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Criticality experiments for validation of cross sections: The neptunium case

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ABSTRACT

The ^{237}Np neutron-induced fission cross section has been measured recently in a large energy range (from eV to GeV) at the n_TOF facility at CERN. When compared to previous measurement the n_TOF fission cross section appears to be higher by 5–7% beyond the fission threshold. To check the relevance of n_TOF data, we simulate a criticality experiment performed at Los Alamos with a 6 kg sphere of ^{237}Np. This sphere was surrounded by enriched uranium ^{235}U so as to approach criticality with fast neutrons. The simulation predicts a multiplication factor k_{eff} in better agreement with the experiment (the deviation of 750 pcm is reduced to 250 pcm) when we replace the ENDF/B-VII.0 evaluation of the ^{237}Np fission cross section by the n_TOF data. We also explore the hypothesis of deficiencies of the inelastic cross section in ^{235}U which has been invoked by some authors to explain the deviation of 750 pcm. The large distortion that should be applied to the inelastic cross sections in order to reconcile the critical experiment with its simulation is incompatible with existing measurements. Also we show that the $\bar{\nu}$ of ^{237}Np can hardly be incriminated because of the high accuracy of the existing data. Fission rate ratios or averaged fission cross sections measured in several fast neutron fields seem to give contradictory results on the validation of the ^{237}Np cross section but at least one of the benchmark experiments, where the active deposits have been well calibrated for the number of atoms, favors the n_TOF data set. These outcomes support the hypothesis of a higher fission cross section of ^{237}Np.

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1. Introduction

^{237}Np is a long lived (2 My) radioactive isotope abundantly produced in nuclear power plants. Therefore its incineration is of interest to reduce the long-term radiotoxicity of the final disposal. This could be in principle achieved in fast reactors, where a significant fraction of the neutron energy spectrum extends beyond the first chance fission threshold. However establishing the feasibility of this option requires a better knowledge of the neutron cross sections specially for fission. This motivated in the past and also recently several measurements of the fission cross section. Although some data sets are consistent within the estimated uncertainty, which is usually about 3–4%, some significant discrepancies show up between others, in particular the n_TOF recent measurement which exhibits values higher by about 6% when compared to several previous measurements (Paradela et al., 2010).

In order to shed light on this problem we have used a critical experiment to check the cross section discrepancies. It consisted of a spherical-like assembly made of a ^{237}Np sphere surrounded by a spherical shell of Highly Enriched Uranium (HEU) in which the neutron multiplication factor k_{eff} is measured with an accuracy of 0.36%. Actually k_{eff} is an integral quantity depending on many neutron reaction parameters; however many of them are accurately known, and its sensitivity to the ^{237}Np fission cross section is large enough (1.5×10^{-3}/%) to get a good test. Even if it cannot be considered as a definitive test, due to its integral nature, it is a good indicator of the reliability of the ^{237}Np (n,f) cross section.

In Section 2 we give an overview of the existing data concerning the ^{237}Np fission cross section. In Section 3 we describe the critical experiment, its simulation with the cross sections as inputs, and how the recent data sets for the ^{237}Np fission cross section affect k_{eff} and how they compare with the experimental value. In Section 4 we explore the hypothesis of a deficient inelastic cross section of ^{235}U which has been invoked by some authors to explain the mismatch on k_{eff} for the critical assembly including ^{237}Np. In Section 5 we examine the impact of an hypothetical increase of the $\bar{\nu}$ of ^{237}Np on k_{eff}. In Section 6 we show how the fission rate measurements discriminate between the cross section data sets.

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2. Status of ^{237}Np fission cross section

Several experiments based on the time-of-flight technique were dedicated to the measurement of this cross section in the fast neutron region. Several monoenergetic measurements also exist, specially close to 14 MeV, but we list below those which cover a larger neutron energy range. Meadows (Meadows et al., 1983) measured ^{237}Np (*n*,*f*) from 1 to 10 MeV in Argonne Fast Neutron Generator (FNG) laboratory. Later Lisowski (Lisowski et al., 1998) measured it in Los Alamos Meson Physics Facility (LAMPF) covering the broad neutron energy range from 1 to 400 MeV. Then Shcherbakov (Shcherbakov et al., 2002) performed another measurement at the Gneiss neutron source in Gatchina. More recently, Tovesson (Tovesson and Hill, 2007) measured this cross section at Los Alamos Neutron Science Center, covering both subthreshold and above threshold fission from 100 keV to 200 MeV. A comparison of the results from the above-mentioned experiments is displayed in Fig. 1 as a ratio to the fission cross section of ^{235}U. Within the experimental uncertainties, all these data are consistent with each other, except the Behrens measurement (Behrens et al., 1983).

