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Evaluation of the neutron induced reactions on ^{235}U from 2.25 keV up to 30 MeV

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Abstract. An evaluation of fast neutron induced reactions on ^{235}U is performed in the 2.25 keV–30 MeV incident energy range with the code EMPIRE–3.2 Malta, combined with selected experimental data. The reaction model includes a dispersive optical model potential (RIPL 2408) that couples seven levels of the ground-state rotational band and a triple-humped fission barrier with absorption in the wells described within the optical model for fission. EGSM nuclear level densities are used in Hauser-Feshbach calculations of the compound-nuclear decay. The starting values for the model parameters are retrieved from the RIPL-3 database. Excellent agreement is achieved with available experimental data for neutron emission, neutron capture and fission, which gives confidence that the quantities for which there is no experimental information are also predicted accurately.

In the fast neutron region of the evaluated file, the fission cross section is taken from Neutron Standards, and neutron capture includes fluctuations observed in recent experiments. Other channels are taken directly from model calculations. New evaluation is validated against ICSBEP criticality benchmarks with fast neutron spectra with excellent results.

1. Introduction

The Nuclear Energy Agency of the OECD started a new international collaboration called CIELO (Collaborative International Evaluated Library Organisation) with the main goal to improve our understanding of neutron reactions on key isotopes that are important in nuclear applications [1–3]. A central role of this project is taken by ^{235}U, which is the major nuclear fuel in energy applications.

While existing ^{235}U evaluations (e.g., ENDF/B-VII.1 [4], JEFF-3.2 [5], and JENDL-4.0 [6]) perform very well for many applications, several discrepancies have been highlighted between ^{235}U integral and differential data (e.g., for prompt fission neutron spectra of thermal ^{235}U(n,f) [7,8]), or between evaluated data from different libraries (e.g., between ^{235}U inelastic cross sections [9]). Such challenges could only be solved by a new and comprehensive evaluation of all neutron induced reactions on ^{235}U. This is the subject of the current contribution.

2. Evaluation methodology

The methodology considers available differential data as the primary constraint, supplemented by integral data from benchmark calculations to provide guidance on preferences in the case of discrepant data, or to pin-down quantities for which no experimental information is available and are not constrained otherwise by the theoretical models. The aim is to constrain model parameters, which are then used to calculate final cross sections. The internal physical consistency of evaluated data is, therefore, preserved. No least-square fitting of integral benchmarks is undertaken, as we do not aim at producing an adjusted library.

An iterative procedure involving differential data calculated with the advanced reaction models implemented in the nuclear modular system EMPIRE-3.2 [10,11], and modelling of integral benchmarks such as the collection from the Handbook of Criticality Safety Benchmark Experiments [12] was followed to perform the present evaluation. Processing with the NJOY2012.50 code system [13] was done to prepare the corresponding ACE libraries for use with the MCNP Monte Carlo transport code [14] in benchmark calculations.

Differences in integral benchmark performance depending on nuclear data were used to improve the reaction models and to fine-tune the reaction model parameters iteratively. Following this procedure, accurate model based evaluations validated against both differential and integral measurements have been obtained. Efforts have been focused on evaluating the optimal mean values for physical quantities of interest in neutron transport. Evaluation of uncertainties and correlations will be undertaken at a later stage similarly as done before for thorium, manganese, and tungsten evaluations [15,16].

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2.1. Reaction models and parameters

The optical and direct reaction models, the pre-equilibrium exciton model, and the full featured Hauser-Feshbach statistical model have been used to calculate the observables for neutron induced reactions on ^{235}U. The starting values for the model parameters were taken from the Reference Input Parameter Library (RIPL) [17].

