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Summary Report

Consultants' Meeting on

Review Benchmarking of Nuclear Data for the Th/U Fuel Cycle

IAEA Headquarters
Vienna, Austria

20-22 December 2010

Prepared by

R. Capote Noy
IAEA Nuclear Data Section
Vienna, Austria

February 2011

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Nuclear Data Section
International Atomic Energy Agency
Vienna International Centre
PO Box 100
A-1400 Vienna
Austria

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Abstract

A summary is given of the Consultants' Meeting (CM) on Review and Benchmarking of Nuclear Data for the Th/U Fuel Cycle. An IAEA Coordinated Research Project (CRP) on "Nuclear Data for Th/U Fuel Cycle" was concluded in 2005. The CRP activities resulted in new evaluated nuclear data files for ^{232}Th, ^{231,233}Pa (later adopted for the ENDF/B-VII.0 library) and improvements to existing evaluations for ^{232,233,234,236}U. Available nuclear data evaluations for $^{230\text{-}232}$Th, ^{231,233}Pa and ^{232,233,234}U were reviewed including ROSFOND2010, CENDL-3.1, JENDL-4, JEFF-3.1.1, MINSKACT, and ENDF/B-VII.0 libraries. Benchmark results of available evaluations for ^{232}Th and ^{233}U were also discussed. Technical discussions and identified deficiencies are summarized; corrective actions are proposed where deemed necessary. Participants' summary reports presented at the CM are also included in this report.

February 2011

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1. Background

The thorium-based nuclear fuel cycle offers many advantages for future energy production including much smaller build-up of long-lived higher actinides which are the main source of long-term residual radioactivity in nuclear waste, and the fact that world reserves of thorium are much larger than uranium reserves. Furthermore, it is well established that thorium fuel is more proliferation-resistant. The above advantages have resulted in a rising interest in innovative fuel cycle concepts based on thorium.

Numerous activities are in progress in many countries that anticipate the use of thorium-based fuel for accelerator-driven systems applicable to power production and radioactive waste transmutation. Active design efforts are focused on an advanced heavy-water reactor concept in India that uses thorium fuel. The project "Nuclear Data for Th/U Fuel Cycle" was initiated as an IAEA Coordinated Research Project (CRP) in 2002. The main goal of the CRP was the improvement of evaluated nuclear data files that will allow more accurate design calculations of innovative fuel cycle concepts involving the Th/U fuel cycle. The CRP activities resulted in new evaluated nuclear data files for ^{232}Th, ^{231,233}Pa (later adopted for the ENDF/B-VII.0 library) and improvements to existing evaluations for ^{232,233,234,236}U. Basic evaluated nuclear data files as well as the processed libraries in ACE format for the MCNP Monte Carlo code and MATXS format for deterministic codes are available from the IAEA at http://www-nds.iaea.org/Th-U/.

Since the completion of the CRP in 2005, a comprehensive technical document describing the work done was published[1]. Some experience with the use of the data was accumulated and new libraries were released (e.g. JENDL-4.0, ROSFOND-2010). Therefore, it was time to review the differences and new evaluations, and check the overall performance of the files.

The objective of the meeting was to review the status of the evaluated data files, in particular the identified deficiencies and to provide recommendations for actions to further improve the data. Cross sections in the fast neutron energy range, resonance range and of the prompt fission spectra and nu-bar are specifically addressed.

2. Presentations and Discussions

2.1. A. Trkov

A. Trkov presented the uncertainties of integral parameters (such as thermal cross section, resonance integral and ^{252}Cf spectrum-averaged cross section) derived from the covariance information in the files. In the discussion that followed, the uncertainty in the capture cross section averaged over the ^{252}Cf spectrum was considered too high. The reason for this is the large spread of experimental data above 200 keV, which would require a careful review and renormalisation to allow reducing the assigned systematic uncertainty which, in turn, leads to a reduction of the overall uncertainty.

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[1] R. Capote, L. Leal, Liu P., Liu T., P. Schillebeeckx, M. Sin, I. Sirakov and A. Trkov, *Evaluated Nuclear Data For Nuclides Within The Thorium-Uranium Fuel Cycle*, IAEA Technical Report STI/PUB/1435, International Atomic Energy Agency Vienna, 2010.

Available online at http://www-nds.iaea.org/reports-new/tecdocs/sti-pub-1435.pdf.

There was no change in the criticality benchmark results except that two new benchmarks were identified. The ICSBEP benchmark labelled U233-COMP-THERM-004 was presented at the recent ICSBEP meeting. The benchmark model given at the time was found to be deficient. B. Briggs was contacted and he provided the final write-up, including the results with an improved model. The conclusion is that the reactivity is underpredicted by slightly more than 500 pcm.

2.2. S. Kahler

S. Kahler presented benchmark results performed at LANL. With reference to ^{232}Th and ^{233}U systems, this analysis included the ICSBEP benchmarks labelled U233-MET-FAST-004 which contain Th measurements with W reflector and IPPE-FR-MULT-RRR-001 reaction rate ratio measurements from IPPE. He also mentioned results from reaction rate measurements in LANL critical assemblies. He pointed out some systematic discrepancies in Be and polyethylene reflected systems, which were more pronounced in cases with harder spectra. Some of the discrepancies could be caused by ^{232}Th data, but they could just as well result from other effects (e.g. the shape of the fission spectrum) or other material data.

2.3. V. Maslov

V. Maslov made a presentation on prompt-fission neutron spectra. He claimed that for ^{235}U the underestimated spectrum contribution at the low-energy tail is compensated by a bump in the spectrum at higher energies, and hence good agreement attained in the reactivity prediction of criticality. The same is not true for ^{239}Pu and ^{233}U nuclei, therefore larger discrepancies are to be expected. Some indications of such behaviour could indeed be observed, but it was argued in the discussion that they could also be caused by other deficiencies in the data.

V. Maslov made another presentation on the use of surrogate measurements of the fission cross section in nuclear data evaluation. In some cases the surrogate measurements proved very useful, especially when they were supported by direct measurements, but in other cases they led to large discrepancies in the data.

2.4. A. Ignatyuk

A. Ignatyuk presented the intercomparison of the uncertainties in different reactions for the nuclides of interest. In many cases the uncertainties calculated from the covariances were reasonable, but some problematic cases were indicated, e.g. the capture cross section of ^{232}Th above 0.2 keV, which leads to the 12 % uncertainty in the ^{252}Cf spectrum-averaged cross section. Trkov argued that the scattering of experimental data in this energy range is very large, so large systematic uncertainties were assigned to the data in doing the analysis. Capote pointed out that it might be possible to justify renormalisation of some experimental data sets, thus reducing the systematic uncertainties and letting the correlations from the prior (from model calculations) propagate the low uncertainty at energies below 0.2 MeV to higher energies. Also, the uncertainty in the elastic cross section in the unresolved resonance region of about 2 % was considered too small. Trkov said that an ad-hoc correction could easily be implemented.

2.5. R. Capote

R. Capote presented benchmarks results for JENDL-4 supplied by O. Iwamoto as shown in Fig. 1 below. Results for some ^{235}U, ^{239}Pu and ^{233}U fuelled systems are relevant for current review.

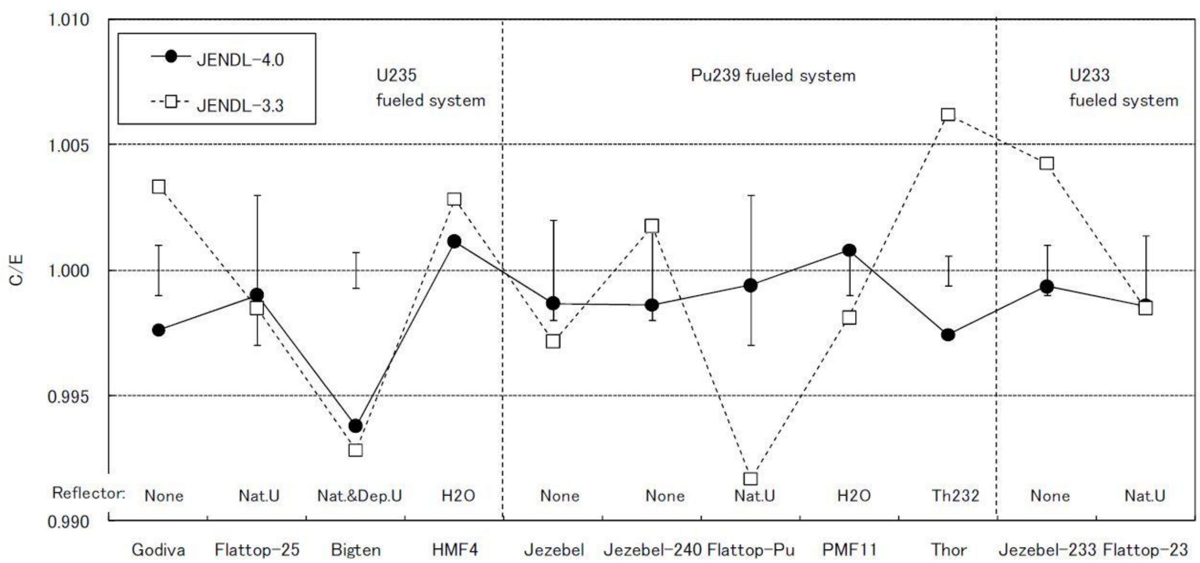

Fig. 1.

3. Overview of Evaluations and Benchmarks

After the formal presentations the discussion continued, focusing on individual nuclides. The fission, capture and (n,2n) cross sections from different sources were compared to experimental data. Where indicated, the inelastic cross sections were also compared. An overview of the discussion about individual nuclei is given below.

3.1. Cross Section Overview

<u>Th-229 cross sections</u>

The resonance parameters in JENDL-4 seem reasonable. The fission cross section measurements of Kobayashi (2001) are discrepant with the old data of Gokhberd (1959). The latter might require renormalisation, in which case the fission cross sections in JENDL-4 above 10 keV should be increased. All things considered, the JENDL-4 evaluation seems to be the best.

Th-230 cross sections

The (n,f) cross sections in JENDL-4 are the best (considering the available experimental data), although at energies above 7 MeV improvement is possible, based on surrogate ratio data, which coincide with direct measurements. The (n,2n) cross section in JENDL-4 is also reasonable. Overall the JENDL-4 evaluation seems to be the best.

Th-231 cross sections

Only the JENDL-4 evaluation is available. Two sets of surrogate measurements for the (n,f) cross section are contradictory. JENDL-4 evaluators seem to follow the Jaunes data closely.

Pa-231 cross sections

The old bomb-shot data of Muir 1971 require renormalisation by a factor of about 0.5. The very recent Oberstedt 2005 data were measured relative to ^{237}Np, but are significantly lower than the data by Plattard and other authors, most of whom were doing measurements relative to ^{239}Pu. The most recent evaluation of the ^{237}Np fission cross section does not help in reducing this discrepancy. In the IAEA evaluation (adopted in ENDF/B-VII.0), the Oberstedt 2005 data (being the most recent) were trusted, partly because the Plattard data show an unreasonable rise above 10 MeV. Most other evaluations are significantly higher. JENDL-4 and MINSK evaluations for the fission cross section are in good agreement below 10 MeV. It is possible that preference for the Oberstedt 2005 data in the IAEA evaluation is not justifiable, in which case the JENDL-4 evaluation would be preferred. A new measurement at the fission "plateau" would be desirable in order to resolve the discrepancies in the evaluated data files.

Differences in the (n,2n) cross sections in different evaluations reflect differences in the fission cross sections.

The capture cross sections below 1 MeV in all evaluations look very similar.

Overall the JENDL-4 evaluation seems to be the most reasonable; the shape of the fission cross section is the basis for this preference.

Pa-232 cross sections

The JENDL-4 fission cross sections in the resolved resonance range seems to follow experimental data best. The (n,2n) cross sections in JENDL-4 look reasonable, and definitely better than in the ENDF/B-VII.0 evaluation.

Overall, the JENDL-4 evaluation seems to be the most reasonable for this nuclide.

Pa-233 cross sections

From the comparison of evaluated and measured data it was evident that the surrogate ratio measurements of the fission cross section are inconsistent with the direct measurements. The JEFF-3.1 evaluation is too high and the older JENDL-3.3 is unreasonable. New surrogate measurements in the energy range 12–17 MeV have been carried out and published by Nayak *et al.*[2], but are not yet compiled in EXFOR. The conclusion was that, in practically all evaluated data files, the fission cross sections below 10 MeV are inconsistent, but the reliability of experimental data should also be verified.

[2] Nayak, B.K., Saxena, A., *et al.*, Phys. Rev. C **78** (2008) 061602.