Recently, an experiment has been performed at the n_TOF neutron facility at CERN with a neutron spectrum covering an unprecedented energy range 0.7 eV–1 GeV (Paradela et al., 2010, 2011). While being in good agreement with previous measurements and evaluated data files below and around the threshold region, above 1 MeV the n_TOF data are 5–7% larger (Fig. 1), and the mismatch is well beyond the uncertainty on the evaluated cross section (about 3%).

This seemingly singular behavior might shed some doubt on the level of accuracy of the n_TOF results. However, one should be aware that several of the previous results are not independent because they have been renormalized to older ones. For example in Tovesson's measurement the cross section has been normalized to the ENDF/B-VI.8 nuclear data at 14.8 MeV because the amount of target material was not known with the desired precision. This evaluated data file is based on Lisowski's measurement which was normalized to Meadows' data over the 1–10 MeV energy range for similar reasons concerning the targets. Therefore the consistency of several experimental data sets, and of the derived evaluated libraries, is in the first place a consequence of these normalizations. And it is worth noting that most of the existing results around 14 MeV, obtained in monoenergetic beam experiments, are not consistent with the ENDF/B-VI.8 value (Paradela et al., 2011).

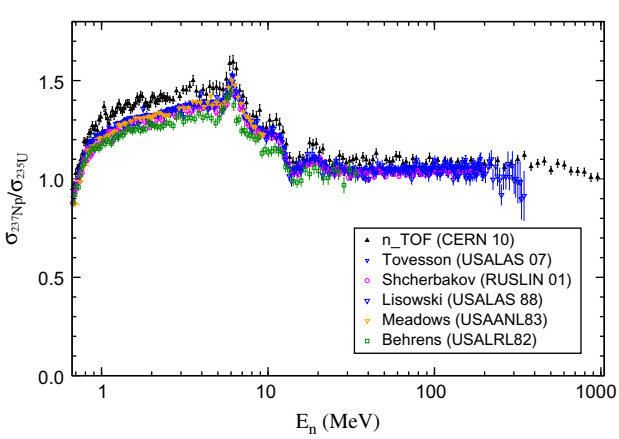

Fig. 1. Comparison of different ^{237}Np fission cross sections measured relative to ^{235}U.

The mismatch between the fission cross sections should be investigated properly. We made one step in this direction by testing them in the simulation of a critical assembly containing a significant quantity of ^{237}Np, and comparing the computed k_{eff} to its value obtained experimentally.

3. The critical ^{237}Np benchmark

3.1. The composite critical experiment

A critical experiment with a composite assembly associating neptunium and HEU has been conducted at Los Alamos with the aim of better defining the critical mass of ^{237}Np (Loaiza et al., 2010; Sanchez et al., 2008). This experiment is based on a ^{237}Np sphere of 6 kg surrounded by nested hemispherical shells of HEU, as sketched in Fig. 2, so as to reach criticality in compact configuration of the system.

^{237}Np is less fissile than ^{235}U due to its threshold, so that only fast neutrons of energy higher than 0.6 MeV can trigger fission. Furthermore ^{235}U contributes to 85% of the fissile mass. Yet 13% of the fissions occur in the neptunium because the neutron flux is higher in the central part. Therefore the k_{eff} is significantly sensitive to the ^{237}Np cross sections, especially to the fission one as fission is the dominant process for multiplying neutrons.

The configuration where the criticality is reached ($k_{eff} = 1$) is determined by extrapolating linearly the inverse of the number of detected neutrons (Sanchez et al., 2008). When all components are in contact, criticality was determined to be $k_{eff} = 1.0026 \pm 0.0036$, that is 360 pcm uncertainty.

3.2. The critical neptunium benchmark

To promote this experiment as a benchmark much easier to simulate, Mosteller and co-workers (Mosteller et al., 2004a,b), applied some simplifications to the geometry by homogenizing some parts with the actual small gaps and by approximating outer structural materials. As the benchmark is very close to the real experiment, the variation of k_{eff} which is expected from the simplifications can be quantified by simulation (Mosteller et al., 2004a,b) and the criticality that would be found if they would be really applied would be $k_{eff} = 1.0019 \pm 0.0036$. This is the reference benchmark value that should be found by simulations based on correct cross sections when using the simplified geometry and composition. This benchmark geometry is represented in Fig. 3 and we used it in all the simulations.