The total, direct elastic and inelastic cross sections and the transmission coefficients for the incident neutron channel were calculated with ECIS06 code [18] (within the EMPIRE-3.2 code system) using a dispersive coupled-channel optical model potential of Capote et al. [19,20] (RIPL 2408 [17]) with the coupling of seven levels of the ground state rotational band as recommended in Refs. [21,22]. Dispersive integrals were calculated analytically [22–24]. Rigid-rotor structure has been shown to be an excellent approximation for odd-mass actinides [22]. Such an optical model is isospin-dependent and approximately Lane consistent [25,26] as demonstrated in Ref. [27]. An excellent agreement with available experimental data was achieved, including total and elastic cross sections and angular distributions.

DWBA calculations on discrete levels from a neighbouring even-even nucleus were used to estimate the cross sections for fragmented vibrational states. These calculations were combined with a compound nucleus contribution to determine the inelastic scattering cross sections and the corresponding angular distributions on low-lying discrete states.

Additionally, we followed the empirical approach proposed by Young et al. [28], who postulated the existence of a series of collective 2^+ and 3^- states (see Table VI in Ref. [28]) at excitation energies in the continuum ($E_x = 1$–4 MeV) with dynamical deformations fitted to reproduce measured emission spectra at 14 MeV on ^{238}U target. Cross sections and angular distributions from these collective states were also calculated by DWBA, and their strength was spread in the continuum using a Gaussian resolution function with $\sigma = 70$ keV.

The statistical model used in the EMPIRE system is an advanced implementation of the Hauser-Feshbach theory [29]. The exact angular momentum and parity coupling is observed. Discrete level schemes retrieved from RIPL [17] are considered up to a defined cut-off energy, and level densities above. To account for the correlation between incident and exit channels in elastic scattering (i.e., the width fluctuation correction), the model proposed by Hofmann, Richert, Tepel and Weidenmüller (HRTW) [30] is used. The importance of considering the influence of direct reactions on calculated neutron inelastic scattering for well-deformed nuclei was recently highlighted [31,32]. This effect increases the neutron inelastic scattering cross section on coupled levels and decrease the elastic cross section, accordingly [32]. However, this effect is weaker for odd targets, due to the higher density of excited levels, and it was neglected in current calculations. Therefore, calculated elastic (inelastic) cross sections may be slightly overestimated (underestimated) for neutron incident energies below 1 MeV.

The level densities, both at equilibrium deformation and at the saddle points, are described with the Enhanced Generalized Superfluid Model (EGSM) [10,11,17] with parameters appropriate for each deformation. The order of symmetry of the nuclear shape at saddles was taken into account by multiplying the level density with specific enhancement factors as defined in Refs. [33,34].

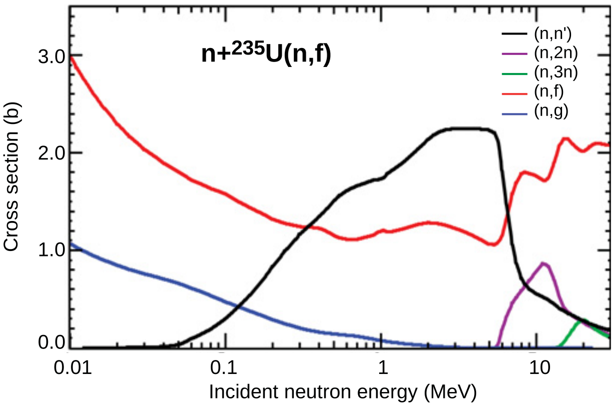

Figure 1. EMPIRE calculated cross sections for major neutron induced reactions on ^{235}U used in current evaluation.

The full γ-cascade in the compound and residual nuclei was considered, including both continuum and discrete transitions, with a maximum multipolarity of 2. Gamma-ray transmission coefficients were obtained from γ-ray strength functions computed using the Modified Lorentzian model (MLO1 closed form) as described in Ref. [17] renormalized to $2\pi\Gamma_\gamma/D_0$ determined from experimentally inferred values of Γ_γ and D_0 [17].

