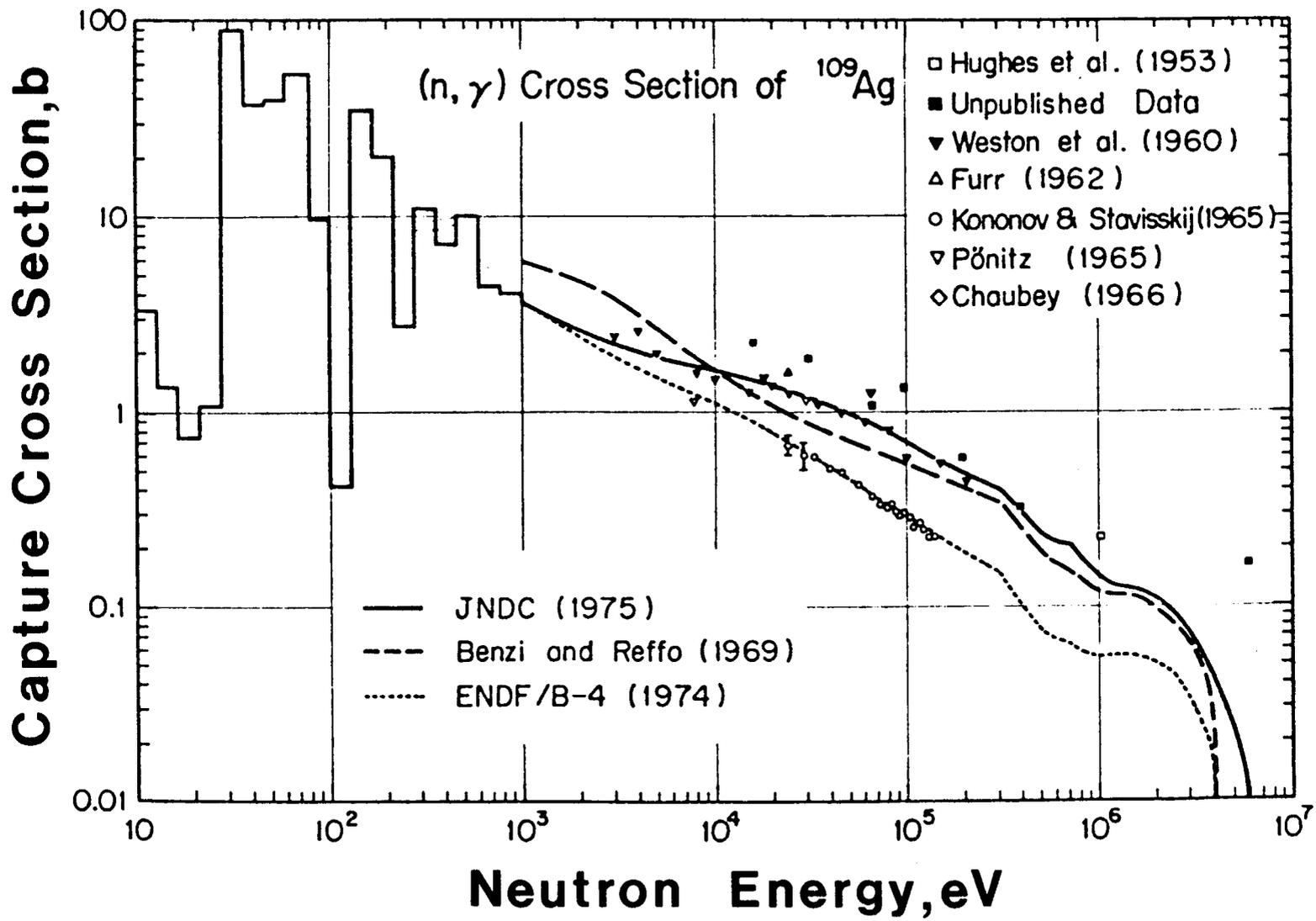


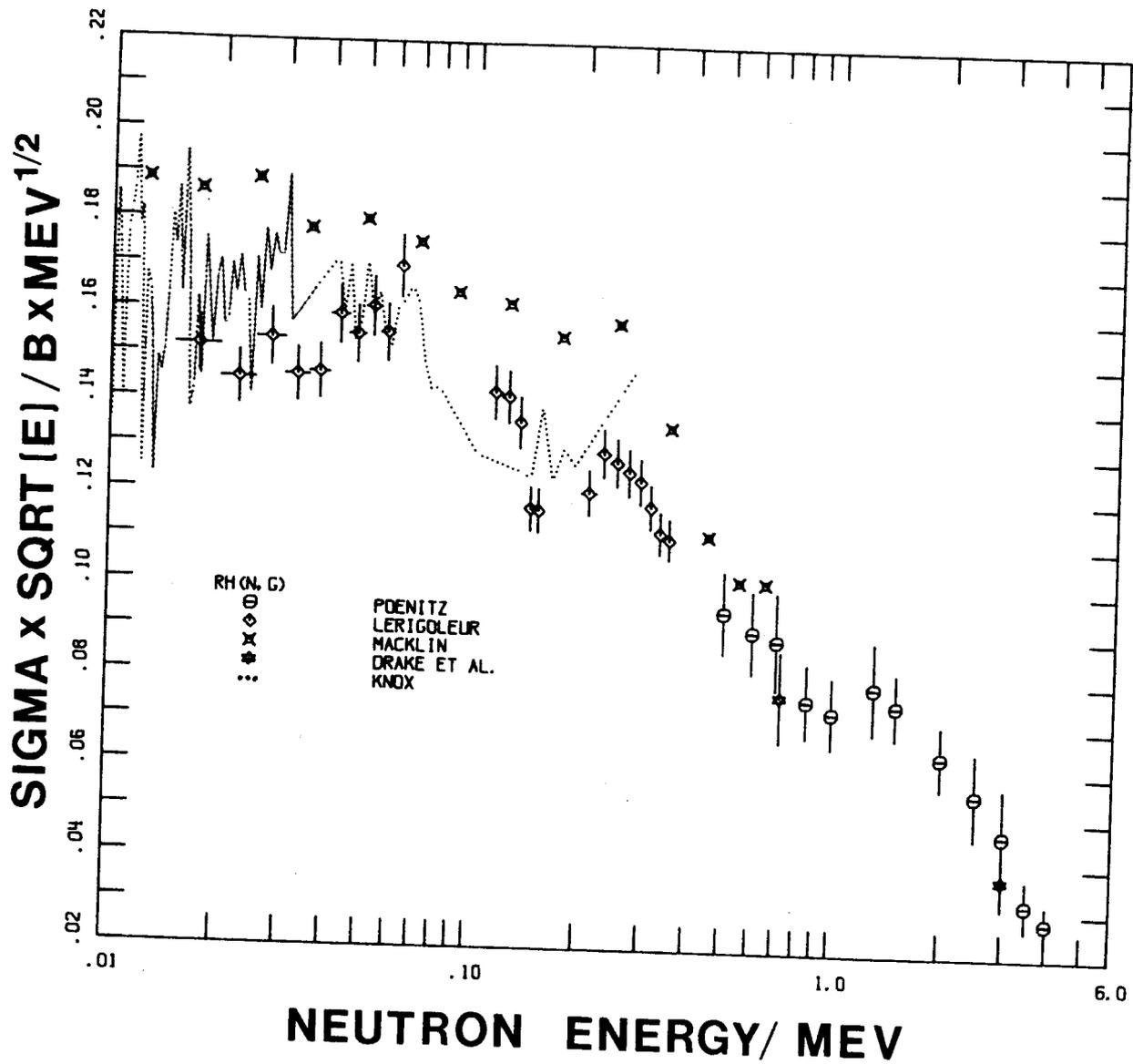