We computed the benchmark with MCNP5 (Monte Carlo code for neutron transport) (X-5 Monte Carlo Team, 2003) driven by MURE (MCNP Utility for Reactor Evolution) (Meplan et al., 2009). The default cross sections were those from the evaluated library ENDF/B-VII.0 (Chadwick et al., 2006). We used 5,750,000 active neutrons distributed over 1200 generations of 5000 neutrons and the 50 first generations were discarded, so that the starting generation was representative of the average distribution.

In such conditions the computed criticality is $k_{eff} = 0.99418 \pm 0.00006$, very close to the value computed by the Los Alamos group (Mihaila et al., 2004). The deviation from the benchmark value is −770 pcm, more than twice the experimental uncertainty. This arouses suspicions about the ^{237}Np cross sections.

It is worth pointing out that the ^{237}Np fission cross section in ENDF/B-VII.0 is essentially based on Tovesson's measurement. As the n_TOF cross section is higher by about 6% above 1 MeV it could help to increase the k_{eff} and bring it closer to the benchmark value. To check this hypothesis we replaced the ENDF/B-VII.0 ^{237}Np fission cross section by the tabulated n_TOF data. All the other cross sections were left unchanged, therefore the total reaction cross

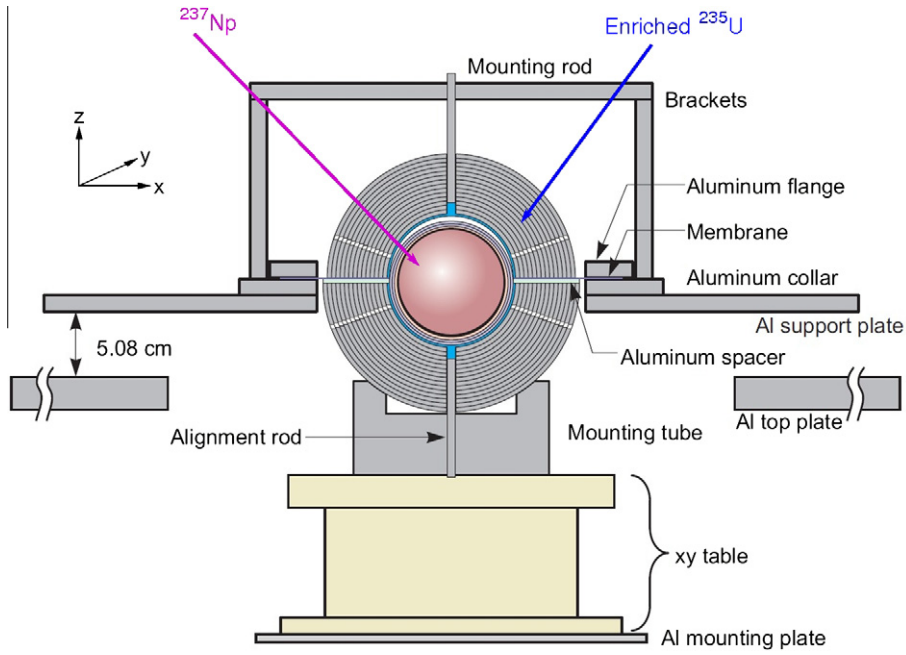

Fig. 2. Detailed scheme of the composite (^{237}Np and HEU) assembly in compact configuration.