Above 10 MeV all evaluations are severely discrepant due to different model parameters used in the calculations, but the lack of experimental data makes it impossible to determine which one is more correct. In order to resolve the discrepancies in the evaluated data files, a new measurement at 14 MeV would be desirable.
The (n,2n) cross section in the Minsk evaluation is considerably lower than in the other evaluated nuclear data files. This is mainly due to the differences in the fission cross section. The ENDF/B-VII.0 and JENDL-4 libraries contain more resonances and are expected to provide a better representation of the capture and fission reactions at lower energies. Overall the ENDF/B-VII.0 and JENDL-4 are of similar quality and seem to be reasonable.

<u>U-232 cross sections</u>
The cross section curve for the fission reaction above the resolved resonance range in JENDL-4 follows experimental data, but the (n,2n) cross section is likely to be too high (possibly by a factor of four). The fission cross section at 14 MeV measured by Fursov is questionable. The CENDL-3 library is the only one that accounts for this data point. The original publication was provided by the IAEA, but no reliable conclusion could be drawn. Large differences exist between the evaluations above 8 MeV.
The (n,2n) cross section in JENDL-4 is very high compared to other evaluations. Since the high-energy region is less important for reactor applications, the details in the shape of the cross sections above the resonance range in JENDL-4 are considered an advantage in favour of this evaluation.

<u>U-233 cross sections</u>
The ORNL resonance evaluation is considered the best available and is adopted in ENDF/B-VII.0 and JENDL-4.
There are new high-resolution fission cross-section measurements from n_TOF that are not reflected in the data files, but they themselves are not sufficient to extend the resonance range to higher energies. Some differences are observed in the (n,2n) cross sections. Above 8 MeV there are differences in the fission cross sections of about 100 mb between different evaluations, related to evaluators' preference for the available experimental data, which are discrepant. This also has a strong influence on the (n,2n) cross section data.
The ENDF/B-VII.0 and JENDL-4 total nu-bar disagree with measured data in EXFOR. They also differ from each other. At the same time there is strong disagreement between inelastic cross sections in both evaluations, but interestingly, both evaluations reproduce well the Jezebel U-233 benchmark (pointing to compensation effects).
The total inelastic cross section in ENDF/B-VII.0 has a very unusual shape below 1 MeV, which is very different from JENDL-4 and other evaluations. From a physical point of view, the behaviour of ^{233}U inelastic cross section is expected to be very similar to the ^{235}U inelastic cross section, both being fissile odd-neutron nuclei. Evaluated inelastic cross sections for ^{235}U are very similar, which is not the case for ^{233}U as shown in Fig. 2 below.

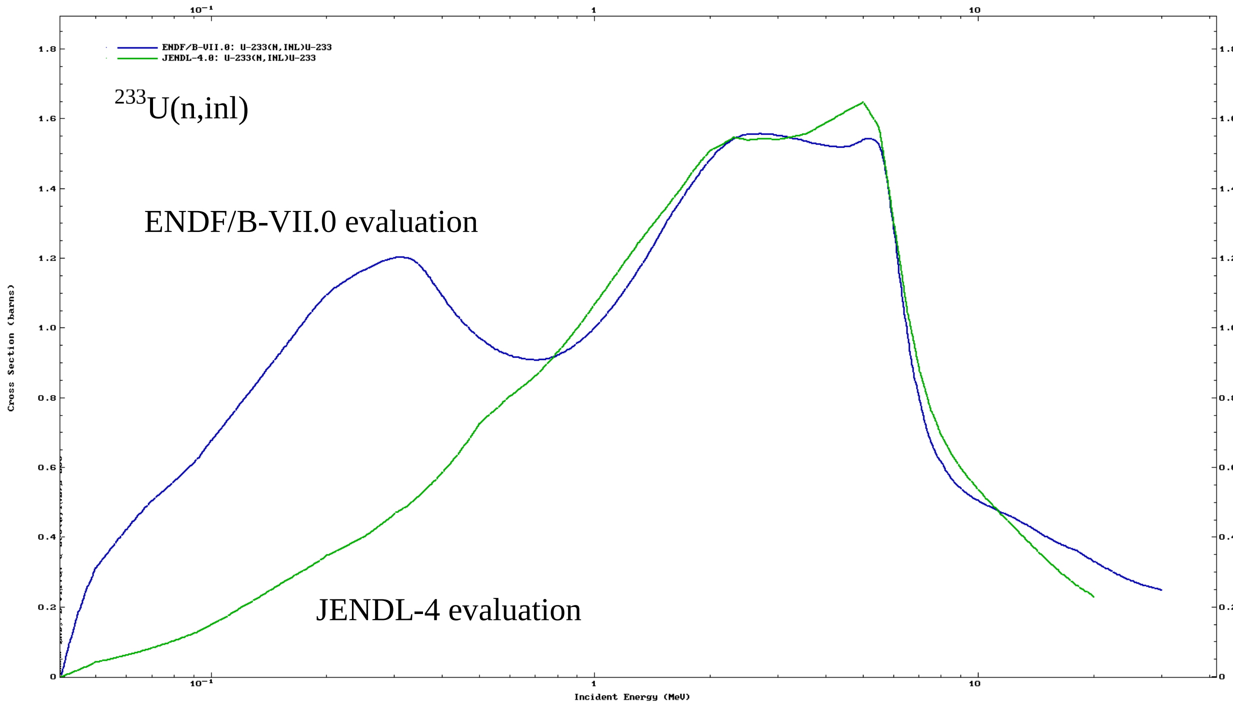

Fig. 2.

ENDF/B-VII.0 evaluations of uranium isotopes and ^{239}Pu were undertaken at Los Alamos National Laboratory by P.G. Young and collaborators[3]. The ^{233}U evaluation was comprehensively documented in the technical report LA-UR-03-1617[4]; the next paragraph is quoted from the original publication:

»The optical model calculations were performed with the 1996 version of the ECIS coupled-channels optical model code by Raynal. All calculations include coupling of the ground state rotational band of ^{233}U. For the angular distribution comparison given below and for determining the angular distributions in the evaluation, we coupled the lowest 7 rotational states into the calculations and included competition from some 33 other uncoupled ^{233}U states plus a continuum of (n,n′) states. The ^{233}U levels are given in Table 2. These calculations were somewhat time consuming, typically taking roughly 15 minutes per incident energy. For the cross section comparisons given below, which require calculations at a large number of energies, we only coupled the 3 lowest-lying rotational states but included 36 uncoupled states and a continuum. This approximation of only coupling 3 states also was used to generate the neutron transmission coefficients for the reaction theory calculations, described below.»

At the meeting it was agreed that R. Capote would perform some calculations to cross-

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[3] Young, P.G, Chadwick, M.B., MacFarlane, R.E., Talou, P., Kawano, T., Madland, D.G., Wilson, W.B., Wilkerson, C.W., *Evaluation of Neutron Reactions for ENDF/B-VII: $^{232-241}$U and ^{239}Pu*, Nuclear Data Sheets **108** (2007) 2589-2654.

[4] Young, P.G, Chadwick, M.B., MacFarlane, R.E., Talou, P., Los Alamos technical report LA-UR-03-1617 (2003).

check the sensitivity to the nuclear model parameters of inelastic scattering on ^{233}U and check different optical model potentials. These calculations were carried out just after the meeting and results are summarized in Fig. 3 below: the total cross section calculated by Young *et al*. potential (RIPL 2010) with 3 and 7 coupled levels, respectively, is compared with the total cross sections calculated by Capote *et al*. optical model potential (RIPL 2408). A big difference is obtained between total cross section calculated using the same Young *et al.* potential when using 3 or 7 coupled levels. Such differences raise questions about the consistency of the use of only three coupled levels for the calculation of transmission coefficients as discussed in LA-UR-03-1617[4]. Further research is warranted. Young has subsequently re-analyzed this isotope, paying particular attention to the low energy inelastic cross section. This latest analysis reduces the large σ_{inel} bump below 1 MeV. LANL plans to perform additional benchmark calculations with this revised file as part of its on-going data testing activities in support of the planned release of ENDF/B-VII.1 at the end of 2011.

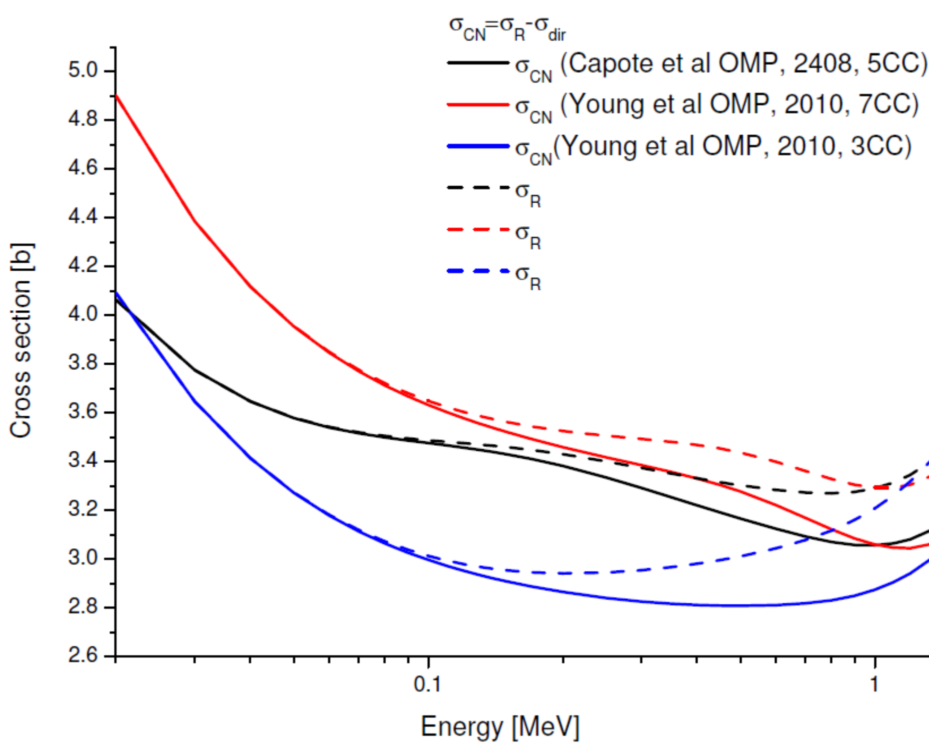


Fig. 3.

<u>Th-232 cross sections</u>

There is a consensus that the ORNL parameters are the best.

The uncertainties in the resonance range in the JENDL-4 file were evaluated independently. The uncertainties in the capture seem too large.

The fission cross section in the plateau region (i.e. below the second chance fission) in the JENDL-4 file is about 8 % higher than in other evaluations. This is partly due to the fact that JENDL-4 evaluators included n_TOF measurements which were not available at the time of the IAEA evaluation (adopted for ENDF/B-VII.0) and are higher than other experimental data.

There are differences in the (n,2n) cross sections at energies above 10 MeV, arising mainly from the discrepancies in the experimental data. In the ENDF/B-VII evaluation (adopted from the IAEA) the Filatenkov data were renormalized (reduced by about 10%), because they were discrepant from other measurements. The JENDL-4 evaluation follows the unnormalised data.

The inelastic cross section data in all evaluations are in good agreement.

3.2. Covariances and Uncertainties Overview

U-233 uncertainties

There is an unreasonably large uncertainty in the quoted total cross section above the resonance range of the candidate evaluation for ENDF/B-VII.1, which can be traced to the large uncertainty in the fission cross section (discussed below). Above 100 keV there is good agreement between the data in all evaluations.

The fission cross section uncertainty is reasonable in the resonance range and above 10 keV in all evaluations, but the uncertainty in the ENDF/B-VII evaluation above the resonance range is very large and exceeds 30 %. Indeed, there are strong fluctuations in the measured data because of the high resolution, but the broad-bin average uncertainty should not be so large.

The evaluated values of the nu-bar uncertainties agree quite well, but differ from the available experimental data by more than the assigned uncertainty. A 0.5 % uncertainty is perhaps too optimistic. It seems that the nu-bar value was adjusted to improve the criticality prediction.

The uncertainty of the capture cross sections seems reasonable in all evaluations.

The uncertainties of the inelastic cross sections look reasonable, but the cross sections in the file differ by a factor of four, particularly the ENDF/B-VII evaluation (see comments on the ^{233}U cross sections in Section 3.1).

Th-232 uncertainties

Some differences in the estimated uncertainties of the total cross sections are observed in the evaluated data files, but they are generally reasonable.

The fission cross section in JENDL-4 is larger than in the other evaluations due to the recent n_TOF data, which are discrepant from other measurements. This ambiguity is reflected in a higher uncertainty assigned to the fission cross sections by the JENDL-4 evaluators. The uncertainty of about 2 % near the second plateau in the ENDF/B-VII.0 evaluation seems over-optimistic.