Pre-equilibrium emission was taken into account by the EMPIRE module PCROSS, featuring a one-component exciton model with gamma, nucleon and cluster emissions [35].

Theoretical fission barriers had been used to calculate neutron-induced fission cross sections, but an accurate prediction of fission cross sections remains elusive [36,37]. Empirical triple-humped fission barriers with shallow tertiary humps have been employed for the compound nucleus ^{236}U and for the lighter uranium isotopes responsible for the multiple fission chances [38,39], as suggested by theoretical barrier studies [40]. The fission coefficients have been calculated with a formalism based on the extension of the optical model for fission [39,41], which describes the direct and indirect transmission across the multi-humped fission barriers.

3. Calculated cross sections

Neutron induced reactions on ^{235}U have been calculated following the reaction modelling reviewed in previous section [38]. All major calculated cross sections (excluding total and elastic) are shown in Fig. 1. Neutron capture is practically negligible above 1 MeV, inelastic cross section is larger than fission from 300 keV up to 5 MeV, and multiple neutron emissions are much lower than fission.

Let's review some of relevant reaction channels. The calculated ^{235}U(n,f) cross section is within 3% of the IAEA library Standard cross section [42,43] as shown in Fig. 2. Such agreement allows for a proper calculation of competing neutron emission channels – (n,n'), (n,$2n$), (n,$3n$) - and neutron capture. Calculated ^{235}U(n,f) cross section was replaced by the Neutron Standard

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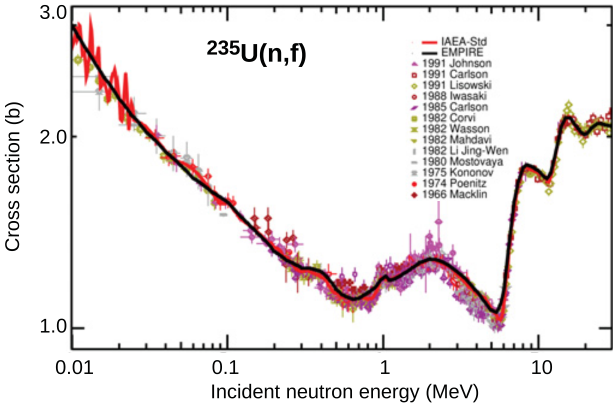


Figure 2. EMPIRE calculated ^{235}U(n,f) cross section in the fast neutron range compared with Neutron Standards (IAEA-Std) [42,43] and selected experimental data from Refs. [44–54] and retrieved from EXFOR [55].

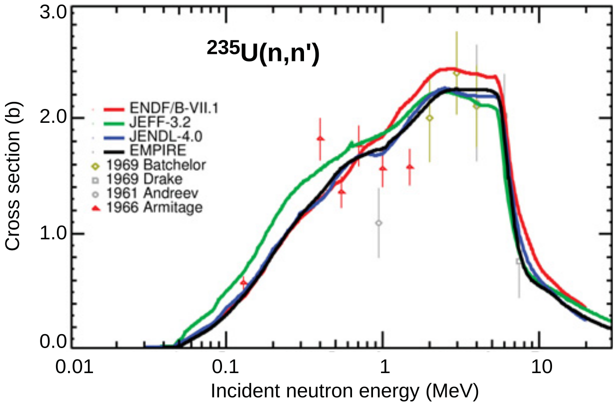


Figure 3. Evaluated cross sections [4–6] for ^{235}U(n,n') compared to current evaluation and experimental data [56–59].

^{235}U(n,f) cross section [42,43] in the IAEA CIELO ^{235}U evaluated file.

EMPIRE calculations of ^{235}U(n,n') cross section are compared with selected evaluations [4–6] in Fig. 3. Large differences between different evaluations from 100 keV up to 2 MeV are observed [9]. The JEFF-3.2 evaluation [5] is consistently larger than all other evaluated data from the threshold up to around 1 MeV. The ENDF/B-VII.1 evaluation [4] is slightly higher than current calculations in the maximum, while JENDL-4.0 evaluation [6] is in very good agreement with current IAEA CIELO evaluation in the whole energy range.