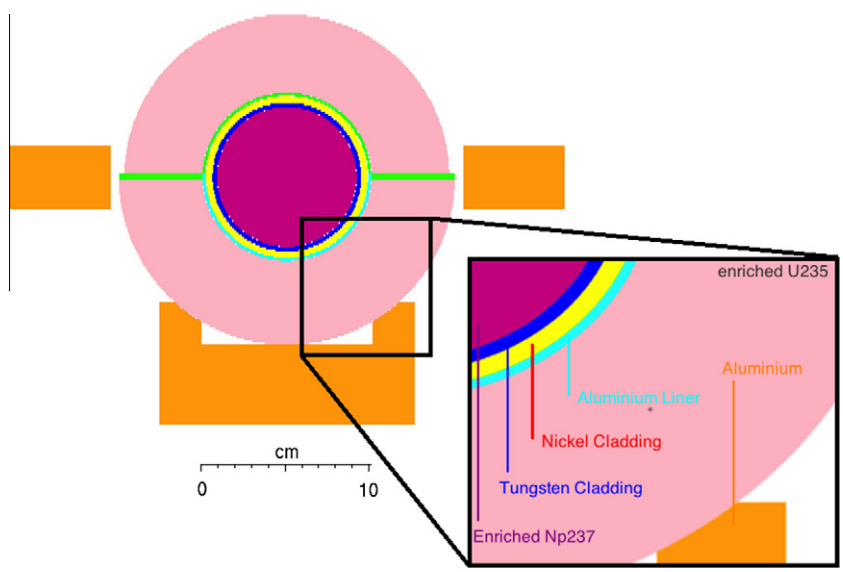

Fig. 3. Neptunium's benchmark geometry. The right-hand side rectangle is an expanded view of a part of the spherical-like assembly.

section was scaled to accommodate the variation of the fission cross section.

After this substitution we obtained $k_{eff} = 1.00435 \pm 0.00006$. Although this slightly exceeds the benchmark value, the deviation is significantly reduced to 250 pcm, 0.7 times the experimental uncertainty. As k_{eff} is sensitive to other parameters, this finding cannot be taken as definitely conclusive on the better fission cross section set. Yet it indicates that the hypothesis of a ^{237}Np fission cross section higher than expected before should be considered seriously.

4. Role of inelastic cross section of ^{235}U

The disagreement between the experimental criticality and its simulated value when using the ENDF/B-VII.0 library, even more so when using ENDF/B-VI.8, has already been noticed (Mihaila et al., 2004; Mosteller et al., 2004a; Chadwick et al., 2006). The authors ascribed this effect to a possible deficiency of the inelastic cross section in ^{235}U. This hypothesis was also formulated following the under-prediction of fission rate ratios ^{237}Np/^{235}U at the center of several critical experiments (Chadwick et al., 2006).

We first study how this cross section affects the k_{eff} in the benchmark configuration, and then we examine how the variation required to match the experimental value compares with measurements of the inelastic cross section.

4.1. Impact of the ^{235}U inelastic cross section

The (n,n') reaction is treated as a set of individual cross sections, one for each excited level in the target nucleus. These levels are

populated at the expense of the incident neutron energy. Changes in the set of inelastic cross sections alter the neutron spectrum. For example, if the cross section for the highest levels is decreased while it is increased for the lowest ones, outgoing neutrons are expected to have a higher average energy: the neutron spectrum is harder. This is illustrated in Fig. 4 where the bell-shaped curves represent the energy spectrum of the neutron flux in the ^{237}Np sphere. The dashed curve, labeled *reduced inelastic*, results from a modification of the inelastic cross sections as described above. As expected, the flux is shifted toward higher energies. The ^{237}Np and ^{235}U fission cross sections are also displayed in order to emphasize the different sensitivity of these nuclei to the change of the neutron spectrum. The fission cross section of ^{235}U is overall flat, so its fission rate is weakly sensitive to shift in the flux. Conversely, ^{237}Np has a fission threshold, hence a harder neutron flux will increase the fission rate (dashed curve).

In order to evaluate if modifications of the ^{235}U inelastic cross sections may reconcile the criticality predicted by the simulation with the ^{237}Np benchmark value, we have performed calculations using various sets of ^{235}U inelastic cross sections. All other cross-sections were left untouched (as in ENDF/B-VII.0), so the total cross section was affected by the variation of the inelastic cross section.

As already noted, the inelastic channel (n,n') involves many cross sections. In MCNP, forty levels are described (MT = 51–90, starting with the first excited level); they sit on top of the continuum (MT = 91) where the energy of the outgoing neutron is smoothly distributed. It is worth noting that in MT = 91, a large number of excited levels are included, mainly located at high excitation energy, and those missed by the recording of the discrete levels as well. Therefore we adopted a random procedure to vary these cross sections. First, we draw randomly the number of affected levels, which are then selected randomly too, and finally for each of these levels the cross section is multiplied by a random factor ranging from 0 to 2.