The nu-bar uncertainty in ENDF/B-VII.0 was adopted from an older BROND evaluation, but the values and the uncertainties are comparable. The uncertainty in the JENDL-4 file is larger by nearly a factor of two, reflecting the spread of the experimental data.

Although JENDL-4 evaluators adopted ORNL resonance parameters, they made a separate evaluation of the uncertainties in the resonance range. Their assigned uncertainties seem too large. From the discussion following the presentation by Ignatyuk it seems that the assigned uncertainty to the capture cross section above the resonance range in ENDF/B-VII.0 is too large. This is supported by the calculated uncertainty of the cross section averaged over the ^{252}Cf spontaneous fission spectrum (presented by Trkov), which is about 12 %, while the discrepancy with the measured value is less than 3 %.

3.3. Benchmarking Overview

It must be emphasized that a major improvement is observed in criticality predictions using the most recent evaluated nuclear data libraries such as JENDL-4 and ENDF/B-VII.0. Most of the criticality benchmarks are predicted within or close to the experimental benchmark uncertainty. There is a slight problem with solution benchmarks of intermediate and thermal systems, which show a decreasing trend with increasing “above-thermal fission fraction” (ATFF). Reaction rate ratios in ^{233}U systems agree well with measurements, but differences are observed in systems containing ^{232}Th. The Grenoble lead-slowing-down experiment is a low-resolution benchmark, which is sensitive to the cross sections in the energy range from 0.1 eV to about 10 keV. Three Th foils of different thicknesses were measured. No major problems with the capture cross sections of ^{232}Th could be identified from the results. For more details please refer to A. Trkov’s summary report in Appendix 3.

ACTIONS

- S. Kahler will re-run the Thor and SB-n benchmarks, separately substituting ^{232}Th and ^{233}U data from JENDL-4, using ENDF/B-VII.0 data for other materials.
- A. Trkov will do a similar exercise for the KBR benchmarks.
- R. Capote will investigate the influence of model parameters on the inelastic cross sections of ^{233}U.
- V. Maslov will draft comments on the influence of the prompt-fission neutron spectra on benchmark calculations

4. Conclusions

An IAEA CRP on “Nuclear Data for Th-U Fuel Cycle” was concluded in 2005, where new evaluations for ^{232}Th and ^{231,233}Pa were produced, and improvements to existing evaluations for ^{232,233,234,236}U made. Former evaluations were later adopted for the ENDF/B-VII.0 library. Since then, new evaluations have become available. Available nuclear data evaluations for $^{230\text{-}232}$Th, ^{231,233}Pa and ^{232,233,234}U were reviewed including ROSFOND2010, CENDL-3.1, JENDL-4, JEFF-3.1.1, MINSKACT, and ENDF/B-VII.0 libraries. Benchmark results for ^{232}Th and ^{233}U nuclei of ENDF/B-VII.0 and JENDL-4 evaluations were also discussed.

Some deficiencies of available evaluations have been identified (see Section 2 for details) and best evaluations recommended.

ANNEX A

IAEA

International Atomic Energy Agency

Consultants' Meeting on

"Review Benchmarking of Nuclear Data for the Th/U Fuel Cycle"

IAEA Headquarters, Vienna, Austria
20-22 December 2010

Meeting Room B0482

Preliminary AGENDA

Monday, 20 December

08:30 - 09:15	**Registration** (IAEA Registration desk, Gate 1)
09:30 - 10:00	**Opening Session**
	Introductory Remarks – Roberto Capote Noy Election of Chairman and Rapporteur Adoption of Agenda
10:00 - 12:30	**Session 1: Presentations by participants**
12:30 – 14:00	***Lunch***
14:00 – 18:00	**Session 2:** • **Presentations by participants (cont'd)** • **Discussion on Th-232 issues** • **Discussion on Pa issues**
19:00	***Dinner at a Restaurant downtown***

Tuesday, 21 December

09:00 - 12:30	**Session 3:** • **Discussion on U-233 issues** • **Discussion on other isotopes**
12:30 – 14:00	***Lunch***
14:00 – 18:00	**Session 4: Recommendations and future actions**

Wednesday, 22 December

09:00 - 17:00	**Session 5: Drafting of report**
	Coffee and lunch break in between
17:00	**Closing of the meeting**

Consultants' Meeting
"Review Benchmarking of Nuclear Data for the Th/U Fuel Cycle"

IAEA Headquarters, Vienna, Austria
20 to 22 December 2010

LIST OF PARTICIPANTS

BELARUS
Vladimir MASLOV
Joint Institute for Power and Nuclear Research-Sosny
99 Academician
A.K. Krasin Str.
Minsk BY-220109
Tel: +375 17 2994441
E-mail: maslov@bas-net.by

RUSSIA
Anatoly IGNATYUK
Institute of Physics and Power Engineering
Bondarenko Sq. 1
249020 Obninsk, Kaluga Region
Tel: +7 484 399 8035
e-mail: ignatyuk@ippe.ru

SLOVENIA
Andrej TRKOV
Jožef Stefan Institute
Java 39
1000 LJUBLJANA
Tel: + 386 1 5885 324
E-mail: andrej.trkov@ijs.si

USA
A.C. (Skip) KAHLER
Los Alamos National Laboratory
Theoretical Division
T-2, Nuclear and Particle Physics, Astrophysics and Cosmology Group
Building 200, Room 215
P.O.Box 1663, MS B214
Los Alamos, NM 87545
Tel. +1-505-606-2042
E-Mail: akahler@lanl.gov

IAEA
Roberto Capote Noy
NAPC Nuclear Data Section
Vienna International Centre
PO Box 100
1400 Vienna
Austria
Tel. +43-1-2600-21713
E-Mail: r.capotenoy@iaea.org

C.1. Improvements to the ^{232}Th evaluation

Andrej Trkov

Jozef Stefan Institute, Java 39, 1000 Ljubljana, Slovenia

Background

The status of the ^{232}Th evaluated nuclear data file is as of June 2010. Compared to the original IAEA evaluation it includes corrections of small errors and minor improvements, mainly to the covariance data to compensate some of the missing uncertainty components. The internal version designation of the current file is "i3c7m".

Scope of File Testing

General testing included:

- Processability with the Pre-Pro codes
- Preparation of 33-group cross-sections and covariances processed with NJOY
- Checking of the uncertainties in the calculated integral parameters such as thermal cross sections, resonance integrals and ^{252}Cf spontaneous fission spectrum averaged cross sections.

Benchmark testing included the following:

- Previously analysed ICSBEP benchmarks
- New ICSBEP benchmark
- Grenoble lead-slowing-down experiment.

Uncertainties in integral parameters

Thermal cross sections

Thermal cross sections are often compared to the values by Mughabghab, but these are really evaluations of information from the literature and not an independent data source. Neutron activation analysis is a widely used technique, for which a database of experimentally measured k_0 and Q_0 constants has been developed and is compiled in the so-called Kayzero database of IUPAC (http://www.iupac.org/web/ins/2001-075-1-500). Assuming that the gamma emission probabilities, isotopic masses and isotopic abundances are known accurately, thermal cross sections can be derived from the k_0 constants. The Q_0 constants are simply the ratios of the resonance integrals and the thermal cross sections. Since the measurements are based on thermal neutron activation, only the capture cross sections can be measured. In Table 1 it is shown that the ^{232}Th thermal capture cross section is in excellent agreement with value derived from the measured k_0 constant in the Kayzero database.

Resonance integrals

By similar arguments as for the thermal capture cross section, the calculated Q_0 value is compared to the measured one in Table 1. Again, excellent agreement is observed.

^{252}Cf *spontaneous fission spectrum averaged cross sections*

The neutron spectrum from spontaneous fission of ^{252}Cf is an accurately determined standard and is well-suited for comparing spectrum-averaged cross-sections σ_{Cf} in certain cases. Unfortunately, the measured data for the spectrum-averaged capture cross section of ^{232}Th are not abundant. The measurement is difficult, because any scattering in the casing and room-return produce low-energy neutrons that perturb the measurement by absorption in the resonances. Special care is needed to minimise the effect and correct for it, as needed.

A single measurement for capture (labelled "absorption") by L. Green is found in EXFOR (entry 10557001). The measurement technique was activation, therefore it is likely that the experimentalists meant the capture cross section, but no further details are given. The uncertainty in the capture cross section in the evaluated data file is large, but the average cross section is in good agreement with the measurement, as seen from Table 1.

Three measurements for the fission cross section exist, one by J. Csikai and Z. Dezso (1985, EXFOR entry 30415001), using ^{27}Al(n,α) and ^{115}In(n,n') monitors to measure the neutron flux, second by J.A. Grundl and D.M. Gilliam (1983, EXFOR entry 12821002), for which no experimental details are given, and third by J.R. Deen and E.L. Draper[1] measured by solid state track detectors. The measurements differ significantly. The "measured" values in Table 1 are the average and "standard deviation" and should be used with caution. Nevertheless, the measured and the calculated values are consistent.

Table 1: Comparison of measured and calculated integral cross sections

Reaction	Measured	Calculated	C/E-1 [%]
σ_0 (n,g)	7.37±0.34 %	7.34 ± 1.5 %	-0.4
Q_0 (n,g)	11.53±3.6 %	11.47 ± 2.4 %	-0.5
σ_{Cf} (n,g)	0.0891±2.4 %	0.0912 ± 12. %	+2.4
σ_{Cf} (n,f)	0.0829±9.1 %	0.0792 ± 2.1 %	-4.5

Benchmark testing

Criticality benchmarks

The changes to the cross sections in the evaluated data file from the older version are small. The previously analysed ICSBEP and other benchmarks are not affected. The list of benchmarks with their ICSBEP designation and number of cases, the short name and a brief description are listed in Table 2. The predicted values of the multiplication factors were compared to the reference benchmark values. The results are shown in Fig. 1. Note that the lattice LWBR SB-2½ contains no Th, therefore a slightly positive bias in the calculated results is not due to the Th data. Some of it

[1] J.R. Deen, E.L. Draper, Jr., Measurement of Fission Product Yields and the Energy Integral Fission Cross Section of Thorium-232 in a Californium-252 Fission-Neutron Spectrum, Nucl.Technol. **25**, 416 (1975).

most likely comes from ^{233}U and some from the “last-minute changes” to the Zr data (compare ENDF/B-VII.b2+ and ENDF/B-VII.0).

A new ICSBEP benchmark U233-COMP-THERM-004 named ETA-II containing Th in the fuel was presented at the last ICSBEP meeting in Ljubljana, 2010. The computational model for MCNP provided by the authors was found to be deficient (for example, it did not use thermal scattering law data; correction of this deficiency resulted in a negative bias in the results of several 100 pcm). Rigorous development of a new model is beyond the scope of the present analysis; calculations with the existing model (but including thermal scattering data) is included in Fig. 1. Since the cross section data did not change, the results of ENDF/B-VII are applicable for the present version of the file.

Table 2: List of ICSBEP criticality benchmarks included in the analysis.