(n,2n) reaction is the main competition to fission above 7–8 MeV of neutron incident energy; calculations are compared with selected evaluations in Fig. 4. Preequilibrium emission plays an important role in multiple neutron emission calculations, and the high energy (HE) tail seen in (n,2n) evaluations and current calculation comes from the contribution of non-compound reactions (either from the direct inelastic scattering to discrete levels or continuum, or from the pre-equilibrium emission). The HE tail in (n,2n) EMPIRE calculations above 15 MeV is higher than in other evaluations due to the inelastic scattering to states in the continuum. EMPIRE calculation agrees much better with the ENDF/B-VII.1 and JENDL-4.0 evaluations up to 15 MeV. Experimental data are well described by the current calculations.

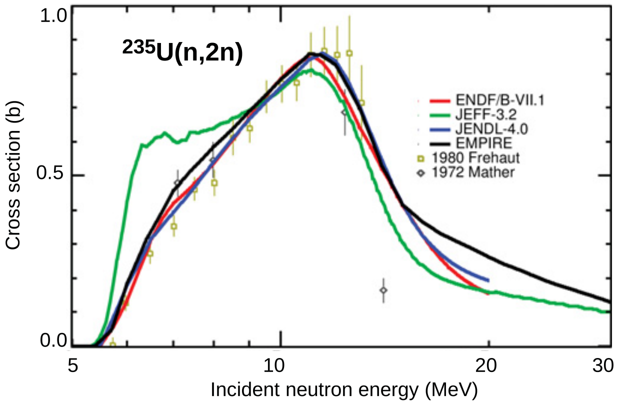


Figure 4. Evaluated cross sections [4–6] for ^{235}U(n,2n) compared to current evaluation and experimental data [60,61].

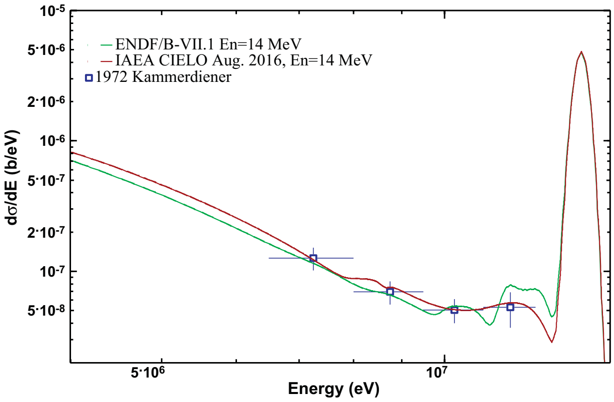
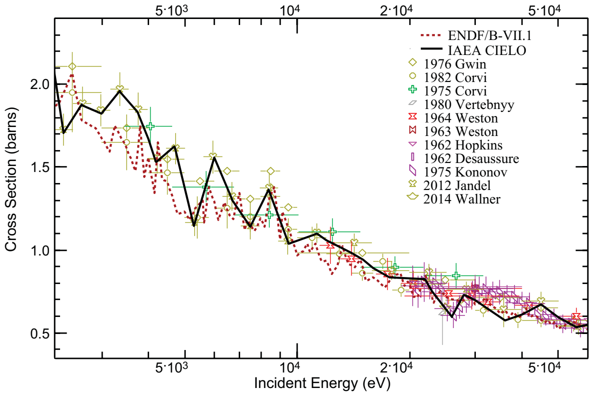


Figure 5. Evaluated neutron emission spectra at 14 MeV incident neutron energy vs Kammerdiener experimental data [62].

