High Energy Evaluations of the ^{239}Pu(n,f) Cross Section

Allan D. Carlson
Neutron Physics Group
National Institute of Standards & Technology

Presented at
The AFRI/NPWG Meeting
Brookhaven National Laboratory
November 17, 2011

BACKGROUND INFORMATION

- The earliest $^{239}Pu(n,f)$ ENDF/B evaluations used very straight-forward simple procedures to obtain the cross section.
  - For example with the ENDF/B-IV evaluated by Hunter and Stewart (1973):
    - Only 5 absolute measurements were used.
    - Only 10 ratio measurements to the $^{235}U(n,f)$ cross section.
    - One 1 ratio measurement to the $^{238}U(n,f)$ cross section and it was converted to a ratio to the $^{235}U(n,f)$ cross section.
    - The ratios to the $^{235}U(n,f)$ cross section were evaluated by drawing a smooth curve through the data. Then converted with an evaluated $^{235}U(n,f)$ cross section.
  - The problems with this evaluation:
    - Uncertainty information was not directly used.
    - Full use was not made of the ratio data. Both cross sections should have an impact.
    - Uncertainty/covariance not obtained directly from the evaluation.
  - Only estimates were obtained for the uncertainty.
    - 0.1- 1 MeV 3% 1-20 MeV 6%

THE ENDF/B-V ^{239}Pu(n,f) EVALUATION

•Original evaluation by Kujawski and Stewart (1978).

•Almost the same database as was used for the ENDF/B-IV evaluation.

- •One data set was added.
- •Between 12 and 20 MeV the same database as for ENDF/B-IV.

•An improvement since some correlations between the ^{235}U(n,f) and ^{239}Pu(n,f) cross sections were included.

•Uncertainties were about 3% between 12 and 20 MeV.

•A significant improvement was the conversion of the ratio using an evaluation by Poenitz of the ^{235}U(n,f) cross section.

- •More objective since all measurements since 1965 were used.
  - •data before 1965 were generally poorly documented or had large uncertainties.
- •Separate evaluations of the shape and normalization were each done taking into account weighting based on experimental uncertainties.

THE ENDF/B-VI ^{239}Pu(n,f) EVALUATION

•The original evaluation was part of the standards evaluation by the CSEWG Standards Subcommittee (1987). Details of the evaluation will be given later.

•The standards evaluation was an evaluation of all the standards simultaneously except the H(n,n), ^{3}He(n,p) and C(n,n) standards.

- •The H(n,n) standard was presumed to be so well known that little improvement in that cross section would result from the evaluation process.
- •There are only a small number of measurements of the ^{3}He(n,p) and C(n,n) standards relative to the other standards so they would have little impact on the evaluation.

• The ^{239}Pu(n,f), ^{238}U(n,f) and ^{238}U(n,γ) cross sections, though they are not standards, were used in the evaluation since there are high quality measurements of those cross sections relative to the standards and there are very accurate absolute measurements of those cross sections. Thus they have an impact on the cross sections and the uncertainties of the standards.

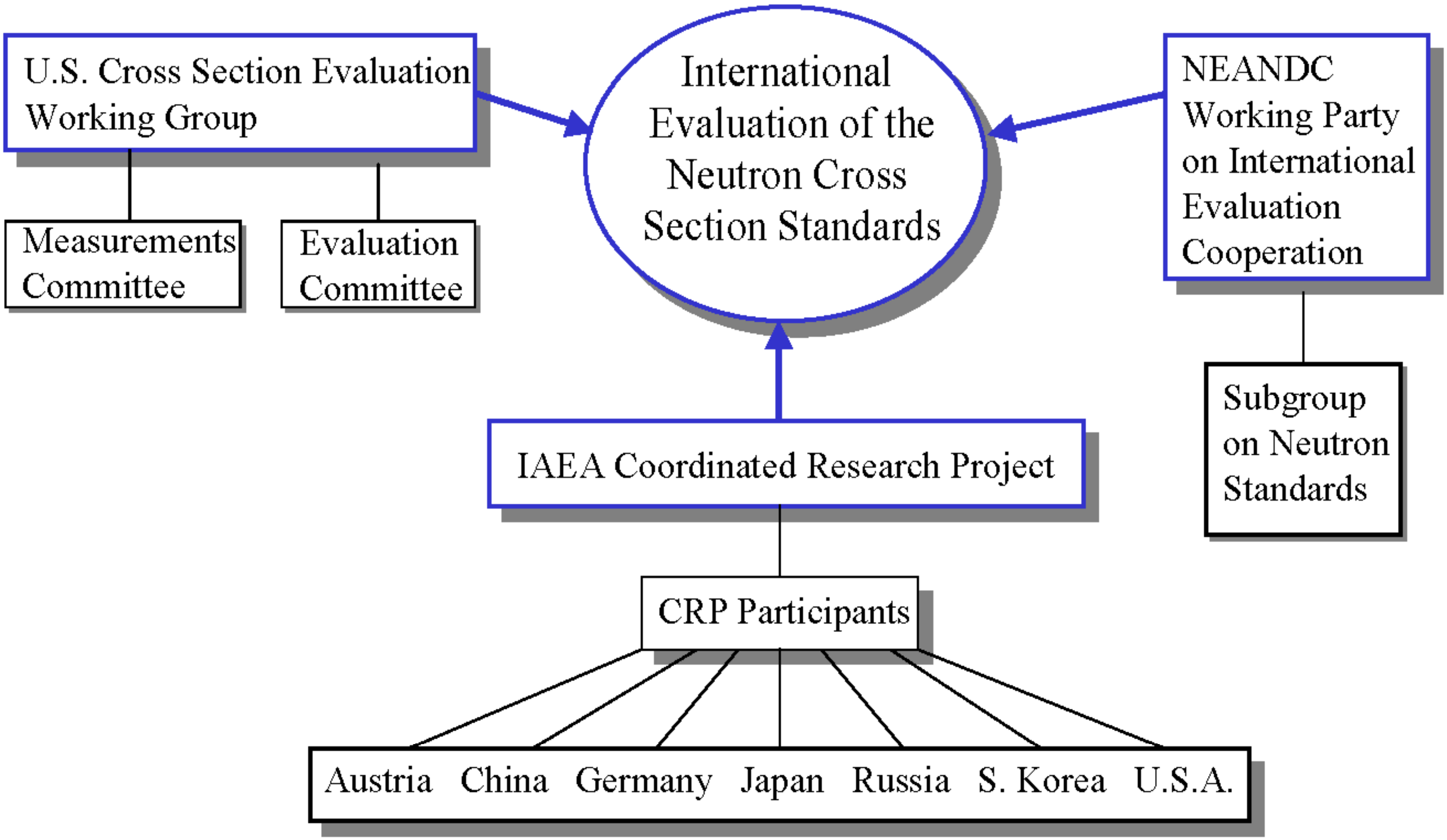
- •There is also the benefit that improvements should occur for these important nuclear application cross sections.

THE ENDF/B-VI ^{239}Pu(n,f) EVALUATION (cont.)

- In 1990 a MOD to this evaluation was made by Young, MacFarlane and Arthur. Part of the energy range of the standards evaluation was replaced by an evaluation using GLUCS with a covariance analysis.
  - They wanted to include 2 new important measurements not available for the standards evaluation.
  - The database included only ^{239}Pu(n,f) ratio measurements to the ^{235}U(n,f) standard and absolute ^{239}Pu(n,f) measurements.
  - Except for the 2 new measurements, the data were taken from the standards database.
  - The ratio measurements and absolute data were evaluated separately.
    - The ratio results were converted using the ENDF/B-VI ^{235}U(n,f) standard cross section.
  - The evaluation was obtained from a smooth curve through the GLUCS output.