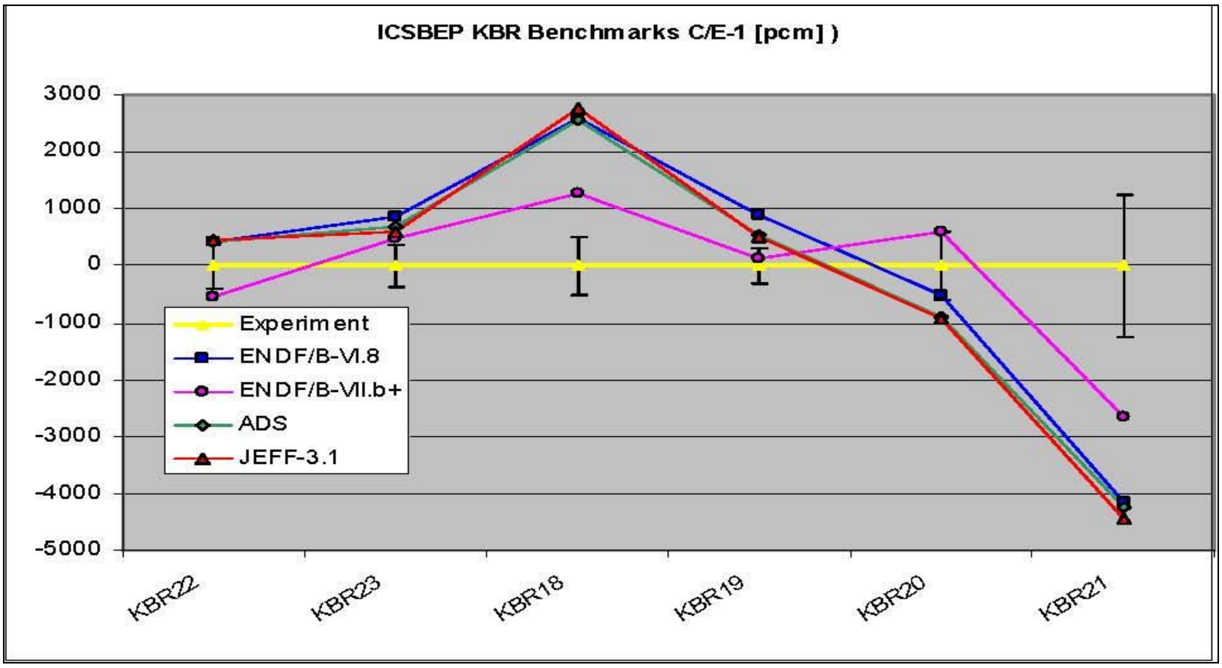
ICSBEP Identifier	Cases	Short name	Description
HEU-MET-FAST-068	1	KBR22	U/Th metal, PE = polyethylene
HEU-MET-INTER-008	1	KBR23	U/Th metal, PE
IEU-COMP-FAST-002	1	KBR18	90%$^{235}UO_2$ + Th metal + 36%$^{235}UO_2$
IEU-COMP-INTER-001	1	KBR19	90%$^{235}UO_2$ + Th metal + 36%$^{235}UO_2$, PE
	1	KBR20	90%$^{235}UO_2$ + Th metal, PE
IEU-COMP-THERM-005	1	KBR21	36%$^{235}UO_2$ + Th metal, PE
PU-MET-FAST-008	1	THOR	Pu sphere/Th-reflector
HEU-COMP-THERM-015	1	LWBR SB-1	93%$^{235}UO_2$ + ZrO_2, ThO_2 blanket
	1	LWBR SB-5	93%$^{235}UO_2$ + ZrO_2, ThO_2 blanket
U233-COMP-THERM-001	1	LWBR SB-2	97%$^{233}UO_2$ + ZrO_2, ThO_2 blanket
	1	LWBR SB-2½	97%$^{233}UO_2$ + ZrO_2, no blanket
	1	LWBR SB-3	97%$^{233}UO_2$ + ZrO_2, UO_2 + ThO_2 blanket
	1	LWBR SB-4	97%$^{233}UO_2$ + ZrO_2, UO_2 + ThO_2 blanket
	1	LWBR SB-6	97%$^{233}UO_2$ + ZrO_2, ThO_2 blanket
	1	LWBR SB-7	97%$^{233}UO_2$ + ZrO_2, UO_2 + ThO_2 blanket
U233-COMP-THERM-004	1	ETA-II	D_2O moderated ^{233}UO2 – $^{232}ThO_2$ lattice (new)
LEU-COMP-THERM-060	10	RBMK	Th absorbers, cases 19-28 (not used)
U233-SOL-THERM-006	1	ORCEF	Th as impurity only, (not used)
U233-SOL-THERM-008	1	ORNL	Th as impurity only, (not used)
U233-SOL-THERM-009	1	ORNL	Th as impurity only, (not used)
U233-SOL-THERM-012	1	ORCEF	Th as impurity only, (not used)
U233-SOL-THERM-013	1	ORCEF	Th as impurity only, (not used)

Grenoble lead-slowing-down experiment

The Grenoble lead-slowing-down experiment is an interesting low-resolution measurement that probes the energy range from about 0.1 eV to approximately 10 keV. At lower energies the uncertainty is large due to low counts. At higher energies the error increases due to increased sensitivity to the lead cross sections.

Three experiments were performed with Th foils of different thickness, namely 175, 1000 and 4000 μm. Radiative capture reaction rates were measured by detecting the prompt-gamma rays from the sample. Normalisation was done to the average reaction rates in the energy range 4 – 10 keV, because at these energies the capture cross section is known rather accurately from two recent independent measurements, one performed at the n_TOF facility at CERN and the other at IRMM in Geel and excellent agreement exists between them.

Comparison between measurements and calculations is shown in Figures 2-4. In Figure 2, two calculations are presented where the data for lead were taken from the ENDF/B-VII and the JENDL-3.2 library, respectively. No essential differences are observed. Within the limits of experimental uncertainties, no obvious deficiencies of the ^{232}Th data are noted.



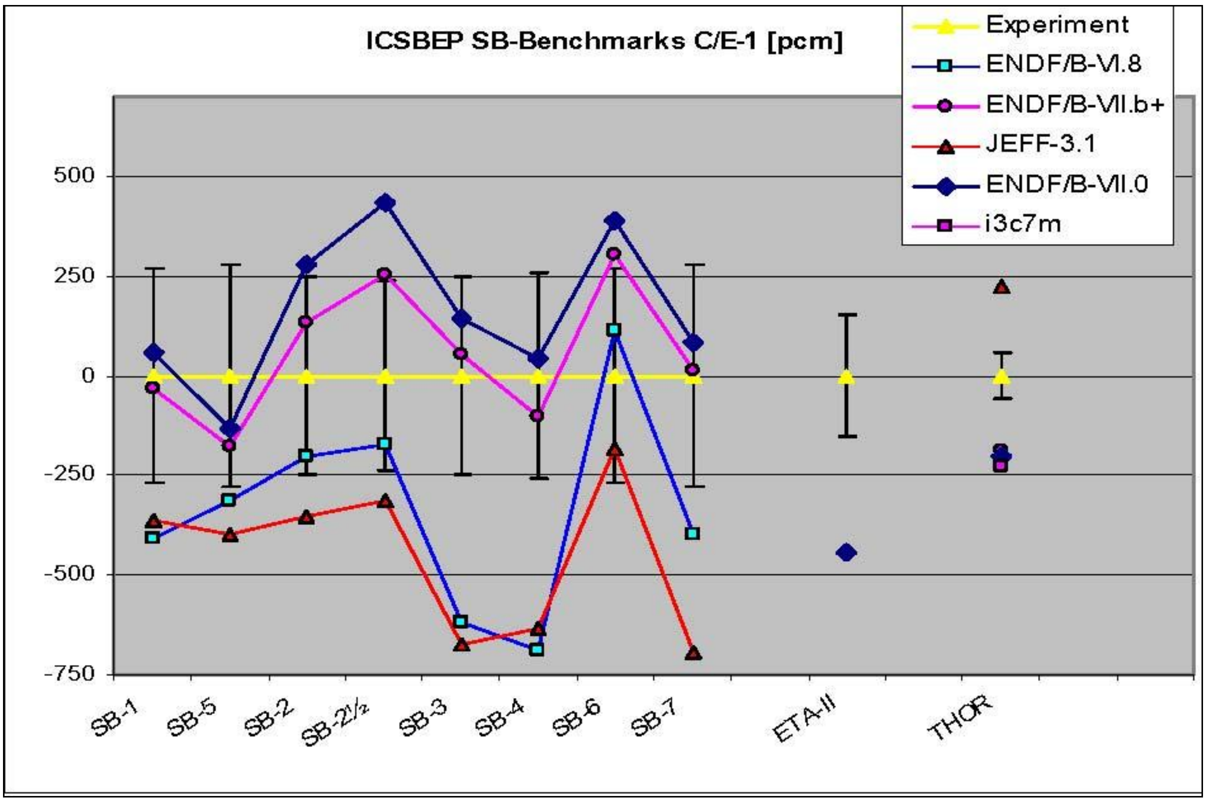


Fig.1. C/E-1 (pcm) for the ICSBEP Criticality benchmarks

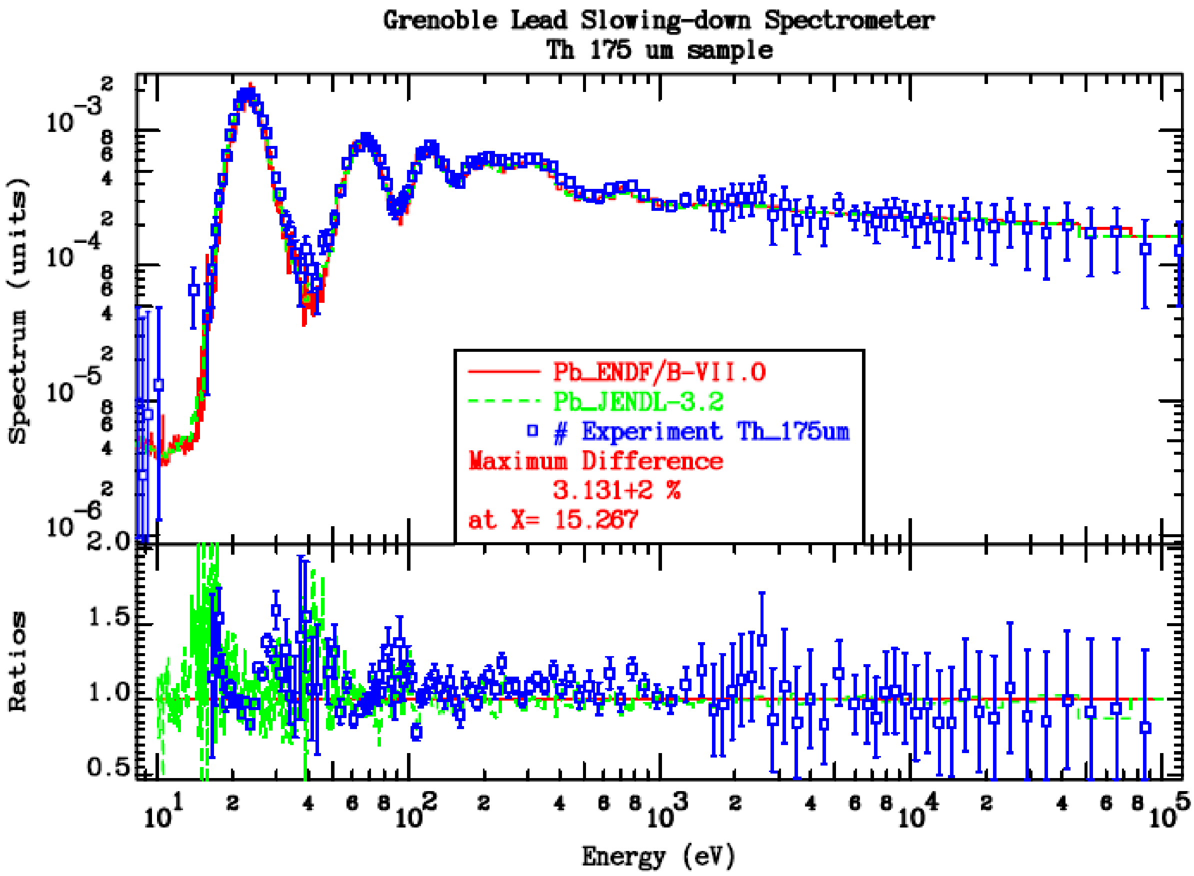


Fig.2. Grenoble lead slowing-down benchmark results.

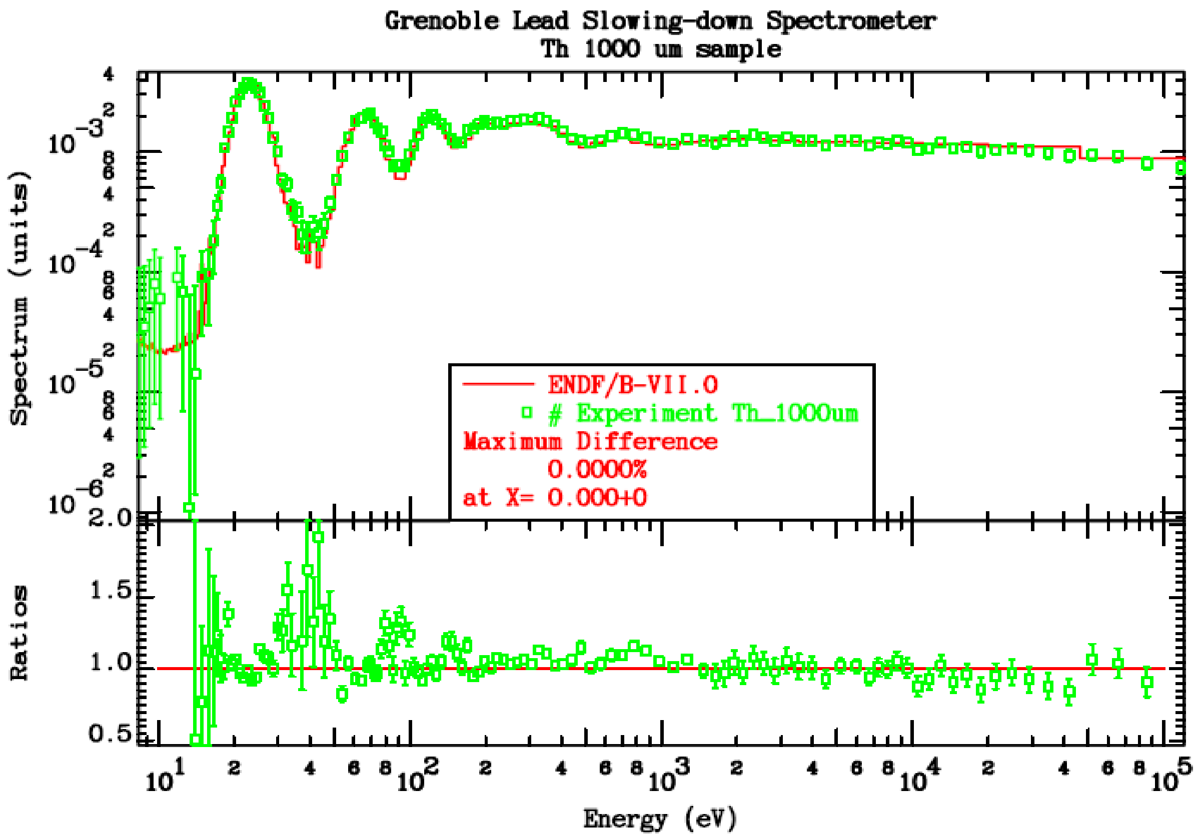


Fig.3. Grenoble lead slowing-down benchmark results.