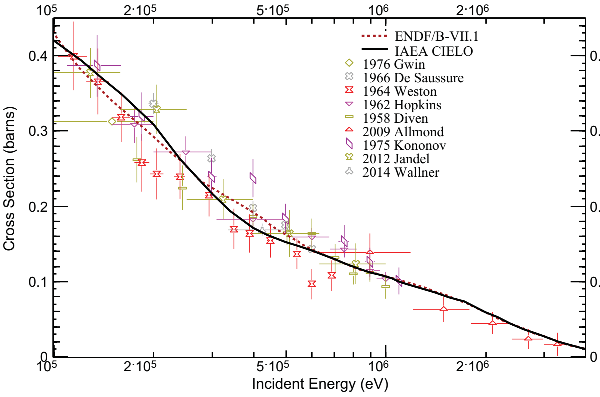
The assumed collective levels in the continuum also lead to improved description of Kammerdiener experimental data [62] on neutron emission spectra at 14 MeV as shown in Fig. 5, as well as to improved Monte Carlo simulations of the time-of-flight spectra in pulsed-sphere experiments performed at the Lawrence Livermore National Laboratory (LLNL) for the ^{235}U target [63].

Calculated neutron capture cross sections using the statistical model agrees well on average with existing evaluations at energies from 10 keV up to 1 MeV as shown in Ref. [38]. However, recent experimental data for ^{235}U(n,γ) reaction have been published by Jandel et al. [73] showing fluctuations from 2 keV up to 200 keV. Additionally, unique AMS ^{235}U(n,γ) measurements using well-defined neutron spectra with average energy equal to 25 and 426 keV have been published by Wallner et al. [74] relative to very well known neutron capture cross section on gold and ^{238}U targets.

Neutron capture cross sections were modified to follow Jandel fluctuations as shown in Fig. 6(a), as statistical model can not describe those fluctuations. Additional reduction of capture was needed to agree with Wallner data; the relatively large reduction around 25–35 keV was confirmed by Kononov et al. [53], Vertebniy et al. [68] and Corvi [65] datasets, which were recalculated from measured α ratio using the ^{235}U(n,f) standard cross section [42,43]. There is some discrepancy in data near 35 keV, lower data was preferred as done in ENDF/B-VII.1 evaluation.



(a) Above the resolved resonance range upto 100 keV.



(b) Fast neutron range above 100 keV.

Figure 6. Evaluated ^{235}U(n,γ) cross section compared to experimental data retrieved from Refs. [53,64–77].

Finally, some reduction of capture from 300 to 500 keV following Wallner measurement [74] was made as shown in Fig. 6(b). In that energy region, measured Wallner cross section is in agreement within uncertainties with Weston [69], Jandel [73], De Saussure [75], and Hopkins [71] data, which were also recalculated from measured α ratio using ^{235}U(n,f) standard cross section [42,43].

The agreement of several experimental datasets with Wallner data [74] at both 25 and 426 keV supports the changes made in evaluated capture data. It should be noted that changes made in capture and fission cross sections in the evaluated file are well within uncertainties of the measured total cross sections. No modification of the elastic cross section was made in the evaluated file. Therefore, small differences are reflected in the summed total cross section.

4. Integral benchmarks

Traditionally, nuclear power applications rely heavily on computational means to optimize the design, ensure safety and determine quantities of interest that are difficult to measure directly. Accurate nuclear data that reproduce integral observables are of utmost importance.

Requirements for criticality safety gave rise to an international project of collecting detailed and verified information about reactor experiments that were performed in different laboratories in the world over several decades. This information can be used for the validation of computational methods, as well as nuclear data. The current issue of the Handbook of International Criticality Safety Benchmarks Evaluation Project (ICSBEP) [12] contains 558 evaluations representing 4798 critical, near-critical or subcritical configurations. The current scope of testing was limited to selected critical benchmark cases from the ICSBEP collection.

Table 1. List of highly-enriched uranium (HEU) fuel bare assemblies from the ICSBEP Handbook [12].