- This work is in agreement with the 1987 standards evaluation result except:
  - near 9 MeV where it is about 4% higher.
  - above 15 MeV where it is a few percent higher.
- These higher values result from the higher cross sections for the new measurements.
- The uncertainties on the GLUCS output varied from below a percent from 1 to 5 MeV to above 1% at 15 MeV.

THE EVALUATION OF THE STANDARDS THAT PRODUCED THE ENDF/B-VII STANDARDS



COMPARISON OF THE EVALUATION PROCESS FOR ENDF/B-VI vs ENDF/B-VII STANDARDS

- DATABASE
  - For the ENDF/B-VI evaluation, the database for the least-squares simultaneous evaluation was developed by Poenitz. More than 400 experiments are in that database. Each experiment was carefully checked to see if there were missed corrections, the need for updating of data (such as half-life data or improved standards), etc.

  - For the ENDF/B-VII evaluation, more than 30 additional data sets were added to the database. The database was extended up to 200 MeV from the previous 20 MeV limit.

Typical GMA Experiment Input

```
5231984U5(N,F)            A.D.CARLSON ET AL.
84GEEL,PROC.
1 1 0 1 16   67  8  0  0  0  0
 UNCERTAINTIES
1 U5 MASS
2 FF DET. BIAS
3 FLIGHT PATH (GEOM.)
4 COL. AREA (GEOM.)
5 FF EXTRAPOLATION
ENERGY DEP. UNCERT.
3 STATISTICS
4 FROM TIMING UNCERT.
5 DUE TO TRANS. OF     MATERIALS IN BEAM
6 DUE TO UNCERT. IN    ADC ZERO FOR BLACK DET.
7 DUE TO UNCERT. IN    DEAD TIME CORR.
8 FROM UNCERT. IN BD   EFFICIENCY
9 FROM UNCERT. IN      FISSION CHAMBER BKGND
10 FROM UNCERT IN      BD BKGND
11 FROM UNCERT.IN SCATT. FROM MATERIAL IN F.C.
1.2  .0  .0  .2  .5  .0  .0  .0  .0  .0 1  1  1  1  1  0  0  0  0  0
 .00  .00  .00
 .00  .00  .00
 .00  .00  .00
 .50  .50  .50
 .50  .50  .50
 .50  .50  .50
 .50  .50  .50
 .50  .50  .50
 .50  .50  .50
 .50  .50  .50
 .50  .50  .50
 0  0  9  1  1  1  1  1  1  1  1
.3097E+00 .1227E+01  .2  2.3  1.0  .4  .6  .5  .1  1.0  .3  .4  .1  2.2
.3245E+00 .1242E+01  .2  2.4  1.0  .4  .5  .5  .1  1.0  .3  .4  .1  2.2
.3391E+00 .1223E+01  .2  2.0  1.0  .3  .5  .5  .1  1.0  .3  .4  .1  2.1
```

Systematic Uncertainties

Experimental Data:
E, σ, ΔE, Res, Stat, ΔT, Trans, ADC0, DTC, BD eff, FC Bkg, BD Bkg, FC scatt Corr

UNCERTAINTY COMPONENTS FOR SET 523

5231984U5(N,F)   A.D.CARLSON ET AL.   84GEEL,PROC.
1 1 0 1 16 67 8 0 0 0 0
UNCERTAINTIES
1 U5 MASS
2 FF DET. BIAS
3 FLIGHT PATH (GEOM.)
4 COL. AREA (GEOM.)
5 FF EXTRAPOLATION
ENERGY DEP. UNCERT.
3 STATISTICS
4 FROM TIMING UNCERT.
5 DUE TO TRANS. OF   MATERIALS IN BEAM
6 DUE TO UNCERT. IN   ADC ZERO FOR BLACK DET.
7 DUE TO UNCERT. IN   DEAD TIME CORR.
8 FROM UNCERT. IN BD   EFFICIENCY
9 FROM UNCERT. IN   FISSION CHAMBER BKGND
10 FROM UNCERT IN   BD BKGND
11 FROM UNCERT.IN SCATT. FROM MATERIAL IN F.C.
1.2 .0 .0 .2 .5 .0 .0 .0 .0 .0 1 1 1 1 1 0 0 0 0 0

TABULATED UNCERTAINTIES FOR SET 523

En	Sig	DE	Res	Stat	Tm. u.	Mat Tr	ADC 0	DTC.	BD eff	FC Bk	BD Bk	FC Sc	Tot. U.
.3097E+00	.1227E+01	.2	2.3	1.0	.4	.6	.5	.1	1.0	.3	.4	.1	2.2
.3245E+00	.1242E+01	.2	2.4	1.0	.4	.5	.5	.1	1.0	.3	.4	.1	2.2
.3391E+00	.1223E+01	.2	2.0	1.0	.3	.5	.5	.1	1.0	.3	.4	.1	2.1
.3534E+00	.1213E+01	.2	2.1	1.0	.3	.5	.5	.1	1.0	.3	.4	.1	2.1
.3686E+00	.1224E+01	.2	2.1	1.0	.1	.6	.4	.1	1.0	.3	.4	.1	2.1
.3840E+00	.1205E+01	.2	2.0	1.0	.2	.5	.4	.1	1.0	.3	.4	.1	2.1
.3996E+00	.1179E+01	.2	2.0	1.0	.1	.5	.4	.1	1.0	.3	.4	.1	2.1
.4171E+00	.1214E+01	.2	2.3	1.0	.1	.7	.4	.1	1.0	.3	.4	.1	2.2
.4367E+00	.1201E+01	.2	2.3	1.0	.4	.9	.4	.1	1.0	.3	.4	.1	2.3
.4556E+00	.1134E+01	.2	1.9	1.0	.7	.7	.3	.1	1.0	.3	.4	.1	2.3
.4727E+00	.1132E+01	.2	1.8	1.0	.7	.6	.3	.1	1.0	.3	.4	.1	2.2
.4897E+00	.1123E+01	.2	1.8	1.0	.4	.5	.3	.1	1.0	.3	.4	.1	2.1
.5065E+00	.1118E+01	.2	1.6	1.0	.4	.5	.3	.1	1.0	.3	.4	.1	2.1