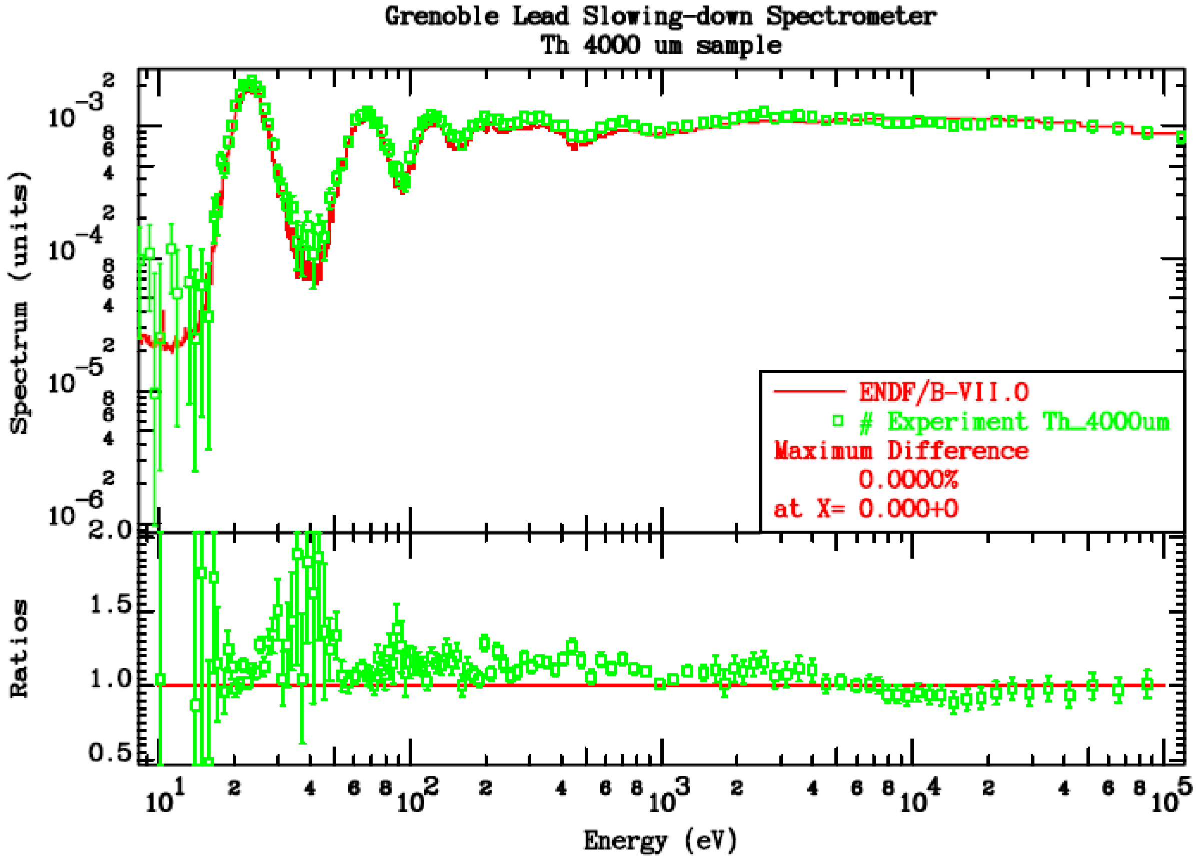


Fig.4. Grenoble lead slowing-down benchmark results.

Conclusions

The new evaluated nuclear data file for ^{232}Th was tested. The file is based on the IAEA evaluation but includes a number of small corrections. The overall performance of the file is not affected, but processability is improved. The file has been processed with NJOY99.336 to produce an ACE library for MCNP, plot cross sections and plot covariances in ECCO-33 and Vitamin-J 175-group structure. The validation package includes:

- The corrected evaluated nuclear data file for Th-232.
- ACE library file for MCNP.
- Series of plots comparing ENDF/B-VII.0 and the new corrected file.
- Series of plots of all cross sections and double-differential data generated with the ACER module of NJOY.
- Series of plots of covariances in the ECCO 33-group structure.

Acknowledgement

The additional work on ^{232}Th evaluation was supported by the National Nuclear Data Center, Brookhaven National Laboratory.

C.2. Benchmark Eigenvalue Calculations of ^{233}U and/or ^{232}Th Critical Systems

A. C. (Skip) Kahler
Los Alamos National Laboratory
Theoretical Division
T-2: Nuclear and Particle Physics, Astrophysics and Cosmology Group

Abstract. This report summarizes the results of MCNP continuous energy Monte Carlo eigenvalue calculations performed for a variety of ^{233}U and ^{232}Th bearing benchmarks. Calculated eigenvalues, using ENDF/B-VII.0 cross sections, are generally accurate for unmoderated systems. For intermediate and thermal spectrum systems a decreasing trend in calculated eigenvalue is observed with increasing Above-Thermal Fission Fraction. This summary report was prepared as a contribution to an IAEA sponsored Consultants meeting to "Review Benchmarking of Nuclear Data for the Th/U Fuel Cycle".

Eigenvalue calculations have been performed for a variety of International Criticality Safety Benchmark Evaluation Project (ICSBEP) critical benchmarks containing ^{233}U and/or ^{232}Th. These calculations were performed with the MCNP continuous energy Monte Carlo code and include unmoderated and either water or beryllium moderated systems. Most of these calculations use ENDF/B-VII.0 cross sections, but in selected instances JENDL-4.0 cross sections for ^{90,91,92,94,96}Zr, ^{232}Th and/or ^{233}U have been used in conjunction with ENDF/B-VII.0.

Table 1 shows the calculated eigenvalues for a suite of U233-MET-FAST (UMF) benchmarks. Among these, UMF1 and UMF6 are historical LANL experiments known as Jezebel-23 and Flattop-23. One other benchmark in this Table, designated Pu-MET-FAST-008, or PMF8, is the historical LANL "Thor" experiment; an unmoderated, spherical Pu core surrounded with Thorium.

Table 1: Calculated Eigenvalues for Fast ^{233}U and ^{232}Th Benchmarks

Benchmark Name	Benchmark Eigenvalue	ENDF/B-VII.0	Comment
UMF1	1.000(1)	0.99964(4)	Unreflected (^{233}U Jezebel)
UMF2	1.0000(10) 1.0000(11)	0.99907(4) 1.00050(4)	^{233}U core, 1.2 cm or 2.0 cm thick HEU reflector
UMF3	1.000(1) 1.000(1)	0.99450(4) 1.00016(4)	Same core as UMF2, 2.3 cm or 5.3 cm thick ^{nat}U reflector
UMF4	1.0000(7) 1.0000(8)	1.00459(4) 1.00500(4)	Same core as UMF2, 2.4 cm or 5.8 cm thick ^{nat}W reflector
UMF5	1.0000(30) 1.0000(30)	0.99427(4) 0.99248(4)	Same core as UMF2, 2.0 cm or 4.2 cm thick Be metal reflector
UMF6	1.0000(14)	0.99928(4)	^{233}U core, 19.9 cm thick ^{nat}U reflector (Flattop-23)
PMF8	1.0000(6)	0.99800(3)	Spherical Pu core, cylindrical Th reflector (Thor)
Note: All benchmarks are described in the International Handbook of Evaluated Criticality Safety Benchamrk Experiments. This handbook is maintained by the International Criticality Safety Benchmark Evaluation Project.			

The calculated eigenvalues for either bare ^{233}U or ^{233}U reflected by uranium are generally very accurate (case 1 of UMF3 being a notable exception). The tungsten reflected assemblies calculate high, a known deficiency that is expected to be remedied with revised isotopic tungsten evaluations scheduled to appear in ENDF/B-VII.1 later in 2011. The beryllium reflected assemblies calculate low, a result also seen to an even greater degree in thermal and intermediate spectrum solution assemblies. Further study and resolution of this issue is beyond the scope of this report. The thorium reflected benchmark's, PMF8/Thor, calculated eigenvalue is somewhat low, particularly compared to the reported experimental uncertainty. Additional comments on this benchmark are provided below, where calculated eigenvalues with either ENDF/B-VII.0 or JENDL-4.0 ^{232}Th are discussed.

Figure 1 shows the calculated eigenvalues for a variety of thermal and intermediate spectrum ^{233}U solution systems. Inclusion of bare, water reflected, polyethylene reflected and beryllium reflected systems provides a suite of benchmarks that span virtually the entire Above-Thermal Fission Fraction (ATFF) space.

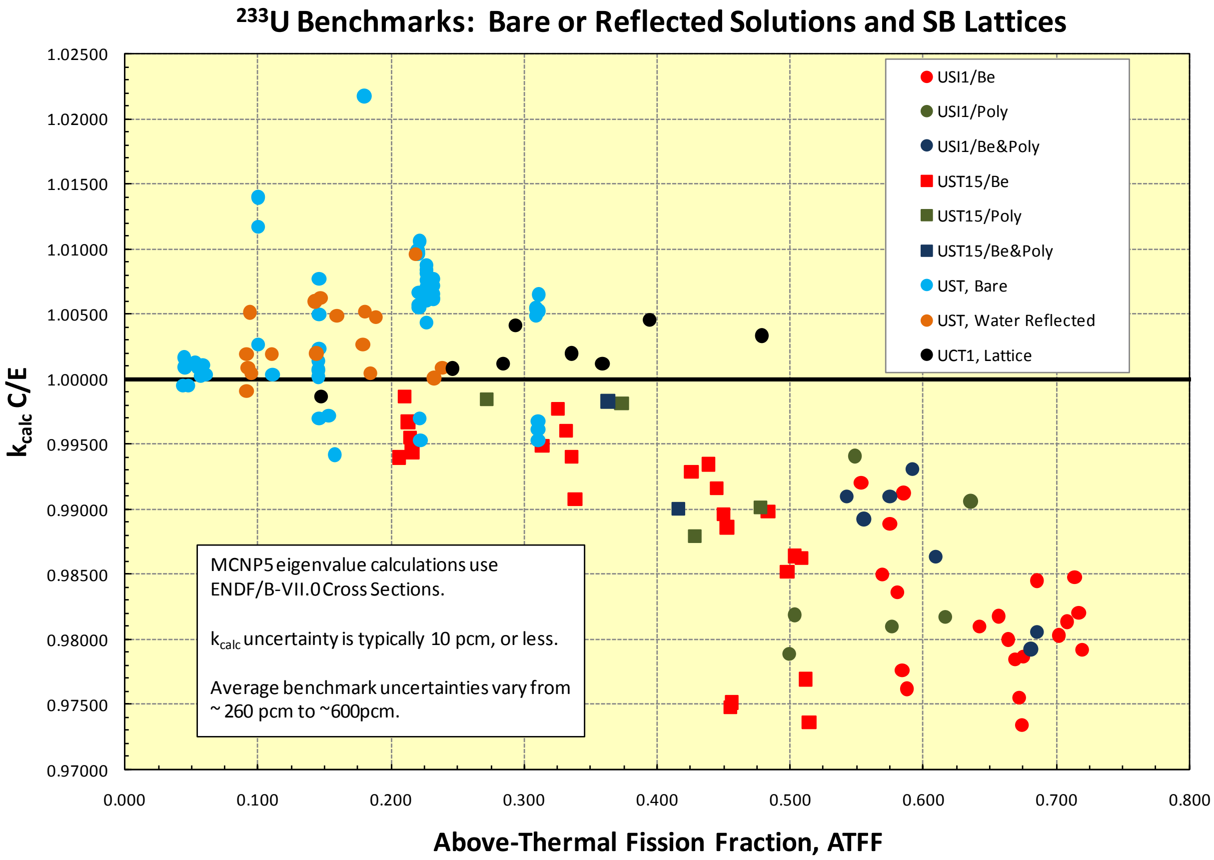


Fig. 1: Calculated Eigenvalues for Thermal and Intermediate Spectrum ^{233}U Benchmarks

Also included in this suite (black circles) is the U233-COMP-THERM-001 benchmark; a set of lattice experiments performed in support of the United States' Light Water Breeder Reactor (LWBR) program. It is likely that the neutronic properties of a ^{233}U fueled commercial reactor would be similar to those of this benchmark. Although there is a wide range of calculated eigenvalues for these benchmarks, spanning a nearly 4.5% range with an obvious decreasing calculated eigenvalue versus increasing ATFF trend, the lattice (UCT1) ATFF values are such that the calculated eigenvalues are fortuitously close to unity.