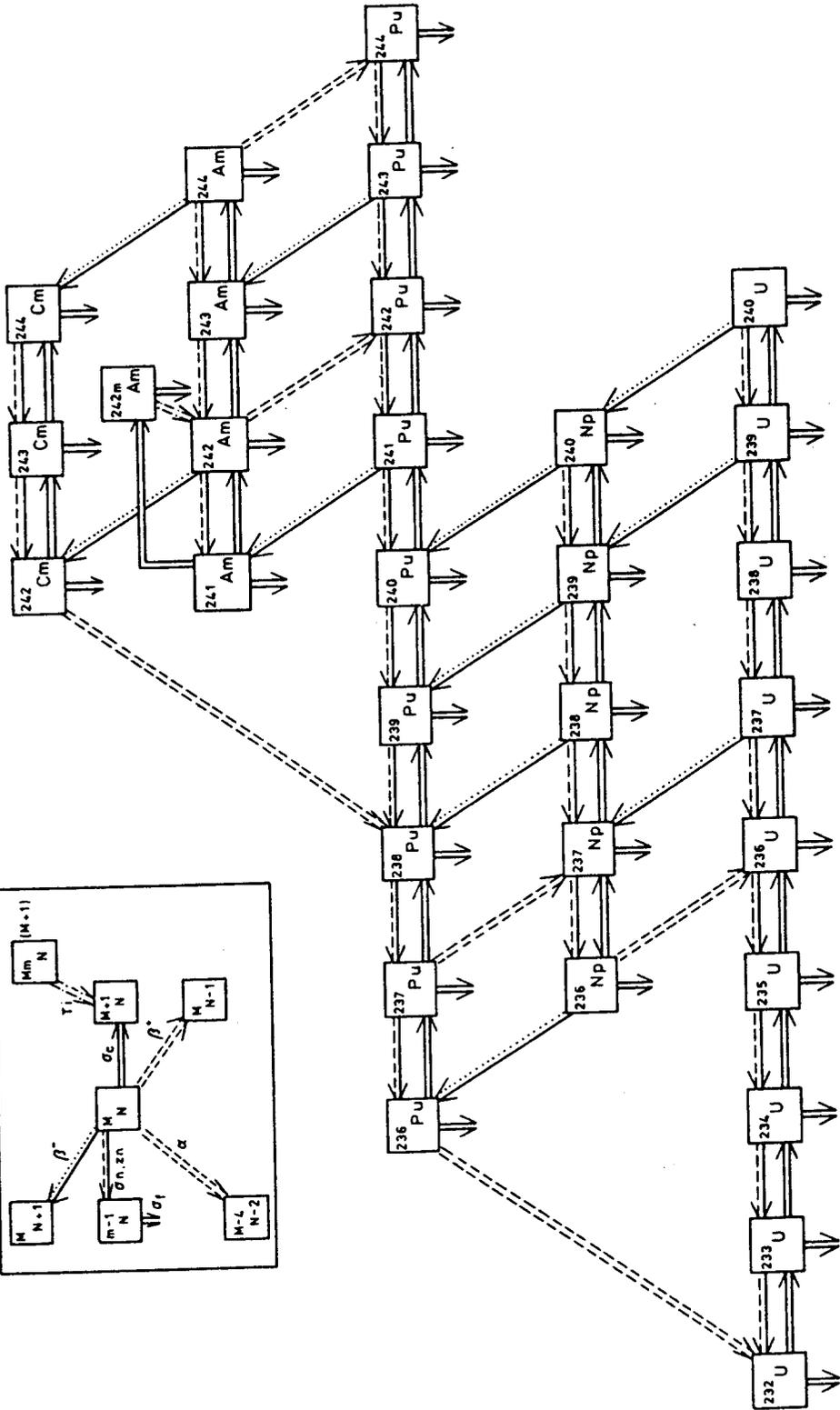
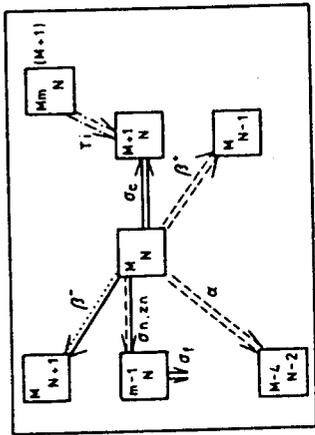