SIMULTANEOUS EVALUATION DATABASE

$^{6}Li(n,t)$ → ^{6}Li total cross section

$^{6}Li(n,n)$ → ^{6}Li total cross section

$^{10}B(n,\alpha_0)$ → ^{10}B total cross section

$^{10}B(n,\alpha_1)$ → ^{10}B total cross section

$^{10}B(n,n)$ → ^{10}B total cross section

$Au(n,\gamma)$

$^{235}U(n,f)$

$^{238}U(n,f)$

$^{238}U(n,\gamma)$

$^{239}Pu(n,f)$

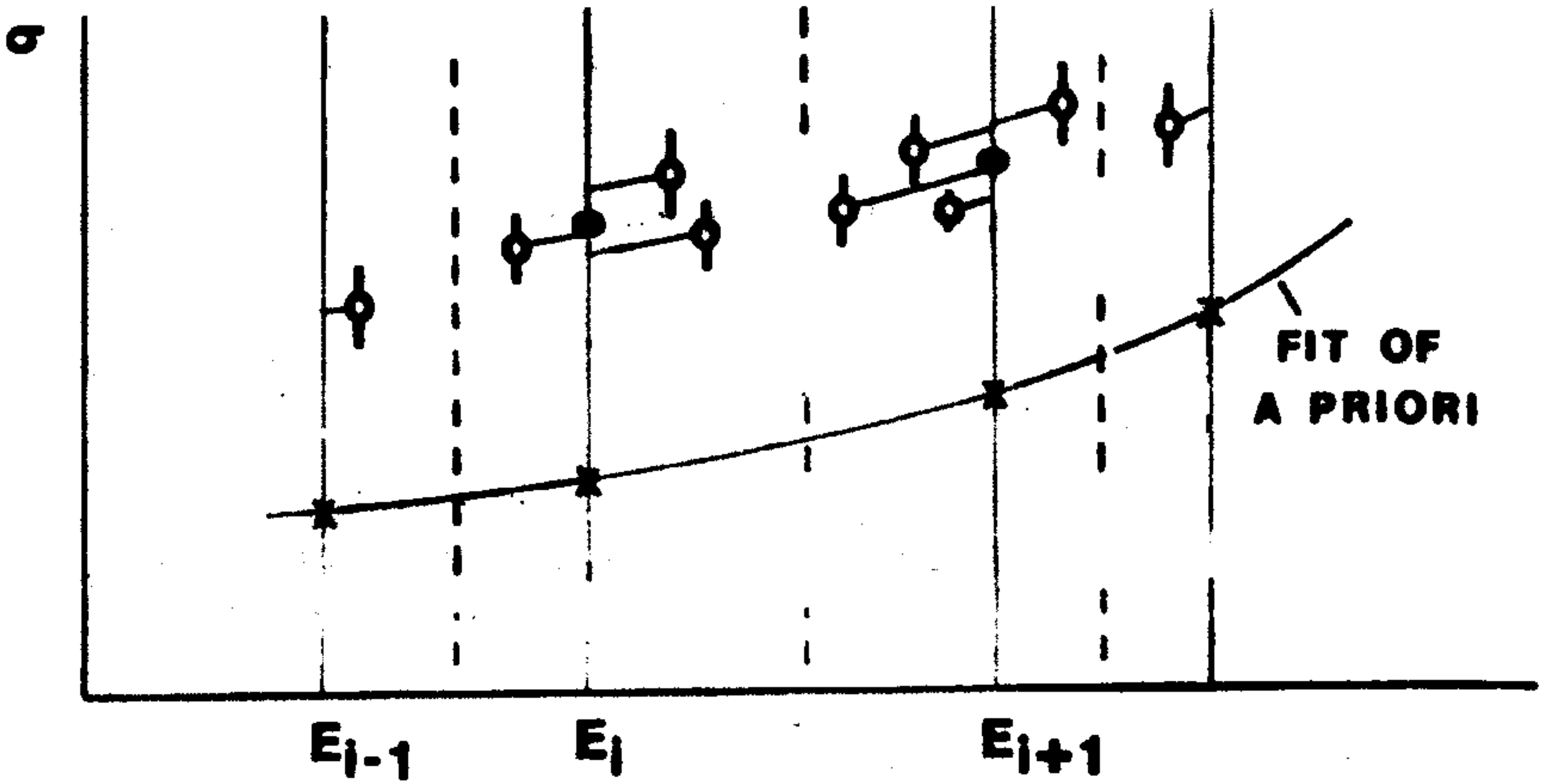
DATA TYPES USED IN THE SIMULTANEOUS EVALUATION

Type	Data Type	Example
1	Absolute cross section	$\sigma_{nf}(^{235}U)$
2	Cross section shape	$c*\sigma_{n\alpha}(^{6}Li)$, c unknown
3	Absolute cross section ratio	$\sigma_{nf}(^{238}U)/\ \sigma_{nf}(^{235}U)$
4	Ratio shape	$c*\sigma_{nf}(^{235}U)/\sigma_{n\alpha}(^{6}Li)$ c unknown
5	Sum of cross sections	$\sigma_{tot}(^{6}Li) = \sigma_{nn}(^{6}Li) + \sigma_{n\alpha}(^{6}Li)$
6	Spectrum averaged cross section	$\sigma_{nf}(^{235}U)$ averaged over ^{252}Cf spont. fission spect
7	Absolute ratio of cross section/ sum of cross sections	$\sigma_{nf}(^{235}U)/\ \sigma_{n\alpha}(^{10}B)$, where $\sigma_{n\alpha}(^{10}B) = \sigma_{n\alpha_0}(^{10}B) + \sigma_{n\alpha_1}(^{10}B)$
8	Shape of Type 5 data	
9	Shape of Type 7 data	

TOTAL NUMBER OF DATA SETS FOR REACTIONS AND THEIR RATIOS IN THE GMA DATABASE. VALUES IN BRACKETS ARE THE NUMBER OF ABSOLUTE CROSS SECTIONS.

	^{6}Li(n,t)	^{10}B(n,α_0)	^{10}B(n,α_1)	^{10}B(n,α)	Au(n,γ)	^{238}U(n,γ)	^{235}U(n,f)	^{239}Pu(n,f)	^{238}U(n,f)
^{6}Li(n,t)	18 (7)								
^{10}B(n,α_0)	0	5 (4)							
^{10}B(n,α_1)	1 (0)	12 (10)	11 (2)						
^{10}B(n,α)	4 (0)	0	0	5 (2)					
Au(n,γ)	3 (3)	0	6 (3)	4 (4)	27 (21)				
^{238}U(n,γ)	2 (2)	0	9 (5)	4 (4)	10 (9)	14 (11)			
^{235}U(n,f)	14 (0)	0	2 (1)	25 (0)	12 (10)	12 (6)	68 (52)		
^{239}Pu(n,f)	2 (0)	0	0	19(0)	0	1 (0)	19 (14)	22 (19)	
^{238}U(n,f)	2 (1)	0	0	0	0	0	34 (29)	3 (1)	18 (11)

ENERGY GRID AND PROCEDURE FOR OBTAINING "EXPERIMENTAL" VALUES ON AN ENERGY GRID USING GMA



COMPARISON OF THE EVALUATION PROCESS FOR ENDF/B-VI vs ENDF/B-VII STANDARDS

- **Least-squares simultaneous evaluations**.
- Such evaluations are model independent so they can be applied to both the light and heavy nuclide standards. Ratio data can be evaluated simultaneously with cross section and other types of data.
  - For the ENDF/B-VI evaluation:
    - only one code, the generalized least-square code GMA was used.
    - GMA was checked against GLUCS and the results agreed.
    - A problem was observed when correlated discrepant data were used.
      - The resulting output was too low. The problem was first found with model-independent least-squares analyses. This is the Peelle's Pertinent Puzzle (PPP) problem. To remove this problem, discrepant data greater than three standard deviations from the output results were down weighted. This also led to χ^2 per degree of freedom of about 1.

COMPARISON OF THE EVALUATION PROCESS FOR ENDF/B-VI vs ENDF/B-VII STANDARDS (cont.)

Least-squares simultaneous evaluations (cont.)

- For the ENDF/B-VII evaluation:
  - considerably more work was done on comparisons of least-squares codes than that during the ENDF/B-VI evaluation. Comparisons were done with the GMAP, GLUCS, PADE and SOK codes for simple cases. When used under the same conditions there was general agreement on the results.
  - The SOK and GMAP codes were compared using the full standards database. The same covariance matrices of the uncertainties was used for each analysis. The agreement between the two fits was generally good.
  - The use of medium energy range correlations for groups of points several standard deviations from the output results was more often used for the ENDF/B-VII evaluation. The length of the correlation component was determined from the energy dependency of the discrepancy.
  - For the ENDF/B-VII evaluation, PPP was handled more properly.
    - To do so, uncertainties were expressed as factional uncertainties for input to GMA (now renamed GMAP). Several other PPP reduction methods were also compared in a number of trials. The agreement was within about 0.3%

COMPARISON OF THE EVALUATION PROCESS FOR ENDF/B-VI vs ENDF/B-VII STANDARDS (cont.)