The Japanese nuclear data community has recently released the latest generation of their evaluated nuclear data file, JENDL-4.0. Additional eigenvalue calculations for the UCT1 and PMF8/Thor benchmarks have been using a mix of ENDF/B-VII.0 and one or more of $^{90,91,92,94,96}Zr$, ^{232}Th and ^{233}U JENDL-4.0 cross sections are tabulated below.

Table 2: Calculated Eigenvalues for Fast Spectrum ^{233}U and ^{232}Th Benchmarks

Benchmark Name	Benchmark Eigenvalue	ENDF/B-VII.0	JENDL-4.0 ^{232}Th plus ENDF/B-VII.0	JENDL-4.0 ^{233}U plus ENDF/B-VII.0	JENDL-4.0 ^{iso}Zr plus ENDF/B-VII.0	JENDL-4.0 ^{iso}Zr, ^{232}Th & ^{233}U plus ENDF/B-VII.0
PMF8 (Thor)	1.0000(6)	0.99800(9)	0.99906(9)	---	---	---
UCT1.1_SB1	1.0006(27)	1.00183(9)	1.00199(10)	---	0.99971(9)	0.99969(9)
UCT1.2_SB2	1.0015(25)	1.00486(10)	1.00497(10)	1.00127(11)	1.00233(10)	0.99909(11)
UCT1.3_SB25	1.0000(24)	1.00459(10)	---	1.00060(10)	1.00279(10)	0.99915(10)
UCT1.4_SB3	1.0007(25)	1.00269(8)	1.00338(8)	0.99905(8)	1.00101(8)	0.99779(8)
UCT1.5_SB4	1.0015(26)	1.00233(8)	1.00291(8)	1.00130(8)	1.00061(8)	0.99980(8)
UCT1.6_SB5	1.0015(28)	1.00017(9)	1.00032(9)	---	0.99840(9)	0.99859(9)
UCT1.7_SB6	0.9995(27)	1.00367(10)	1.00420(10)	0.99937(10)	1.00158(10)	0.99768(10)
UCT1.8_SB7	1.0004(28)	1.00162(9)	1.00281(9)	0.99758(9)	0.99941(9)	0.99655(9)
UCT4	1.0017(18)	0.99853(8)	0.99931(8)	0.99761(8)	0.99923(9)	0.99881(8)

Pu-MET-FAST-008 (PMF8, also known as “Thor”) is a Pu core reflected by thorium. The ENDF/B-VII.0 calculated eigenvalue is 200 pcm low, a relatively large bias given the claimed 60 pcm experimental uncertainty. Use of JENDL-4.0 ^{232}Th cross sections reduces this bias to just under 100 pcm. Since unreflected Pu assembly calculated eigenvalues with ENDF/B-VII.0 $^{239,240}Pu$ cross sections are accurate, this suggests that the JENDL-4.0 ^{232}Th cross sections are better suited for fast system eigenvalue calculations.

Calculated eigenvalues for the U233-COMP-THERM-001 (UCT1) benchmark with various cross section data sets are also shown in Table 2. The various configurations defined in the UCT1 benchmark represent a set of critical experiments performed at the Bettis Laboratory in the US in support of the Light Water Breeder Reactor Program. This benchmark consists of water moderated and reflected fuel rods set on a hexagonal pitch. The rods contained various combinations of $^{233}UO_2$-ZrO_2, $^{235}UO_2$-ZrO_2, $^{233}UO_2$-$^{232}ThO_2$ or $^{232}ThO_2$. UCT4 was also performed at Bettis, with $^{233}UO_2$-$^{232}ThO_2$ fuel rods in a D_2O moderating environment plus $^{235}U(1.3)O_2$ driver rods in an H_2O moderating and reflecting environment. There is much less Zr in this benchmark, appearing only in the rod clad and end-plugs.

UCT1 benchmark eigenvalues were calculated with the MCNP continuous energy Monte Carlo program, using the cross sections identified in the various Table columns. The calculated eigenvalues with ENDF/B-VII.0 cross sections are all larger than the defined benchmark eigenvalue. While each is within 2σ of the expected eigenvalue, having all 8 values be too large, by an average of ~200 pcm, indicates a bias in some aspect of the modeling or cross sections. The calculations were repeated, substituting either ^{232}Th, ^{233}U or $^{90,91,92,94,96}Zr$ (designated ^{iso}Zr in the Column label) from JENDL-4.0 while retaining ENDF/B-VII.0 cross sections for the remaining nuclides.

Continuing with UCT1 results, differences in ENDF/B-VII.0 and JENDL-4.0 ^{232}Th are minimal, as the average calculated eigenvalue increased by less than 50 pcm after switching from ENDF/B-VII.0 to JENDL-4.0. Hence, in contrast to the fast region nuclear data, it appears that the thermal region ENDF/B-VII.0 and JENDL-4.0 ^{232}Th cross sections are similar. However, the impact of changing from ENDF/B-VII.0 ^{233}U or ENDF/B-VII.0 ^{90,91,92,94,96}Zr to the corresponding JENDL-4.0 data is much more significant. For the six UCT1 benchmarks containing ^{233}U the ENDF/B-VII.0 calculated eigenvalue was ~210 pcm too large whereas after switching to JENDL-4.0 ^{233}U the calculated eigenvalue is ~75 pcm low. Similarly, the impact of using JENDL-4.0 ^{90,91,92,94,96}Zr is to reduce the calculated eigenvalue bias of ~200 pcm to ~0 pcm. Finally, we show the impact of simultaneously changing ^{90,91,92,94,96}Zr, ^{232}Th and ^{233}U from ENDF/B-VII.0 to JENDL-4.0. The individual affects noted above are nearly additive, as the ~200 pcm ENDF/B-VII.0 calculated eigenvalue bias becomes ~-220 pcm after these substitutions. Unfortunately, these results are not sufficiently comprehensive to recommend one data set over another, as it appears the use of ENDF/B-VII.0 plus either JENDL-4.0 ^{233}U or JENDL-4.0 ^{90,91,92,94,96}Zr yields more accurate calculated eigenvalues than pure ENDF/B-VII.0, but the combination of both produces an overcorrection. Further studies of other Zr bearing benchmarks absent ^{233}U are warranted, but are beyond the scope of the current effort.

Calculated eigenvalues for the UCT4 benchmark are biased low for all cross section combinations. The ENDF/B-VII.0 to JENDL-4.0 calculated eigenvalue change for ^{233}U is similar to that observed in the UCT1 benchmark, with a decrease of ~100pcm. Since the already low ENDF/B-VII.0 based calculated eigenvalue gets worse with JENDL-4.0, one might argue that the more accurate ^{233}U file for criticality benchmark calculations comes from ENDF/B-VII.0, but drawing this conclusion on such limited data is questionable and a larger suite of ^{233}U bearing benchmarks must be calculated to confirm such an assertion. Other potential deficiencies in the ENDF/B-VII.0 ^{233}U file, particularly with respect to inelastic scattering cross sections below 1 MeV, were identified during this Consultants meeting. LANL (P. Young) has recently developed a new ^{233}U file with a more realistic low energy inelastic scattering cross section profile. This file will be tested later this year and if so judged may be a candidate for inclusion in the ENDF/B-VII.1 library scheduled for release at the end of calendar year 2011.

C.3. Update of ^{233}U, $^{229-232}$Th and $^{230-233}$Pa fission data

V. M. Maslov

Joint Institute for Nuclear and Energy Research, 220109, Minsk-Sosny, Belarus

1. Background

The influence of the ^{235}U(n,f) and ^{239}Pu(n,f) prompt fission neutron spectra (PFNS) on modeling of integral benchmarks was estimated in [1-3] to be significant. For ^{233}U(n,f) PFNS similar sensitivities could be envisaged. For the variety of Th/U fuels and systems, ranging from metal fast to deep thermal solutions, large positive/negative swings in calculated K_{eff} can be expected. Th/U fuelled core criticality calculations would be sensitive to the modelled soft tail of fission neutrons or deficiency of hard tail fission neutrons, as revealed in [1-3] for U and Pu fuels. The deficiency of the ^{233}U(n_{th},f) PFNS, adopted for the ENDF/B-VII.0 (Fig. 1) could be traced back to the "propagation" of calculated ^{235}U(n_{th},f) PFNS shape at E_n= 0.5 MeV [1]. At higher energies, average energies of PFNS coincide only at ~5 MeV, at other E_n the average energies and spectra shapes are drastically different (Fig. 2). However, it might be argued that the response of the criticality benchmark calculations for the ^{233}U thermal and fast systems would be similar to that observed for the PFNS of ^{239}Pu, as demonstrated in [2, 3]. Our approach allowed to solve the longstanding problem of inconsistency of ^{235}U integral data testing and differential prompt fission neutron spectra data [1], a similar approach may hold for ^{233}U. Using modified PFNS, we may avoid arbitrary tweaking of neutron cross sections or neutron multiplicities for Th/U fuel-cycle related nuclides (^{233}U, ^{229,230,231,232}Th and ^{230,231,232,233}Pa) to compensate the ill-defined shape of ^{233}U PFNS [5]. For metal fast benchmarks much would depend on the inelastic scattering cross section of ^{233}U, which should be considered rather uncertain at the moment [4]. The unrealistic evaluations of poorly investigated cross sections related to the Th/U fuel cycle could be excluded by consistent analysis of the available fission data base.

The evaluation of ^{229,230,231,232}Th(n,f) and ^{230,231,232,233}Pa(n,f) cross sections could be supplemented by description of surrogate and ratio surrogate fission data, coming from transfer reactions. These are pick-up, stripping or inelastic scattering reactions of charged-particle beams with stable actinide targets, which tremendously enhance the scarce neutron-induced fission cross section data base for the Th-Pa chain of nuclei. Consistent description of non-fissile ^{230,232}Th(n,f) neutron-induced and surrogate fission data allows to quantify the consistency of surrogate, ratio surrogate and neutron-induced fission data. That, in turn, would allow to predict the fission cross sections in keV-energy range of fissile ^{229,231}Th target nuclides. A similar approach in case of ^{231,233}Pa(n,f) reactions with a less diverse data base, allows prediction of fissile targets cross sections of ^{230,232}Pa. Unfortunately, even in the newest data library of JENDL-4.0 [5] the consistency of the fissilities for chain of nuclides is not observed, adverse effect on (n, γ) and (n, xn) prediction is unavoidable. In clear-cut cases like ^{232}Th(n,f) and ^{230}Th(n,f), the fission chances partitioning was done in a simplified manner, the relative contributions of (n,f) and (n,xnf) reactions are defined as a phenomenological, non-Hauser-Feshbach, fit to the observed fission cross sections. In the less clear-cut cases of ^{231}Pa(n,f) and ^{233}Pa(n,f), the fission chances contribution prediction is not correlated with the fissilities of $^{A+1-x}$Pa nuclides, emerging in (n,xnf) reactions.