All the sets generated in this way are not acceptable because the inelastic cross section of ^{235}U also affects its critical mass which is well known. In other words the modification of the inelastic cross section should not change the computed criticality of a HEU sphere. We checked this feature by running a MCNP calculation with each of the proposed cross section sets over the GODIVA benchmark (HEU-MET-FAST-001) describing a bare HEU sphere reaching criticality (LaBauve et al., 2010). If the resulting k_{eff} deviated by more than 50 pcm from the unperturbated calculation (k_{eff} = 1.0000) the cross section set was rejected and another was generated randomly. Among 8000 sets generated, only 551 were validated using the GODIVA benchmark, and were then used for the computation of the ^{237}Np benchmark.

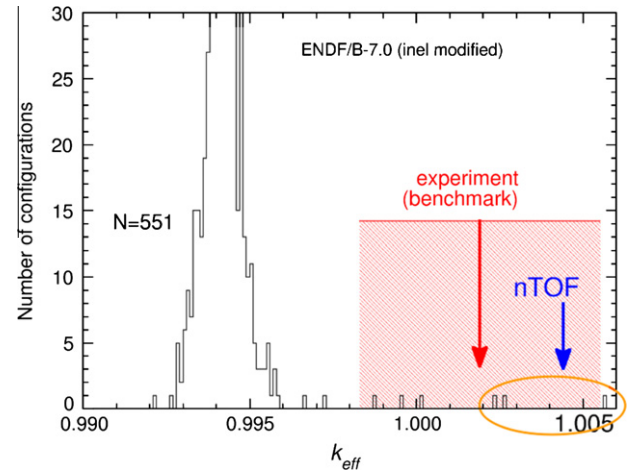

Fig. 5. Spectrum of k_{eff} for the ^{237}Np benchmark computed with acceptable random variations of the inelastic cross section of ^{235}U. The red arrow indicates the benchmark value and the red hatched area the associated uncertainty. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Fig. 5 displays the k_{eff} obtained in these 551 computations. As expected it peaks at the unperturbated value for the ^{237}Np benchmark and spreads rather narrowly around its average. Nevertheless, a few values reach the experimental range. This means that some specific modifications of the inelastic cross section comply with the conservation of the ^{235}U critical mass and have a good agreement with the measured criticality for the ^{237}Np benchmark.

However, the examination of these particular sets reveals that they are always generated by highly depressing the continuum contribution (MT = 91). As already noted, the continuum part contains a large number of continuous excited levels, mainly located at high excitation energy, and is a significant part of the inelastic cross section. Therefore the required shift of the energy spectrum can only be obtained by reducing strongly its contribution in favor of lower lying levels. This is illustrated in Fig. 6, similar to Fig. 5, but where the histograms show the k_{eff} distributions associated to a reduction of the continuum part by 0%, 10%, 25% and 40%.

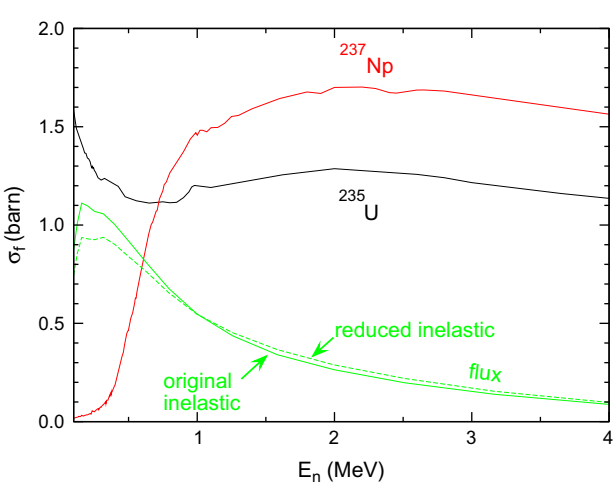

Fig. 4. Effect of a distortion of the inelastic cross section on the neutron flux and fission cross sections of ^{237}Np and ^{235}U.