- **R-matrix evaluations**
  - It was realized that useful data could be added to the evaluation process by adding R-matrix evaluations to the process. They would be used for reactions leading to the ^{7}Li and ^{11}B compound nuclei. This work could then provide improvements in ^{6}Li(n,t) and ^{10}B(n,α) standards. So in addition to neutron cross section data, a large database of charged-particle, polarization and differential cross section data could then be used.
    - For the ENDF/B-VI evaluation, only one code, EDA was used
    - For the ENDF/B-VII evaluation:
      - two codes were used, EDA and RAC. These two codes work differently. The fits used different expressions for the $\chi 2$ minimized function.
        - For EDA only a normalization uncertainty and a statistical uncertainty are associated with each point in a data set.
        - For RAC, all uncertainties can be handled and it includes correlation components.
      - For a simple ^{6}Li(n,t) cross section test, agreement in principle, except for a small PPP effect with RAC, was obtained between the codes using SAMMY as an intermediary.

R-MATRIX EVALUATION DATABASE

^{6}Li Total		^{10}B Total	
$^{6}Li(n,n)$	Integral Data	$^{10}B(n,n)$	Integral Data
$^{6}Li(n,n)$	Differential Data	$^{10}B(n,n)$	Differential Data
$^{6}Li(n,n)$	Polarization Data	$^{10}B(n,n)$	Polarization Data
$^{6}Li(n,t)$	Integral Data	$^{10}B(n,\alpha_0)$	Integral Data
$^{6}Li(n,t)$	Differential Data	$^{10}B(n,\alpha_0)$	Differential Data
$^{6}Li(n,t)$	Polarization Data	$^{10}B(n,\alpha_1)$	Integral Data
$^{4}He(t,n)$	Differential Data	$^{10}B(n,\alpha_1)$	Differential Data
$^{4}He(t,t)$	Differential Data	$^{7}Li(\alpha_0,\alpha_0)$	Differential Data
$^{4}He(t,t)$	Polarization Data	$^{7}Li(\alpha,\alpha_1)$	Differential Data
		$^{7}Li(\alpha,n)$	Differential Data

COMPARISON OF THE EVALUATION PROCESS FOR ENDF/B-VI vs ENDF/B-VII STANDARDS (cont.)

•Evaluation procedure

- For the ENDF/B-VI evaluation,
  - The evaluation of the standards (except the H(n,n), ^{3}He(n,p), and C(n,n) cross sections) and the ^{239}Pu(n,f), ^{238}U(n,f), ^{238}U(n,g) was done by combining the results of a simultaneous evaluation (GMA) and R-matrix analyses (EDA).
  - An energy grid was defined which is the same for all cross sections involved in the evaluation, and the fitting parameters were the values of the cross sections for these grid points.
  - It was assumed that the individual fitting for the simultaneous and R-matrix evaluations would include computations of sums that could be combined to produce the same overall output parameters as would have been obtained from a global least-squares fit of all the input data.
  - To avoid some problems, it was decided that the boron and lithium experimental data would be separated into two uncorrelated groups, one to be used in the R-matrix analysis and the other in the simultaneous analysis.
  - Ratio measurements and correlated data were used in the simultaneous evaluation.

ENDF/B-VI STANDARDS EVALUATION PROCEDURE

THERMAL DATA FOR
^{233}U, ^{235}U, ^{239}Pu, ^{241}Pu

THERMAL CONSTANTS EVALUATION

^{6}Li+n, ^{10}B+n, Au(n,γ),
^{235}U(n,f), ^{238}U(n,f)
^{238}U(n,γ), ^{239}Pu(n,f)

SIMULTANEOUS EVALUATION

COMBINING PROGRAM

FINAL RESULTS

^{6}Li+n, ^{10}B+n, CHARGED PARTICLE DATA

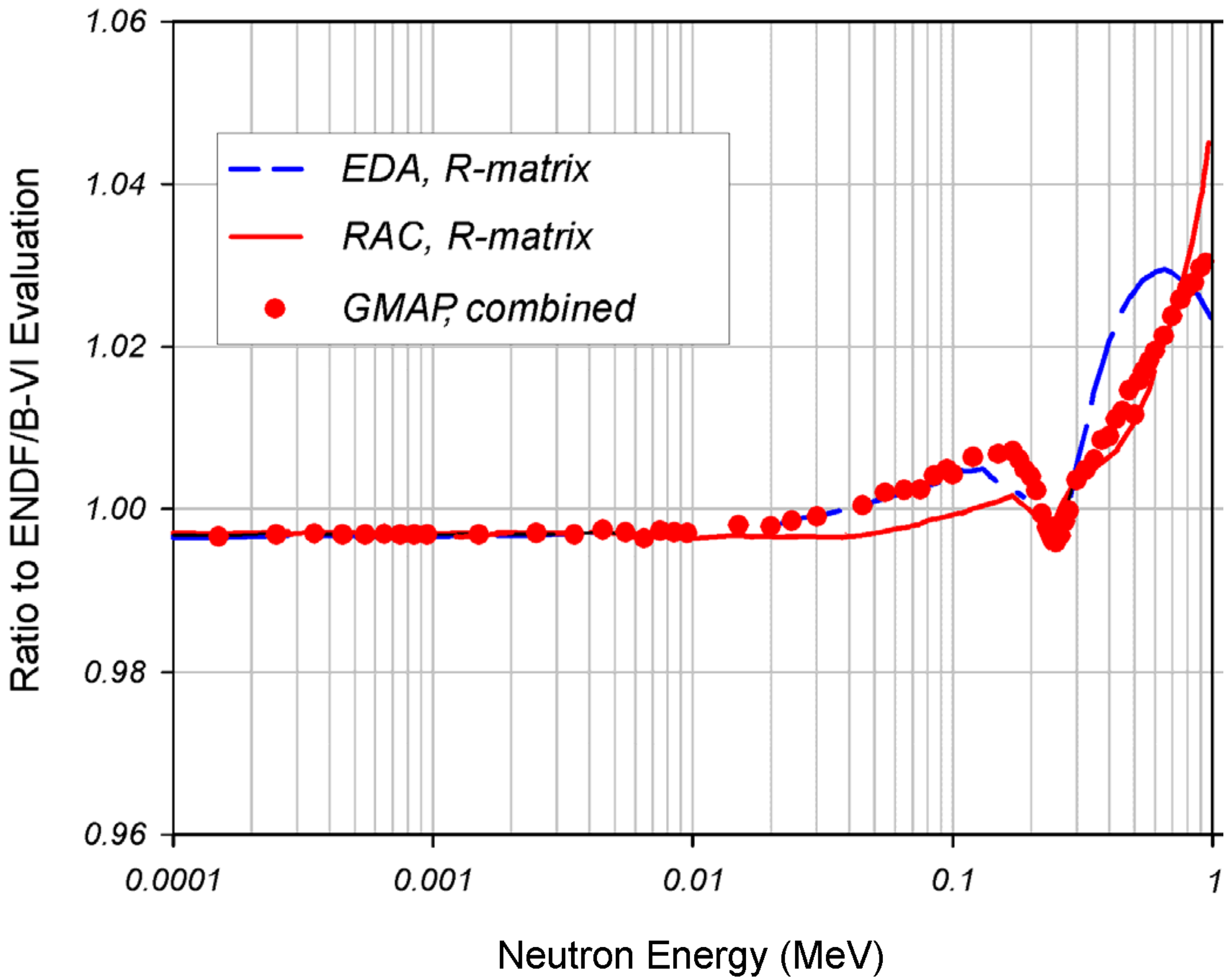
R-MATRIX ANALYSES

COMPARISON OF THE EVALUATION PROCESS FOR ENDF/B-VI vs ENDF/B-VII STANDARDS (cont.)