No.	ICSBEP Label	Short name	Common name
1	`HEU-MET-FAST-001`	hmf001	Godiva
2	`HEU-MET-FAST-008`	hmf008	VNIIEF_bare
3	`HEU-MET-FAST-015`	hmf015	VNIIEF_UnrCy1
4	`HEU-MET-FAST-065`	hmf065	VNIIEF_UnrCy2
5	`HEU-MET-FAST-018`	hmf018	VNIIEF_Sphere
6	`HEU-MET-FAST-051`	hmf051-01	ORCEF-01
7	`HEU-MET-FAST-051`	hmf051-02	ORCEF-02
8	`HEU-MET-FAST-051`	hmf051-03	ORCEF-03
9	`HEU-MET-FAST-051`	hmf051-15	ORCEF-15
10	`HEU-MET-FAST-051`	hmf051-16	ORCEF-16
11	`HEU-MET-FAST-051`	hmf051-17	ORCEF-17
12	`HEU-MET-FAST-100`	hmf100-1	ORSphere-1
13	`HEU-MET-FAST-100`	hmf100-2	ORSphere-2
14	`HEU-MET-FAST-080`	hmf080	Caliban

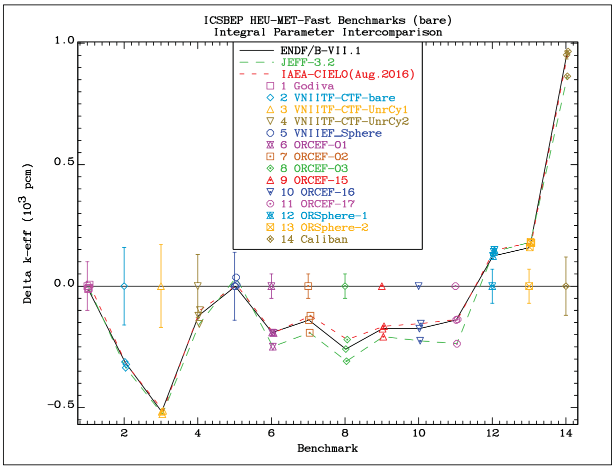


Figure 7. Benchmark results for highly-enriched uranium (HEU) bare assemblies from ICSBEP benchmarks [12]. Symbols linked by lines show the calculated Δk_{eff} values using different libraries.

4.1. ^{235}U bare fast assemblies

There were fourteen bare highly-enriched uranium benchmarks in ICSBEP that were found potentially suitable for the testing. The list is given in Table 1.

Differences Δk_{eff} between the calculated multiplication factors and the reference benchmark values in units pcm (parts per 100 000) are shown in Fig. 7 for ENDF/B-VII.1 [4] and JEFF-3.2 [5] evaluations, together with the results of the IAEA CIELO evaluation (as of August 2016) documented in this work.

There are two benchmark results in excellent agreement: HEU-MET-FAST-001 and HEU-MET-FAST-018. Godiva (HEU-MET-FAST-001) is the only benchmark from Los Alamos national laboratory, NM, USA. Many libraries were tuned to Godiva and its calculated Δk_{eff} lies somewhere in the middle of the calculated results for all fourteen benchmarks. Godiva Δk_{eff} is in excellent agreement with the calculated one for the equivalent Russian spherical benchmark HEU-MET-FAST-018 from VNIIEF, Sarov, Russia.

Unfortunately, the situation is highly unsatisfactory for the remaining benchmarks. They are all extremely sensitive to the ^{235}U nuclear data. The sensitivity profiles available in the DICE package of the NEA Data Bank for the cross sections are very similar. The sensitivities to angular distributions are currently not available for all benchmarks, so a test was made by a direct perturbation of the P_1 coefficient of elastic scattering. The effect on the predicted reactivity was strong, but practically equal for all benchmarks. Sensitivities to higher threshold reaction cross sections are small, so the differences in the emission spectra are not expected to affect criticality selectively for specific benchmarks. In short, any change in the ^{235}U data affects all benchmarks about equally. Therefore, it is not possible to explain large discrepancies well beyond quoted uncertainties and especially a large spread in the calculated Δk_{eff} by nuclear data deficiencies.