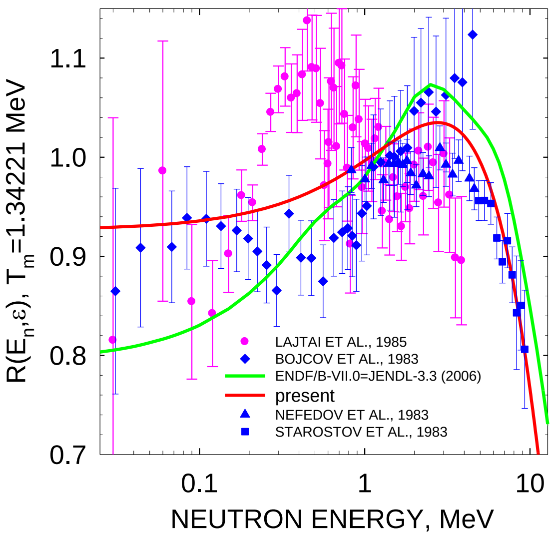


Fig. 1. Prompt fission neutron spectra ^{233}U.

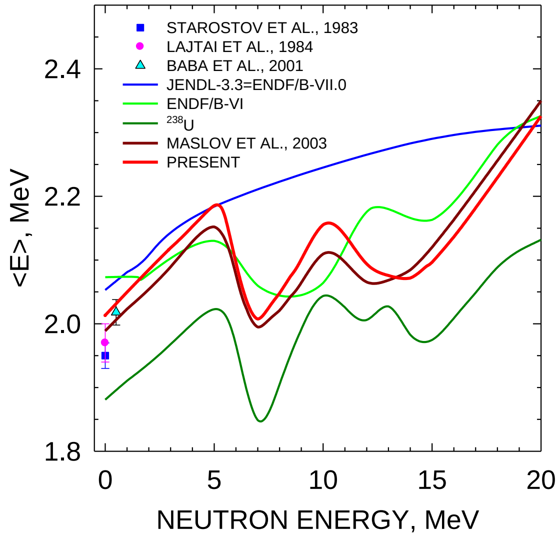


Fig. 2. Average energy of ^{233}U fission neutron spectra

2. ^{232,230}Th(n,f) and ^{231,229}Th(n,f) reactions

First chance fission cross sections trends should be based on consistent description of ^{232}Th(n,f), ^{232}Th(n,2n) [6] and ^{238}U(n,f), ^{238}U(n,xn) data [7, 8], unless and until that is observed, (n,γ) and (n,xn) cross sections should be considered tweaked/tuned. The first positive experience with the theoretical approach in a wide excitation energy range was for the inelastic scattering reactions [6, 7], followed by fission: ^{238}U(α,α'f) and ^{236}U(α,α'f) by Burke *et al.* [9, 10]. These ratio surrogate fission data, which are generally consistent with surrogate data by Britt et al. [11], renormalized by Younes and Britt [12, 13], supported the ^{237}U(n,f) cross section, predicted by Maslov [8] for incident neutron energies up to 20 MeV. The prediction was based on the consistent description of ^{238}U(n,f), ^{238}U(n,xn) cross sections and ^{238}U(n,f) prompt fission neutron spectra in the emissive fission domain [13].

At excitations near fission threshold, surrogate data are model-dependent via assumed neutron-absorption cross sections and different angular momentum spectra of excited and fissioning states in neutron-induced and transfer fission reactions. At excitations higher than emissive fission threshold, the sensitivity of the surrogate data to the angular momentum spectra of the nuclide fissioning in the (n,nf) reaction may again increase. Pre-equilibrium effects which are pronounced in (n,f) reactions, might be another source of possible discrepancies. The recently developed surrogate ratio method largely removes the uncertainty imposed by pre-equilibrium effects and different angular momentum spectra of excited and fissioning states in neutron-induced and transfer reactions. The structure of the fission barrier and the angular moments of the entrance channel states for even-even fissioning nuclides were shown to be significant. The excitation/position of the $K^\pi = 2^+$ abnormal rotational band-head at the inner saddle of even-even fissioning nuclide like ^{238}U are important. For ^{232}Th (n,f) reaction the excitation of $K^\pi = 0^-$ states in the ^{232}Th(n,nf) reaction depends on the mass-asymmetry of the even nuclide ^{232}Th outer saddle shape. It defines the cross section in the 6-10 MeV incident neutron energy range. That, in fact, explains the broad peak in the observed fission cross section (Fig. 3). The partitioning of (n,f) reaction into (n,xnf) contributions differs widely from those of JENDL-4.0. In the latter case the shapes of ^{232}Th(n,f) and ^{232}Th(n,nf) contribution variations at $E_n \geq 10$ MeV are difficult to justify.

Similarly, modeling in case of the ^{230}Th(n,f) reaction produces a similar pattern of first and higher fission chances contributions, again those of JENDL-4.0 are quite different (Fig. 4). The surrogate data for the ^{230}Th(n,f) reaction were inferred using a pick-up reaction ^{232}Th(^{3}He, ^{4}He) by Petit *et al.* [15].

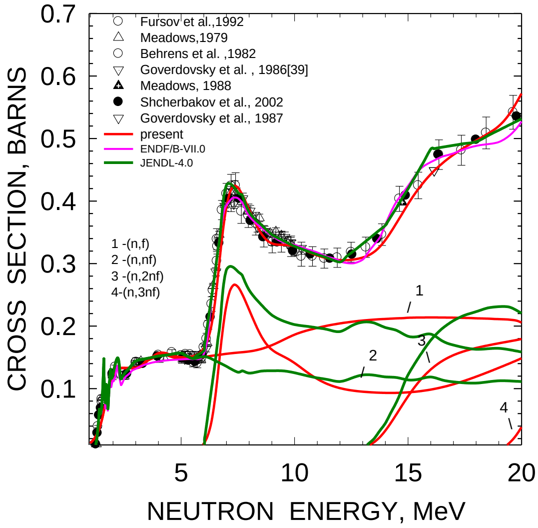


Fig. 3. Fission cross section of ^{232}Th(n,f).

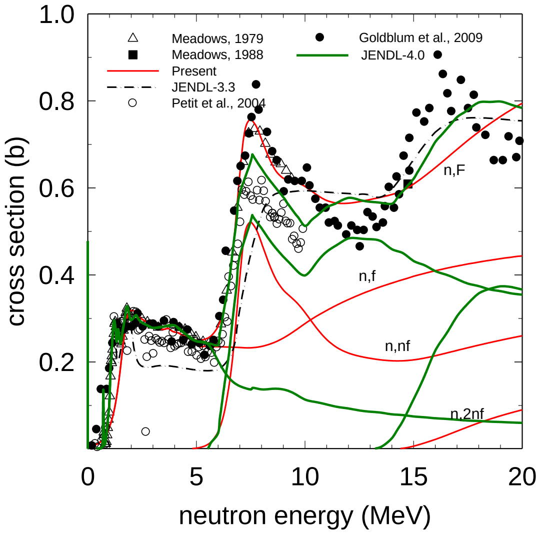


Fig. 4. Fission cross section of ^{230}Th(n,f).

The ratio surrogate data for the ^{230}Th(n,f) reaction are inferred from the ratio of fission probabilities of ^{231}Th and ^{235}U nuclides by Goldblum *et al.* [16]. The desired compound nuclei were populated using the same pick-up reactions ^{232}Th(^{3}He, ^{4}He) and ^{236}U(^{3}He, ^{4}He). With the opening of the ^{230}Th(n,2n) channel, the surrogate data [14] are strongly discrepant from both (n,f) and ratio surrogate data [16] (Fig. 4) which means that in the emissive fission domain the fission cross section cannot be obtained multiplying the measured fission probability (surrogate data) by the neutron absorption cross section. That is a failure of the “factorization theorem” in the emissive fission domain.

Analysis of ^{232}Th(n,f) reaction cross section allows to infer ^{231}Th(n,f) cross section (Fig. 5) [6, 17]. In a similar way, for medium excitations of 10-20 MeV of ^{231}Th nuclide, when pre-fission neutron emission is possible in the reaction ^{232}Th(^{3}He,^{4}He), the ^{229}Th(n,f) cross section could be inferred. Excitation of $K^{\pi}=0^{-}$ states depends on the mass-asymmetry of the even nuclide ^{230}Th saddle shapes. The shape of the surrogate data around E_n~10 MeV resembles that of the present calculated cross section, specifically, the step-like shape at E_n~10 MeV is attributed to the excitation of the two-quasi-particle states in ^{230}Th fissioning nuclide. At $E_n <$ 10 MeV, the ^{230}Th(n,f) cross section is defined by the collective levels lying within the pairing gap of ^{230}Th. The partitioning of ^{230}Th (n,f) reaction into (n,xnf) contributions based on Hauser-Feshbach statistical model is again much different from those of JENDL-4.0 (Fig.4). In the latter the shapes of ^{230}Th(n,f) and ^{230}Th(n,nf) contribution variations at $E_n \geq 10$ MeV are difficult to justify.

Fission barrier structure and transition states ordering of even-even nuclides ^{232}Th and ^{230}Th strongly influence the energy dependence of ^{231}Th(n,f) and ^{229}Th(n,f) cross sections. The present fission cross section and that of JENDL-4.0 for ^{231}Th(n,f) reaction are shown in Fig. 5. The surrogate data for the ^{231}Th(n,f) reaction are inferred from the ratio of fission probabilities of ^{232}Th and ^{236}U nuclides. The desired compound nuclei are populated using inelastic scattering reactions ^{232}Th(^{3}He, ^{3}He’) and ^{236}U(^{3}He, ^{3}He’) by Goldblum *et al.* [15] which are, in fact, ratio surrogate data. The even nuclide ^{232}Th mass-asymmetric saddle shape is probed in first-chance fission domain in [15]. The strong discrepancy of ^{232}Th(t,pf) surrogate data [11-13] with ratio surrogate data [15] has not been explained so far [17, 18]. The p-wave neutron contribution is very sensitive to the position of the $K^{\pi} = 0^{-}$ at the higher outer saddles. It defines the step-like shape of the ^{231}Th(n,f) fission cross section around 200 keV, supported by surrogate data [11-13]. Nothing of the kind is predicted in the evaluation of JENDL-4.0 [5].

A similar cross-section shape is predicted for the ^{229}Th(n,f) reaction cross section. Again, nothing of the kind is predicted in the JENDL-4.0 [5] data library. Our calculation, which is consistent with the neutron-induced fission data by Kobayashi *et al.* [19], is much different from that

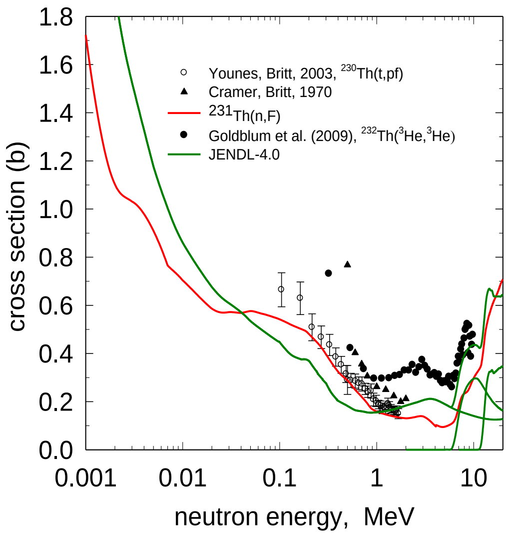


Fig. 5. Fission cross section of ^{231}Th(n,f).

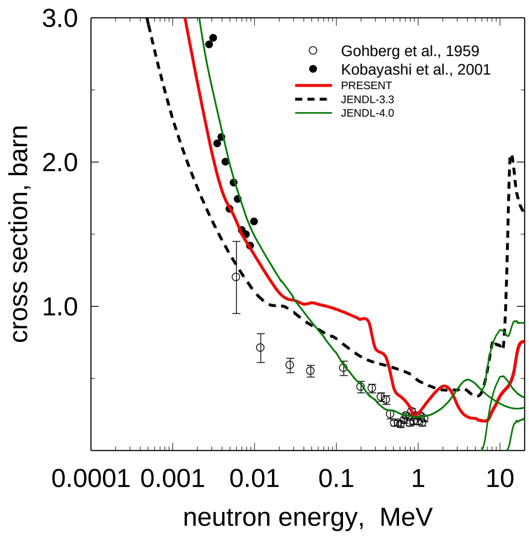


Fig. 6. Fission cross section of ^{229}Th(n,f).

of JENDL-4.0 [1] at higher energies. The shape of the ^{229}Th(n,f) reaction cross section, which could be inferred from data by Gohberg *et al.* [20] supports our prediction at $E_n < 1$ MeV. The diverse set of surrogate, ratio surrogate data for the ^{230,232}Th(n, F) and ^{229,231}Th(n,f) reactions allows to consider their consistency with the data of neutron interactions with respective target nuclides. Measurements support the predicted sharing of the (n,f) cross sections into (n,xnf) contributions, specifically, the increasing trend of the first-chance fission cross section for ^{230}Th and ^{237,239}U [11, 18] targets, posing a problem for the ^{231}Th(n,f) fission cross section.