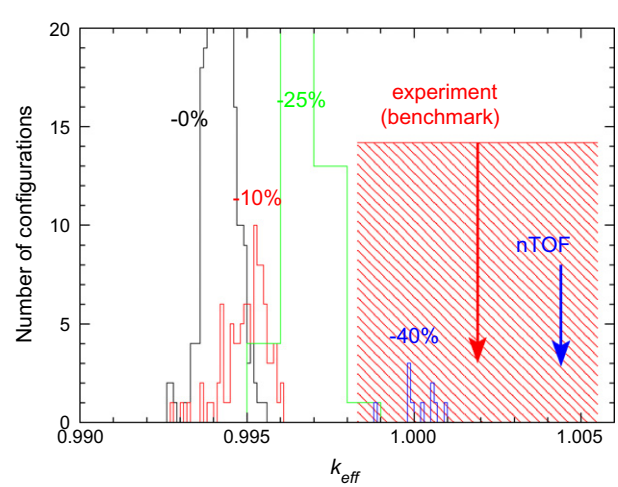

Fig. 6. Same as Fig. 5 but the histograms are associated to different levels of reduction of the continuum part of the inelastic cross section. The reduction factor is indicated by labels on the histograms.

The statistics drop steeply when the level of reduction is enhanced because it becomes more difficult to find cross section configurations conserving the criticality of ^{235}U.

As expected, the more depressed the continuum fraction, the larger the increase in k_{eff}. However a reduction of at least 40% is needed to reach values compatible with the experimental benchmark. This is a large reduction: therefore, in the following, we intend to check the consistency of such an outcome with existing measurements.

4.2. Comparison to previous inelastic measurements

Knitter used a Van de Graaff accelerator to produce monoenergetic neutron beams of 1.5, 1.9 and 2.3 MeV (Knitter et al., 1972) in order to measure the inelastic scattering cross section of ^{235}U. The kinetic energy loss of neutrons was integrated over 200 keV-wide bands from 0.4 MeV to the incident energy minus 0.5 MeV. The results are displayed in Table 1 in the Exp columns.

For MCNP5 with ENDF/B-VII.0 we retrieved the energy spectrum of the inelastically scattered neutrons by considering an infinite slab of ^{235}U of 1 cm thickness and 10 g/cm^3 density hit by neutrons with a kinetic energy of 1.5, 1.9 or 2.3 MeV. All cross sections have been disabled except MT = 51 to 91, representative of the inelastic channels. Therefore the total cross section (MT = 1) contained only these inelastic cross sections. The scattered neutrons were counted on each side of the slab using a F1 tally which scores the number of neutrons crossing a surface.

Fig. 7 shows the inelastic scattering energy spectrum for an incident neutron of 2.3 MeV. The peaks, widened by the recoil kinematics, are produced by populating discrete excited levels of the remaining ^{235}U (MT = 51 to 90), whereas the smooth spectrum represents the continuous part (MT = 91) ending at an energy corresponding to the minimum excitation energy of ≈0.5 MeV.

The spectra were then integrated over bands of 200 keV to allow a direct comparison with Knitter's data. The numbers are reported in Table 1 in the column labeled ENDF/B-VII.0. Fig. 8 shows the comparison for the 2.3 MeV incident energy. The ENDF/B-VII.0 evaluation is close to Knitter's measurement, although a systematic overestimation shows up beyond 0.8 MeV. Measurements from Batchelor (Batchelor and Wyld, 1969), interpolated between 2 and 3 MeV incident energy and integrated over bands of 200 keV for direct comparison, are also plotted. The agreement with ENDF/B-VII.0 is generally even better than in the case of Knitter's data, although a significant discrepancy appears between 0.6 and 1 MeV.

We plotted also in Fig. 8 the spectrum generated by a cross section configuration with 40% reduction of the continuum contribution. We checked that the pattern does not vary significantly among the configurations randomly generated. As expected, the low energy part of the spectrum is depleted and compensated by a strengthening of the part leading to low lying states, hence to high scattered neutrons energy: this will lead to the desired increase of k_{eff} in the ^{237}Np benchmark as some of the low energy neutrons are put beyond the fission threshold. Although such a cross section set seems to be in better agreement with Knitter's measurement above 0.8 MeV, it fails strongly below this energy.