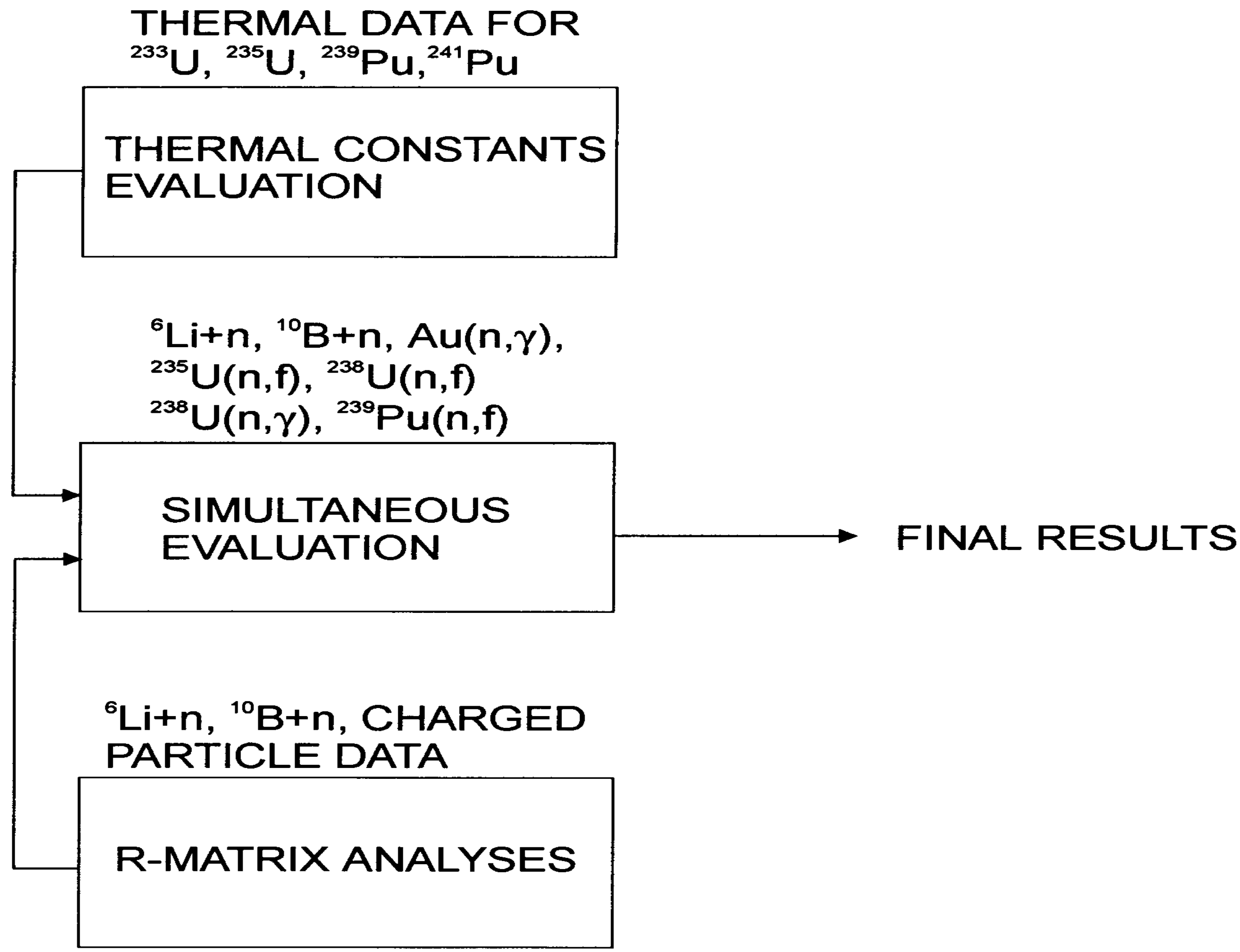
Evaluation procedure (cont.)

- For the ENDF/B-VII evaluation:
  - The procedure is similar to that used for the ENDF/B-VI evaluation except:
    - There are now more than 430 experiments in the database
    - The combining procedure was simpler.
    - The lithium and boron databases were partitioned differently – the only data from those databases used in the GMAP code directly were the ratio data.
    - Two R-matrix codes were used – EDA and RAC.
    - The results from the EDA and RAC analyses were not identical.
      - The differences were the greatest at the highest energies, for example, for the ^{6}Li(n,t) cross section the maximum difference was at 0.5 MeV and it was less than 2%.
      - For the ^{6}Li(n,t), ^{10}B(n,α) and ^{10}B(n,$\alpha_1\gamma$) evaluations, the cross sections from EDA and RAC were averaged (un-weighted) and used as the input to GMAP.
      - At each energy point, half the difference between the EDA and RAC results was treated as a model uncertainty that was an additional component of uncertainty that was added to the components of the total uncertainty for the R-matrix evaluation.