3. ^{233,231}Pa(n,f) and ^{232,230}Pa(n,f) reactions

The theoretical approach employed for the ^{229,230,231,232}Th(n,f) data evaluation may also be used for $^{230\text{-}233}$Pa(n,f) cross section prediction/analysis (Figs. 7-10). The neutron-induced fission database for ^{231}Pa(n,f) and ^{233}Pa(n,f) was enriched by a few data sets [21-23], however they still do not cover the energy range of 0.001-20 MeV. The neutron-induced fission data of ^{231}Pa(n,f) and ^{233}Pa(n,f) might be complemented with surrogate fission data: fission probabilities of ^{231,232,233,234}Pa nuclides, measured in ^{232}Th(^{3}He,d)^{233}Pa, ^{231}Pa(d,p)^{232}Pa and ^{230}Th(^{3}He,d)^{231}Pa at excitation energies 6-11.5 MeV [11-13] and fission probabilities of ^{232}Th(^{3}He,p)^{234}Pa, ^{232}Th(^{3}He,d)^{233}Pa, ^{232}Th(^{3}He,t)^{232}Pa at excitation energies 6-15 MeV [15]. Emissive fission domain data by Petit *et al.* [15], as well as older indirect data by Birgul *et al.* [24] provoke rather "exotic" assumptions about the first and higher fission chances contributions to the observed ^{233}Pa(n,f) and ^{231}Pa(n,f) fission cross sections. Description of these data needs a rather steep decrease of the first-chance fission cross sections and systematically lowered fission probabilities of relevant Pa nuclides [25, 26]. Recent ratio surrogate data on the fission probabilities of ^{232}Th(^{6}Li,^{4}He)^{234}Pa and ^{232}Th(^{6}Li,d)^{236}U by Nayak *et al.* [27], relevant for the E_n=11.5-16.5 MeV, much enhance the measured data base for the ^{233}Pa target nuclide. Another justification for the application of the model, based on consistent description of surrogate, ratio surrogate and neutron-induced fission data, understanding the restrictions of the former, is its general success in case of the ^{229,230,231,232}Th(n,f) reactions. The predicted trend of the ^{231}Pa(n,f) cross section up to E_n=20 MeV, which is similar to that of ^{233}Pa(n,f), is consistent with fissilities of Pa nuclides, stemming from data analysis of ^{232}Th(p,f).

Fission cross section of ^{233}Pa(n,f) measured in [21, 22] between ~1 and ~3 MeV and ~5 and ~8.5 MeV, are rather discrepant with fission data [15], extracted from the transfer reaction ^{232}Th(^{3}He,p)^{234}Pa for E_n=0.5 ~ 10 MeV, surrogate for ^{233}Pa(n,f) reaction (Fig.7). The surrogate data [15] are appreciably higher than direct ^{233}Pa(n,f) data [21-23] both around (n,f) and (n,nf) fission thresholds (Fig. 7). The evaluation of JENDL-4.0 roughly describes data of [21-23] up to (n,nf) threshold and follows surrogate data [15]. It also predicts a steep slope at higher energies up to the ^{233}Pa(n,2nf) reaction threshold. That trend, based on very low fission probability of ^{233}Pa is strongly discrepant with the surrogate data of ^{232}Th(^{3}He,d)^{233}Pa [11] (Fig.8) as described in the present approach. It looks strange, that the evaluated ^{232}Pa(n,f) reaction cross section of JENDL-4.0 is quite compatible with the present evaluation up to 20 MeV, but very discrepant in case of ^{233}Pa(n,f) reaction. Fig. 8 shows, that to follow the surrogate data by Petit *et al.* [15] the fission probability of ^{233}Pa should be much lower, than predicted by surrogate data of [11].

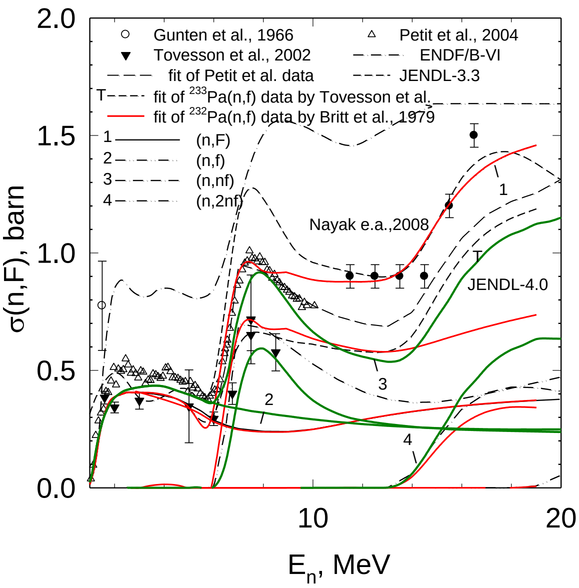


Fig. 7. Fission cross section of ^{233}Pa(n,f)

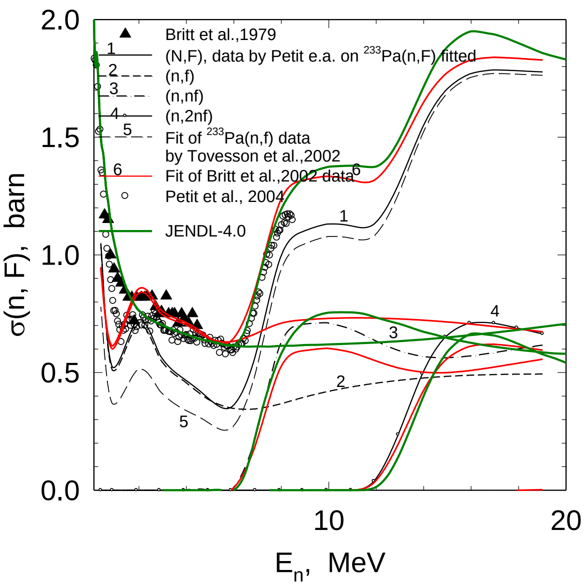


Fig. 8. Fission cross section of ^{232}Pa(n,f)

The present neutron-induced fission cross section of ^{231}Pa(n,f) reaction (Fig. 9) is compatible with measured data by Fursov *et al.* [28] up to E_n~7 MeV, and up to E_n~10 MeV with data by Plattard *et al.* [29]. The data point by Birgul *et al.* [24] at E_n~14 MeV is incompatible with these neutron data trend. The data by Petit *et al.* [15], surrogate for the ^{231}Pa(n,f) reaction, are systematically lower than direct (n,f) data above E_n~7 MeV. To reproduce the data point by Birgul *et al.* [24] contribution of the ^{231}Pa(n,nf) should be much lower than the predicted fission probability for the fissioning nuclide ^{231}Pa. The latter is estimated using fission probability data of ^{230}Th(^{3}He,d)^{231}Pa reaction [11] (Fig. 10). In this case the calculated ^{231}Pa(n,f) cross section would be discrepant with data by Plattard *et al.* [29] at $E_n \geq 7$ MeV and would follow the surrogate data by Petit *et al.* [15]. Fig. 10 compares present calculated cross sections of ^{230}Pa(n,f) with surrogate data [11] and evaluation of JENDL-4.0 [5]. Fig. 10 shows that to follow the data of Birgul *et al.* [24] in Fig. 9, the fission probability of ^{231}Pa should be much lower than predicted by surrogate data of [11].

The predicted trend of the ^{231}Pa(n,f) cross section up to $E_n = 20$ MeV (Fig. 9), which is similar to that of ^{233}Pa(n,f) (Fig. 7), is consistent with fissilities of ^{230,231}Pa nuclides stemming from consistent analysis of ^{232}Th(p,f) and ^{232}Th(p,3n) data analysis [25, 26]. The data base for p+^{232}Th interaction was enlarged by new data on ^{232}Th(p,f) [30] and ^{232}Th(p,3n) [31] for the excitation energy range, corresponding to E_p=15~30 MeV, where the fission probabilities of ^{231}Pa and ^{230}Pa are of major importance.

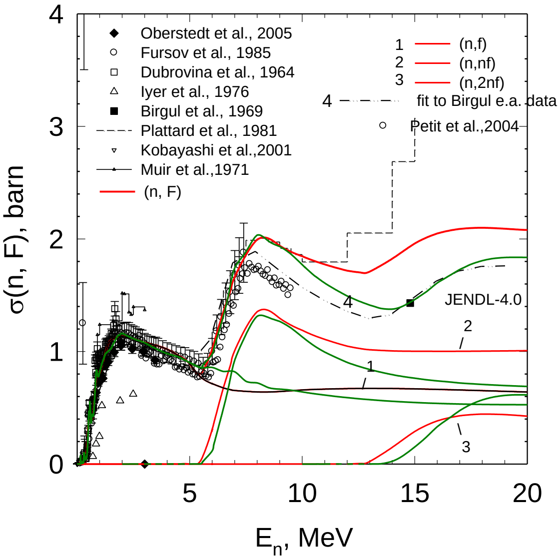


Fig. 9. Fission cross section of ^{231}Pa(n,f).

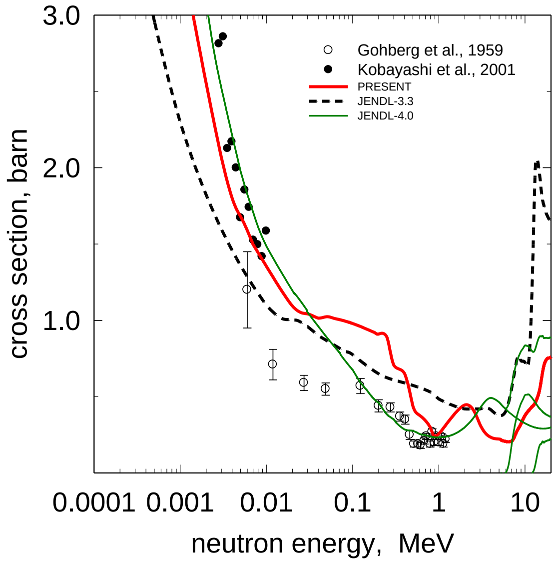


Fig. 10. Fission cross section of ^{230}Pa(n,f).

4. Conclusions

Although the minor discrepancies of the ^{233}U prompt fission neutron spectra (PFNS) in major data libraries are often quoted as a measure of the PFNS uncertainty, these evaluations are strongly discrepant from measured differential PFNS data, lying well outside the biases of different data sets at thermal and fast incident neutron energies [1-3]. The new preliminary evaluation of ^{233}U prompt fission neutron spectrum for thermal neutron energy is more compatible with differential measured data, though some data sets should be assigned larger systematic uncertainty.

It is demonstrated also that the evaluation of ^{229,230,231,232}Th(n,f) and ^{230,231,232,233}Pa(n,f) neutron-induced fission cross sections may be supplemented by description of surrogate and ratio surrogate fission data, coming from transfer reactions. These are pick-up, stripping or inelastic-scattering reactions of charged-particle beams with stable actinides targets, which much enhance the scarce neutron-induced fission cross section data base for Th-Pa chain of nuclei. Consistent description of non-fissile ^{230,232}Th(n,f) neutron-induced and surrogate fission data allows to quantify the consistency of surrogate, ratio surrogate and neutron-induced fission data. That would allow the prediction of the fission cross sections in the keV-energy range of fissile ^{229,231}Th target nuclides. A similar approach in the case of ^{231,233}Pa(n,f) reactions with a less diverse database also allows to predict fissile target cross sections of ^{230,232}Pa. Unfortunately, even in the latest evaluation of JENDL-4.0 [5], the consistency of the fissilities for chain of nuclides is not observed, adverse effect on (n,γ) and (n,xn) prediction would be unavoidable. In clear-cut cases like ^{232}Th(n,f) and ^{230}Th(n,f), the fission chances partitioning was done in a simplified manner in the most recent data library of JENDL-4.0, the relative contributions of (n,f) and (n,xnf) reactions were defined as a phenomenological, non-Hauser-Feshbach, fits to the observed fission cross sections. In the less clear-cut cases of ^{231}Pa(n, F) and ^{233}Pa(n, F) the fission chances contribution prediction is not correlated with the fissilities of $^{A+1-x}$Pa nuclides, emerging in (n, xnf) reactions. These inconsistencies could be avoided in a straightforward application of Hauser-Feshbach statistical theory, once tested in cases of major actinides like ^{232}Th and ^{238}U.

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Nuclear Data Section
International Atomic Energy Agency
Vienna International Centre, P.O. Box 100
A-1400 Vienna
Austria

e-mail: services@iaeand.iaea.org
fax: (43-1) 26007
telephone: (43-1) 2600-21710
Web: http://www-nds.iaea.org