To sort things out, one must stress that one of the main difficulties of such measurements is the subtraction of fission neutrons from the neutron spectra. The lower the energy of the scattered

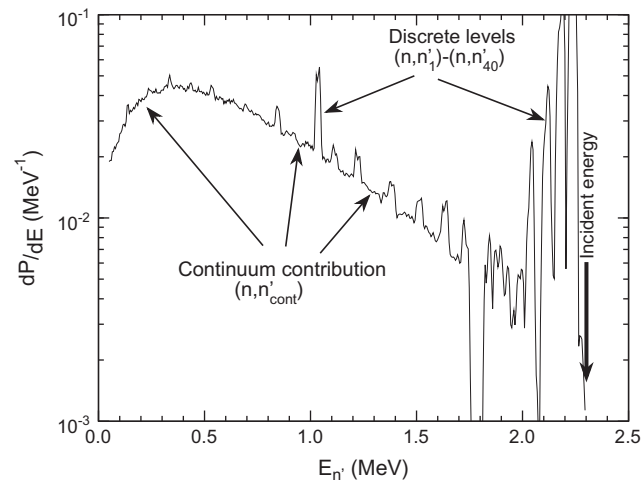

Fig. 7. Energy spectrum of the outgoing inelastically scattered neutron as simulated for ^{235}U using ENDF/B-VII.0 inelastic cross sections (MT = 51–91), for an incident energy of 2.3 MeV (reaction probability).

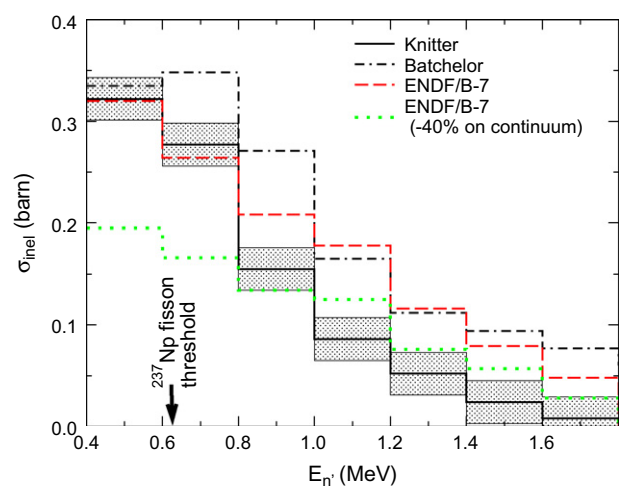

Fig. 8. Comparison of inelastic neutron scattering cross sections of ^{235}U (barn) for energy bands of 200 keV for the outgoing neutron and for an incoming energy of 2.3 MeV. Solid line: Knitterś values with the error bars represented by the hatched rectangles. Dashed line: ENDF/B-VII.0. Dotted-dashed line: measurements from Batchelor. Dotted line: cross section modified with a 40% reduction of the continuum contribution.

Table 1
Comparison between Knitter's inelastic cross section measured for ^{235}U and ENDF/B-VII.0 for energy bands of 200 keV of the outgoing neutron.

$E_n - E_{n'}$ (MeV)	E_n = 1.5 MeV		E_n = 1.9 MeV		E_n = 2.3 MeV	
	Exp $\sigma_{inel} \pm \Delta\sigma_{inel}$	ENDF/B-VII.0	Exp $\sigma_{inel} \pm \Delta\ \sigma_{inel}$	ENDF/B-VII.0	Exp $\sigma_{inel} \pm \Delta\sigma_{inel}$	ENDF/B-VII.0
0.5–0.7	0.117 ± 0.022	0.154	0.046 ± 0.022	0.087	0.008 ± 0.022	0.048
0.7–0.9	0.196 ± 0.022	0.257	0.113 ± 0.022	0.147	0.024 ± 0.022	0.078
0.9–1.1	0.334 ± 0.022	0.321	0.213 ± 0.022	0.205	0.052 ± 0.022	0.115
1.1–1.3			0.294 ± 0.022	0.290	0.086 ± 0.022	0.178
1.3–1.5			0.267 ± 0.022	0.320	0.155 ± 0.022	0.207
1.5–1.7					0.277 ± 0.022	0.264
1.7–1.9					0.322 ± 0.022	0.319

neutrons, the lower the fission contribution: in other terms, we consider the low-energy part results more reliable than the high energy part, an assumption which is corroborated by the consistency of the two measurements, which gets better as one looks at lower scattered neutrons energy. The −40% configuration is strongly descrepant with experimental data in the more reliable region, while it gets closer to Knitter, but further from Batchelor, in the more disputable region.

From this study of the impact of the inelastic cross section we conclude that to reconcile the measurement of the k_{eff} of the ^{237}Np benchmark with simulation only by modifying the inelastic cross section of ^{235}U, one has to reduce the continuum part by about 40% and this is hardly consistent with measurements.