COMPARISON of THE ^{6}Li(n,t) CROSS SECTION FROM THE ENDF/B-VI EVALUATION WITH ENDF/B-VII FITS



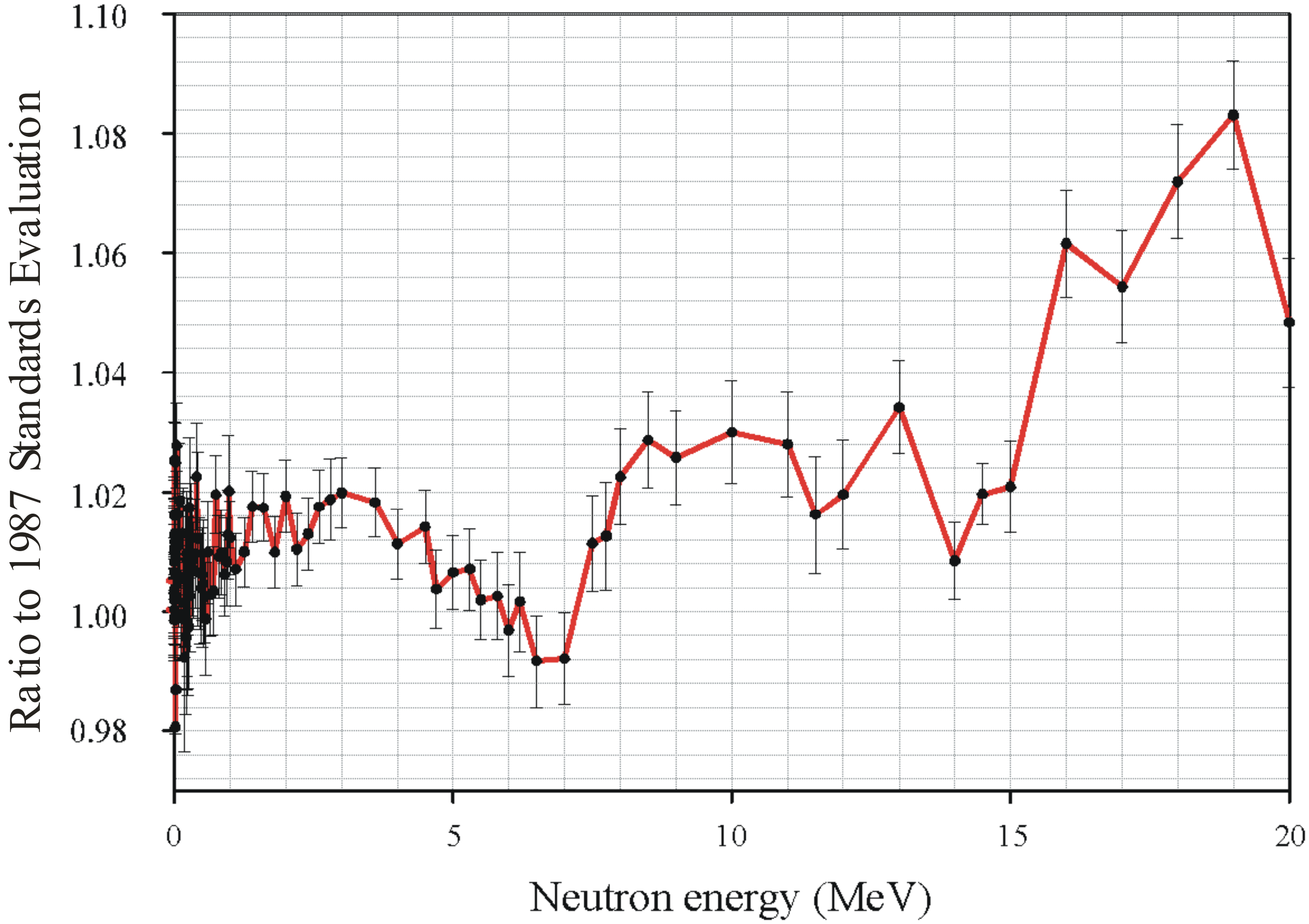
NEW STANDARDS EVALUATION PROCEDURE



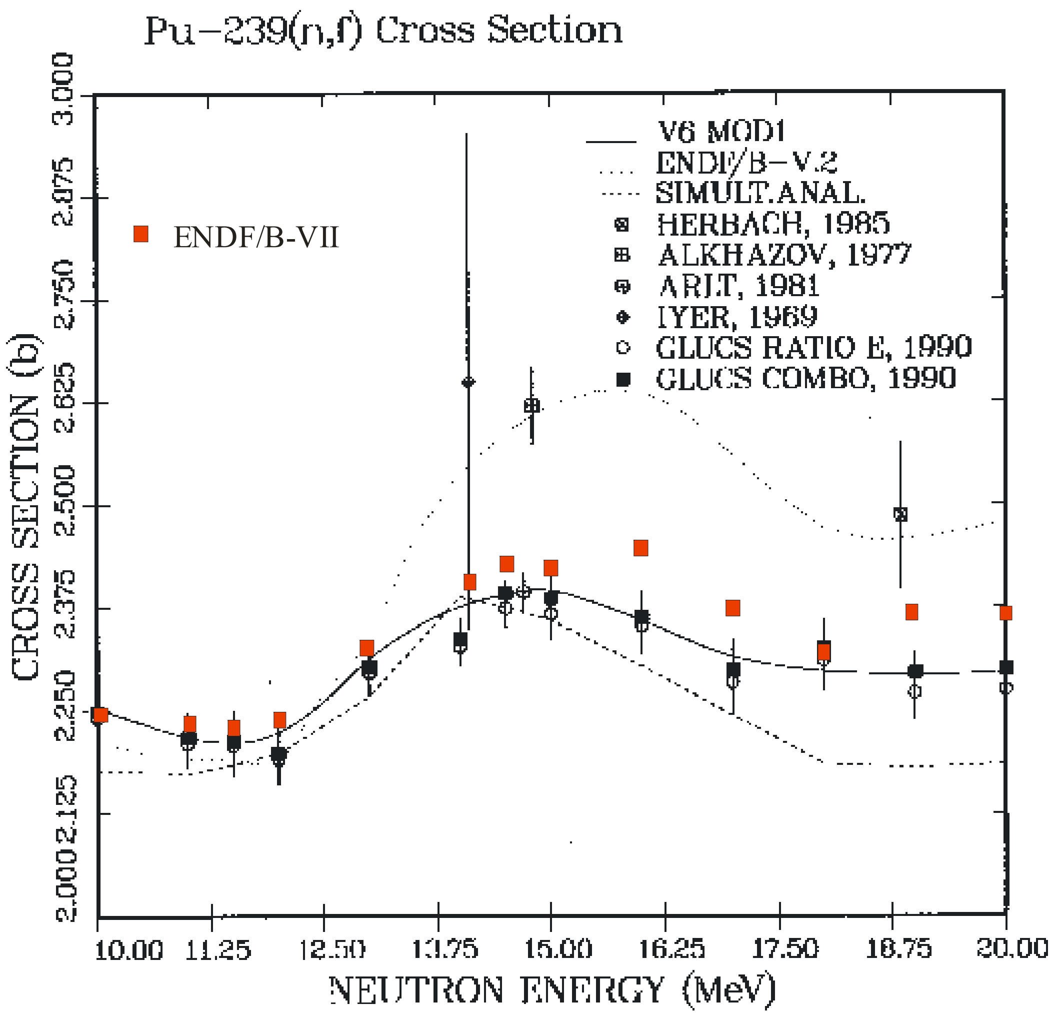
THE NEUTRON CROSS SECTION STANDARDS

Reaction	Energy Range
H(n,n)	1 keV to 20 MeV
^{3}He(n,p)	thermal to 50 keV
^{6}Li(n,t)	thermal to 1 MeV
^{10}B(n,α)	thermal to 1 MeV
^{10}B(n,$\alpha_1\gamma$)	thermal to 1 MeV
C(n,n)	thermal to 1.8 MeV
^{197}Au(n,γ)	thermal, 0.2 to 2.5 MeV
^{235}U(n,f)	thermal, 0.15 to 200 MeV
^{238}U(n,f)	2 to 200 MeV

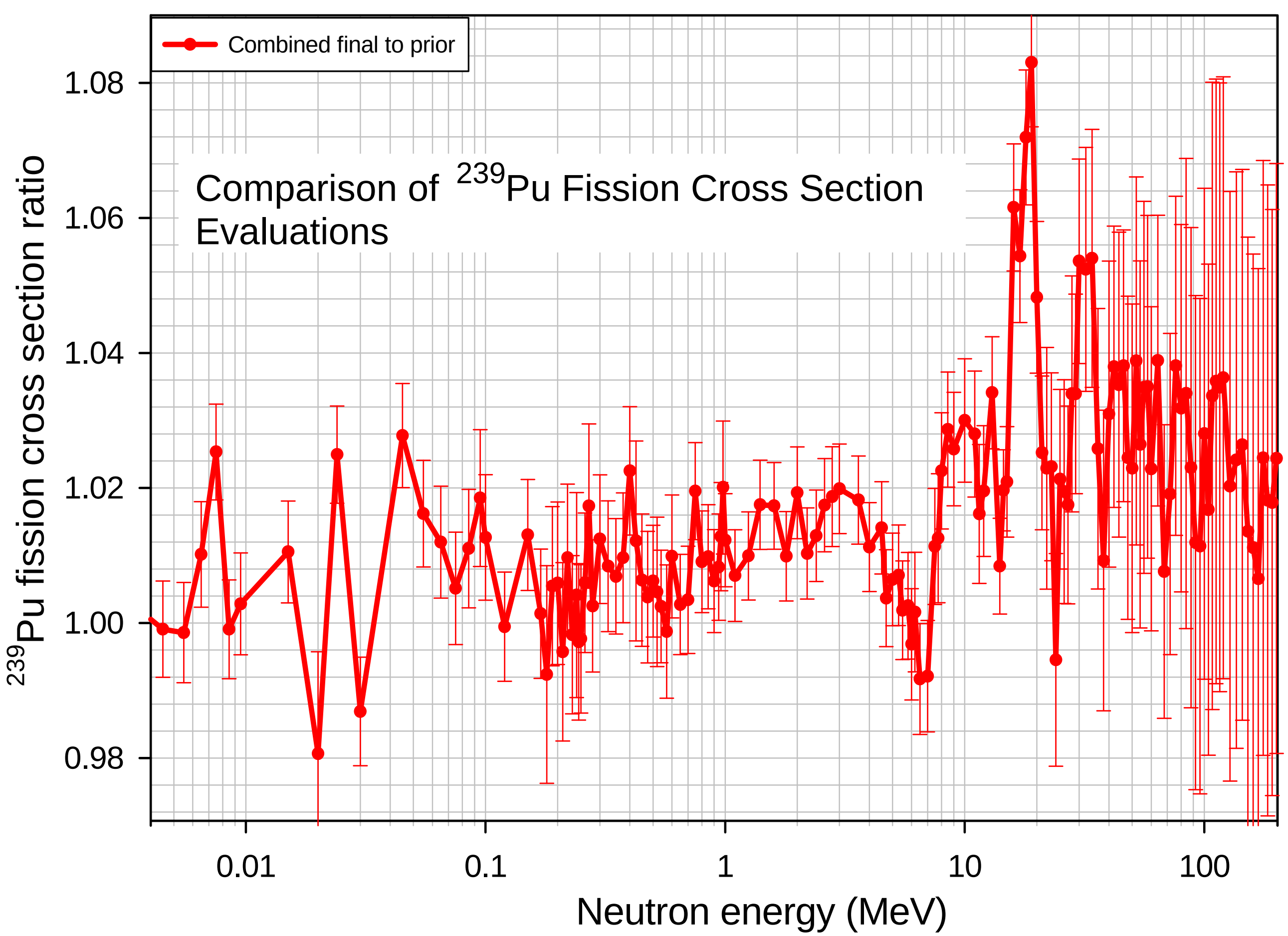
COMPARISON OF THE ENDF/B-VI AND ENDF/B-VII STANDARDS EVALUATIONS of THE ^{239}Pu(n,f) CROSS SECTION UP TO 20 MeV