An action would be appreciated by ICSBEP curators to ask the benchmark evaluators to identify possible reasons for such large discrepancies and to revise benchmark specifications, quoted benchmark uncertainties, or the corresponding MCNP computational models, if needed. Particularly:

- The Russian bare cylinders and spheres HEU-MET-FAST-008, HEU-MET-FAST-015, HEU-MET-FAST-065, and HEU-MET-FAST-018 show a spread of more than 500 pcm (parts per 100 000) with a bias to a lower reactivity using exactly the same nuclear data as the benchmarks mentioned above that showed good agreement. Therefore, HEU-MET-FAST-008, HEU-MET-FAST-015, HEU-MET-FAST-065 benchmark specifications or computational models require attention if we assume that HEU-MET-FAST-001 and HEU-MET-FAST-018 are acceptable.
- The Oak Ridge cylinders HEU-MET-FAST-051 have a relatively small spread, but they are calculated lower compared to Godiva (HEU-MET-FAST-001) by about 250 pcm. The HEU-MET-FAST-051 suite actually includes 17 cases. Since they all show similar trends, only six are included in the analysis. Some of them show unreasonably small uncertainties. On the other hand, the Oak Ridge spheres HEU-MET-FAST-100 cases are high compared to Godiva by about 200 pcm. Overall, the Oak Ridge benchmarks also have a spread of nearly 500 pcm. Therefore, ORNL benchmark specifications or computational models may need some attention.
- The French Caliban cylindrical assembly HEU-MET-FAST-080 is by far the highest, differing by nearly 1000 pcm from the average. A quick review of the input model and the benchmark specifications (O. Cabellos, private communication, December 2016) reveals that the fissile material mass calculated from the volumes and the densities is 0.5% higher than specified in the benchmark description. This would account for about 400 pcm, which would still leave a discrepancy of about 500 pcm. Therefore, in our opinion this benchmark needs a complete review.

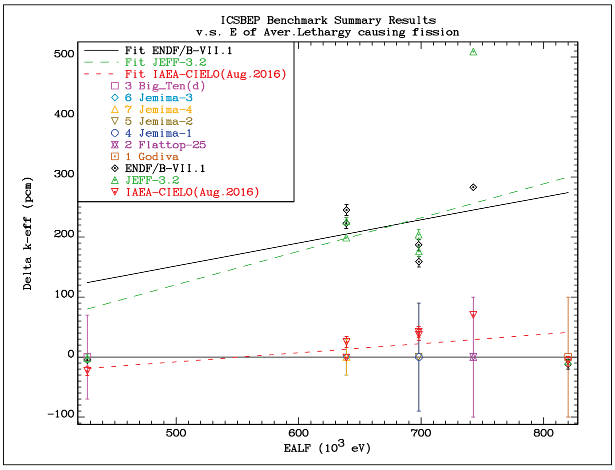


Figure 8. Selected fast ^{235}U cores with ^{238}U contents from ICSBEP benchmarks [12]. Symbols show the calculated values using different libraries; lines show the derived trends of Δk_{eff} as a function of E_α.

The reactivity prediction of current ^{235}U evaluation has been slightly tuned (by less than ±50 pcm using a 0.1% change in nubar) to match the benchmark reactivity value of Godiva, which is somewhere in the middle of predicted reactivities of all benchmarks. Assuming that Godiva is representative of the highly enriched bare assemblies, we believe that the present ^{235}U evaluation represents the best knowledge of the nuclear properties of ^{235}U in the fast energy range in terms of integral performance as well as in terms of measured differential data, consistent with the ^{235}U(n,f) standard cross section [42,43].