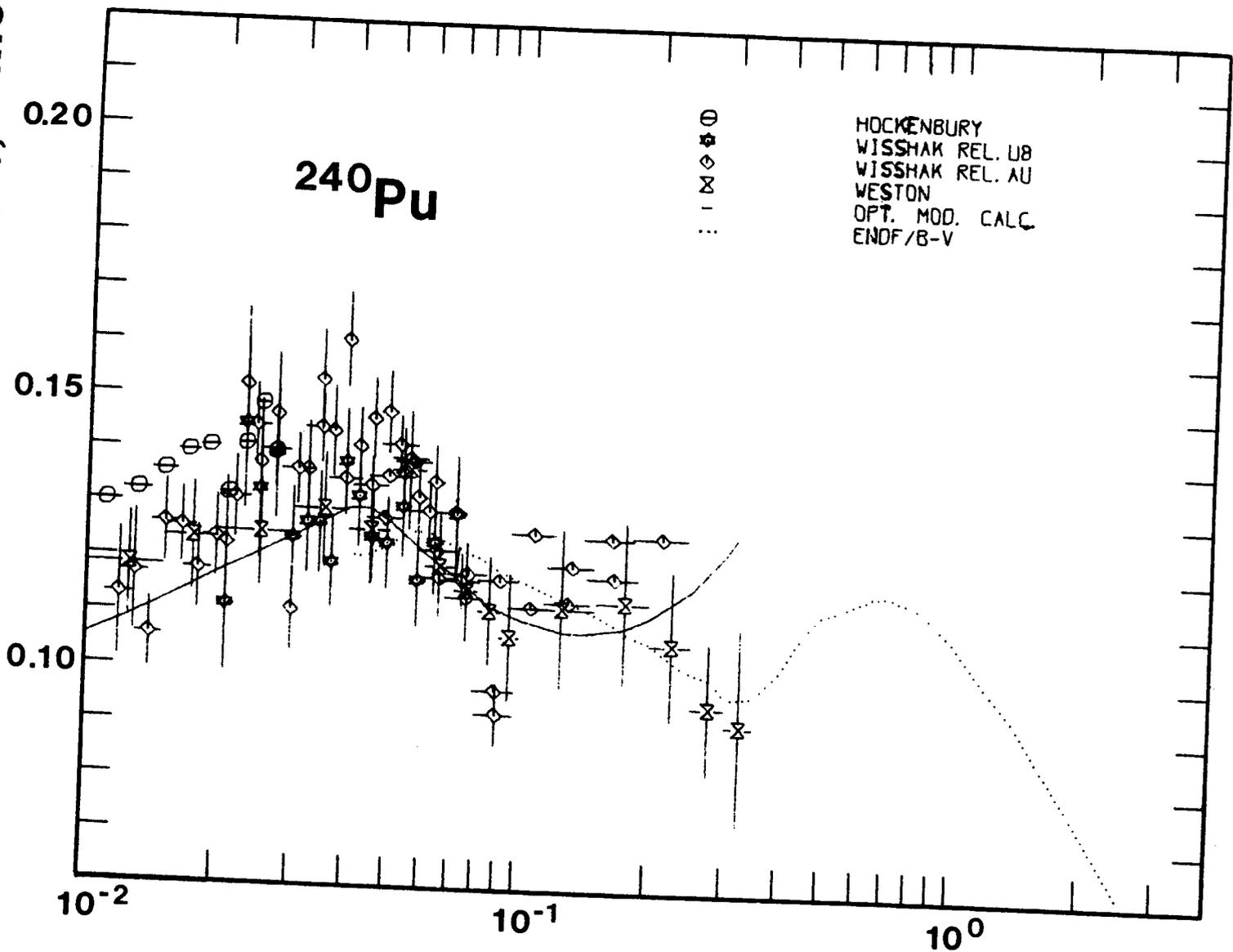








Cross Section  $\times$  SQRT(E), b $\times$ MeV<sup>1/2</sup>



Neutron Energy, MeV

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