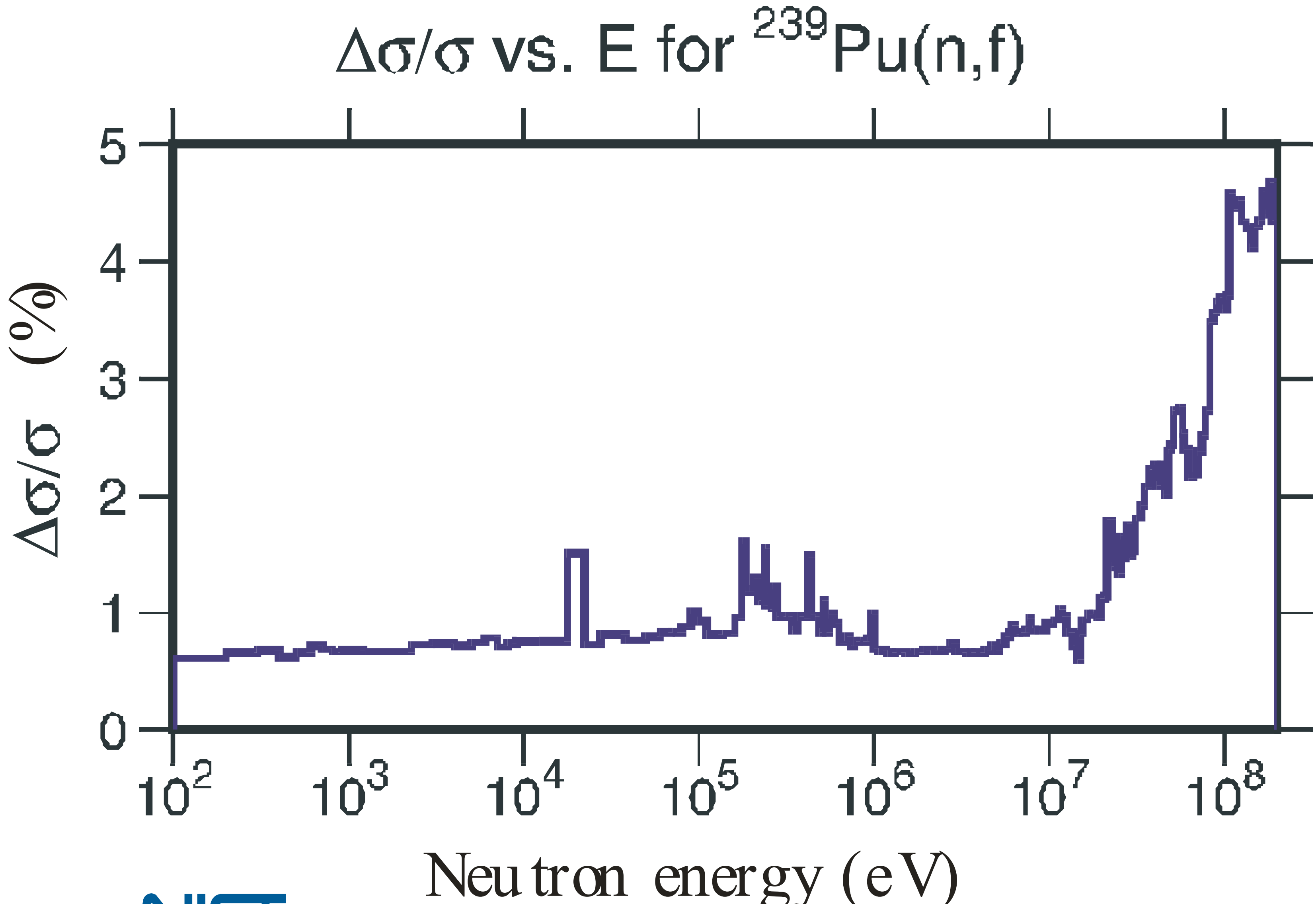
COMPARISON OF VARIOUS EVALUATIONS AND MEASUREMENTS OF THE ^{239}Pu(n,f) CROSS SECTION FROM 10-20MeV