4.2. Other selected fast assemblies containing ^{235}U and ^{238}U

A short list of fast-neutron benchmarks is selected, which are used traditionally to test the performance of uranium nuclear data including both ^{235}U and ^{238}U isotopes. The present ^{235}U evaluated data file was complemented by the newly evaluated ^{238}U file [31,78,79] for this exercise. All other relevant isotopes (mainly ^{234}U) were kept fixed to ENDF/B-VII.1 evaluations [4]. The benchmark list is given in Table 2 and includes different types of fast assemblies with regard to fuel material, reflectors, enrichment and spectral characteristics. From experience these benchmarks are considered to be fairly reliable, since they have been analysed by many authors and reported in a variety of publications. We also included Godiva (HEU-MET-FAST-001) results as a reference.

Calculated Δk_{eff} is plotted in Fig. 8 as a function of parameter E_α, which is defined as the energy of the average neutron lethargy causing fission. E_α works as a spectral parameter, weighted by the fission cross section at a given energy, and is easily retrievable from MCNP outputs. From independent studies we know that the softest neutron spectrum of this list corresponds to the Big_Ten assembly which is very sensitive to neutron energies from 10 to 50 keV. On the other end, the hardest spectrum corresponds to the Godiva assembly which is very sensitive to neutron energies above 100 keV with a mean neutron energy around 700 keV. Flattop-25 and Jemima assemblies are between those two extreme cases.

Table 2. List of main benchmarks for ^{235}U and ^{238}U data validation.

No.	ICSBEP label	Short name	Common name
1	`HEU-MET-FAST-001`	hmf001	Godiva
2	`HEU-MET-FAST-028`	hmf028	Flattop-25
3	`IEU-MET-FAST-007`	imf007d	Big_Ten(d)
4	`IEU-MET-FAST-001`	imf001-001	Jemima-1
5	`IEU-MET-FAST-001`	imf001-002	Jemima-2
6	`IEU-MET-FAST-001`	imf001-003	Jemima-3
7	`IEU-MET-FAST-001`	imf001-004	Jemima-4

From the plot it is evident that the new evaluation solves many of the outstanding problems of the older evaluations:

- The over-prediction of reactivity of the Flattop-25 assembly (HEU reflected by natural uranium) is eliminated without affecting the criticality prediction of Godiva and Big_Ten, the later having a softer spectrum due to the presence of graphite and natural uranium reflector.
- Major improvement is achieved in predicting the reactivity of the Jemima assemblies with uranium fuel of intermediate enrichment.

Derived trends of Δk_{eff} vs. E_α for each library are also plotted in Fig. 8 as black solid line (for ENDF/B-VII.1), and red and green dashed lines (for current evaluation and JEFF-3.2 evaluation, correspondingly). The red dashed line shows that the fitted trend for the current IAEA CIELO ^{235}U and ^{238}U file is practically flat and well within quoted experimental uncertainties, representing a big improvement compared to previous evaluations.

The present IAEA CIELO ^{235}U evaluated data file in combination with the new IAEA CIELO ^{238}U file represent an excellent starting point towards a significantly improved evaluated data for major uranium isotopes.

5. Conclusions

A new evaluation of the neutron induced reactions on ^{235}U target in the 2.25 keV–30 MeV incident energy range proposed by IAEA within the CIELO project [1–3] is presented. Theory based calculations - performed with the code EMPIRE–3.2 Malta - combined with latest experimental data for the capture channel, and with non-model evaluated data from Neutron Standards, accurately reproduce both differential and integral measured data. This was possible by improving the reaction modelling, by using the latest nuclear structure information provided by experiments and microscopic studies, by using the newest and more accurate experimental differential data, and by constraining model parameters using the feedback from criticality calculations of carefully selected benchmark experiments. These aspects are of great importance for the ongoing international projects dedicated to the nuclear data development (e.g., the EC CHANDA [80]).

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