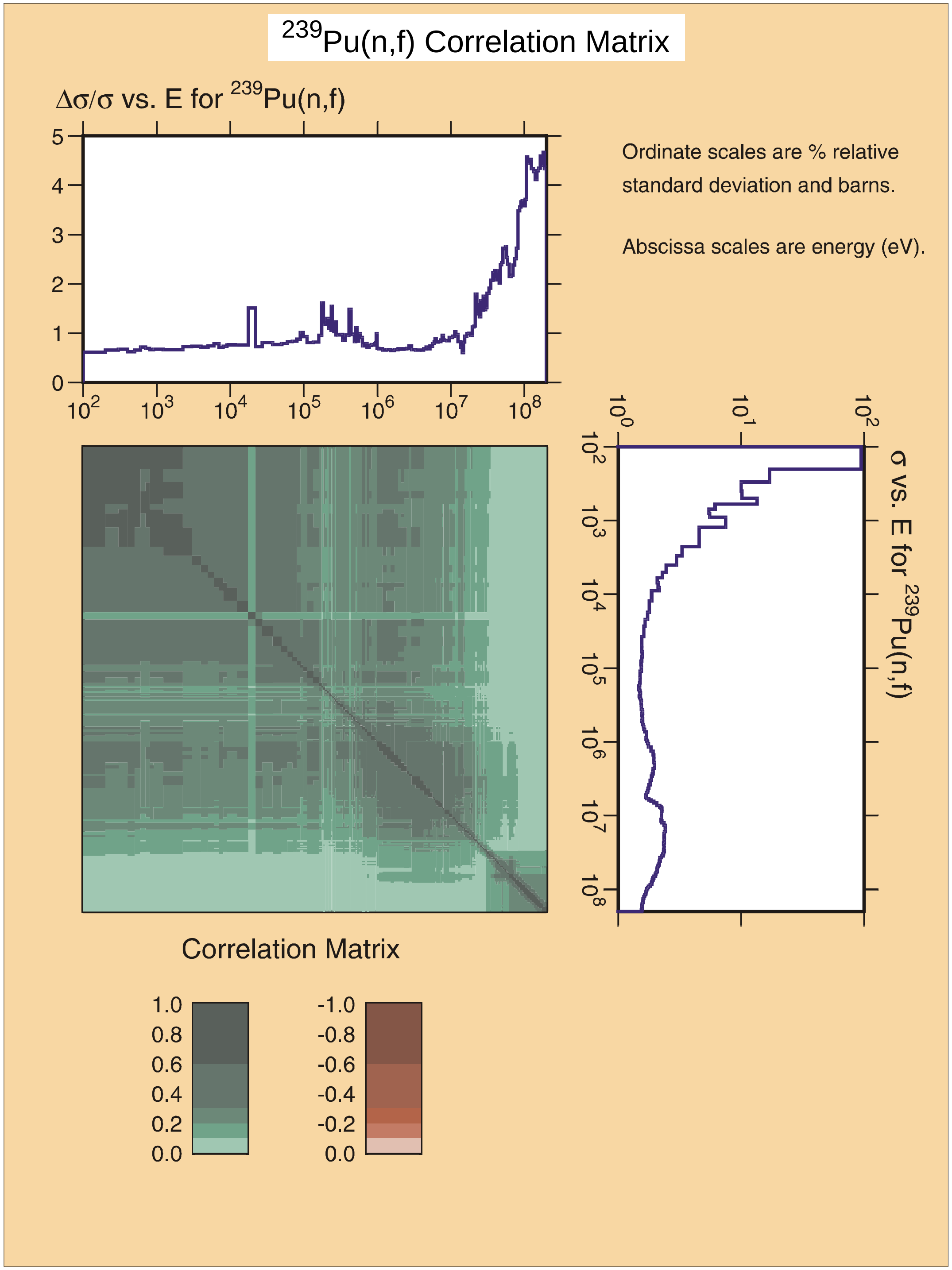


COMPARISON OF THE ENDF/B-VII EVALUATION OF THE ^{239}Pu(n,f) CROSS SECTION UP TO 200 MeV WITH THE ENDF/B-VI STANDARDS RESULTS

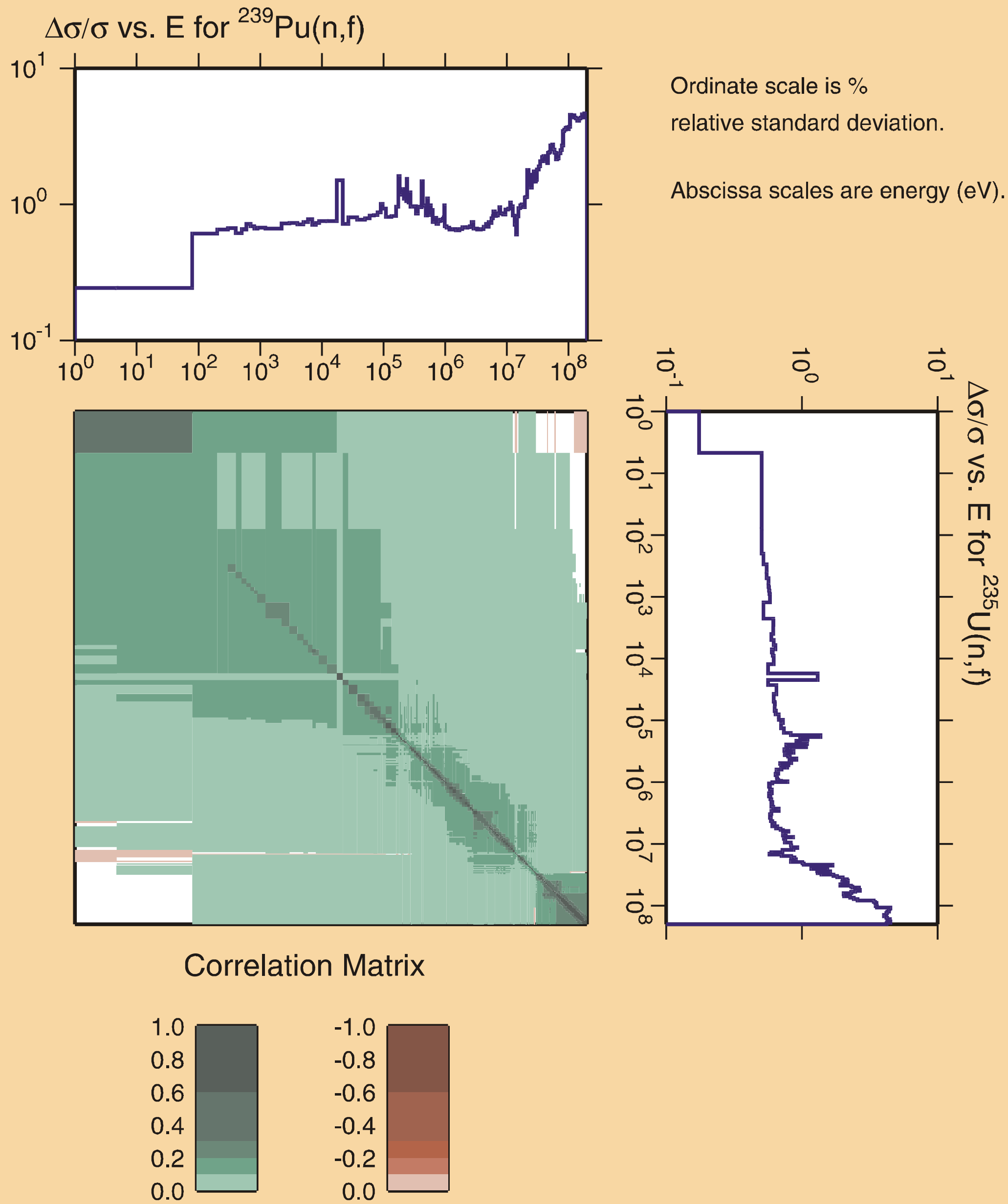


UNCERTAINTY OF THE ENDF/B-VII ^{239}Pu(n,f) CROSS SECTION




239Pu(n,f) Correlation Matrix
Δσ/σ vs. E for 239Pu(n,f)
Ordinate scales are % relative standard deviation and barns.
Abscissa scales are energy (eV).
σ vs. E for 239Pu(n,f)
Correlation Matrix
1.0
0.8
0.6
0.4
0.2
0.0
-1.0
-0.8
-0.6
-0.4
-0.2
0.0

^{239}Pu(n,f) - ^{235}U(n,f) Correlation Matrix



COMMENTS ON THE "LOW" UNCERTAINTIES

•The uncertainties are the average over an interval- not "at" an energy.

•There are a very large number of data sets in the evaluation. Many have small uncertainties. A least squares analysis leads to small uncertainties.

•Normally for a limited size database, unrecognized systematic uncertainties lead to overall uncertainties that are too small and cross sections that are wrong. Our evaluation used a very large database. If one considers the unrecognized systematic uncertainties as normally distributed, their effect should be reduced significantly

•An additional component of uncertainty was added where significant differences for groups of data points were noted. It was noted that this procedure increases the output uncertainties slightly with little effect on the cross sections.

•If correlations among experiments are not taken into account, the uncertainty in an evaluation will be too low. Consider the case of two experiments that are nearly fully correlated. Our database was investigated carefully for correlations such as common detectors.

COMMENTS ON THE "LOW" UNCERTAINTIES (cont.)

•An important result is that it is essential to consider the covariances, not just the variances, in applications of cross sections to practical systems.

- •The use of models in fits leads to the redistribution of the uncertainties between variances and off-diagonal covariances of the uncertainty matrix with a reduction of the variances. As a result, the percent uncertainties are reduced but the uncertainty of the integral quantities sensitive to the evaluated data in a wide energy region is conserved in general.

Conclusions

•The most recent evaluations of the $^{239}Pu(n,f)$ cross section have used the most modern "leading edge" evaluation methods available. New techniques and corrections have been used. Though in some cases the uncertainties seem small, we encourage additional measurements of all the types of data used in the standards evaluation. For each ENDF/B evaluation changes have taken place as a result of new data with, in some cases improved measurement techniques. At the least, new measurement methods may not have the same unknown systematic uncertainties present in the older types of measurements. Also, results that corroborate the present evaluation are valuable.