5. Influence of ^{237}Np $\bar{\nu}$

A criticality experiment is in fact an integral measurement and k_{eff} is sensitive to other parameters, beside cross sections. The average number of emitted neutrons per fission $\bar{\nu}$ is also an important parameter for the criticality and a little variation of $\bar{\nu}$ could highly affect k_{eff}. As the ^{235}U $\bar{\nu}$ is well constrained by HEU critical experiments we only apply variations to the ^{237}Np $\bar{\nu}$.

The simulation shows that a 3% increase on ^{237}Np $\bar{\nu}$ (prompt + delayed) produces a k_{eff} shift of 766 pcm. Although only 13% of fissions occur in the neptunium sphere their contribution in k_{eff} is higher than the ^{235}U fission due to the lower escape probability of the emitted neutrons. Therefore an increase of about 3% of $\bar{\nu}$ is needed to make k_{eff} reach the experimental value.

Fig. 9 shows the $\bar{\nu}$ energy dependence for several experimental sets normalized to the ENDF/B-VII.0 values. For sake of clarity the error bars are not reported in the figure but they lie mostly between 0.5% and 1.5%. Although $\bar{\nu}_{tot}$ is the relevant parameter in critical experiments we plotted in Fig. 9 the ratios of $\bar{\nu}_{prompt}$ as the contribution of delayed neutrons is negligible in those ratios. Most of the experimental values are lower than the evaluated ones, except Veeser's data set around 7 MeV, which is well above the mean energy of neutrons in the critical device. No experimental value reaches the target value 1.03. This comparison does not support the plausibility of an underestimation of the total $\bar{\nu}$ that would explain the mismatch between the simulated and the measured criticality.

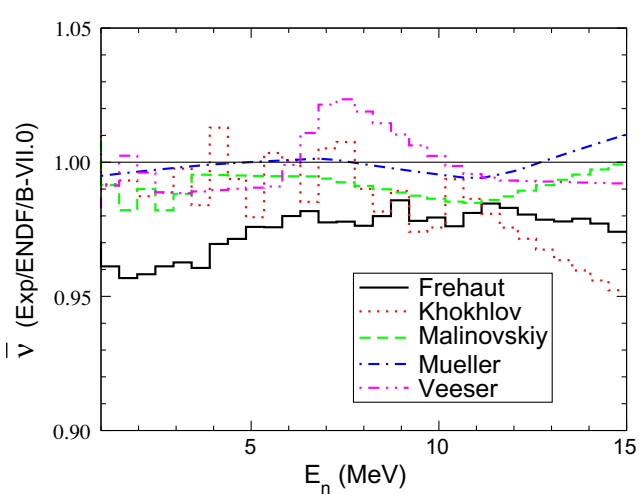

Fig. 9. For fission of ^{237}Np ratio of measured prompt $\bar{\nu}$ to its value given in ENDF/B-VII.0.

6. Fission rate experiments

Fission rate measurements consist of recording the number of fissions, with fission track detectors or inside an ionization chamber, when a sample is subject to a neutron field of known spectrum. The rate is directly related to the fission cross section as it is the folding product over energy of the cross section by the energy spectrum. One can define the averaged cross section as:

$$\langle\sigma\rangle = \frac{\int W(E)\sigma(E)\,dE}{\int W(E)\,dE}$$

where $W(E)$ is the neutron energy spectrum. And the fission rate ratio, or spectral index, as the ratios of averaged cross sections:

$$R_f = \frac{\langle\sigma\rangle_{Np237}}{\langle\sigma\rangle_{U235}}$$

ENDF/B-VII.0 and ENDF/B-VII.1 (Chadwick et al., 2011) libraries have been tested with different fission rate experiments (Kahler et al., 2011). Unfortunately for several of them the description of the experimental details and the method used to calibrate the fission rate is missing. Therefore we discuss only two of the test cases presented in (Kahler et al., 2011), and we add two other examples, with a ^{252}Cf neutron source, where the experimental details are available.

The first situation we consider is the case of fission rates measured at the center of a GODIVA assembly. It is an enriched ^{235}U sphere (LaBauve et al., 2010) and the ratio of fission rates is obtained with ^{237}Np and ^{235}U samples by dividing the former rate by the latter. Table 2 (first line) shows the comparison between this measured ratio, its simulated value when using the ENDF/B-VII.0 library, and the computed value when the n_TOF ^{237}Np fission cross section is used. The ENDF/B-VII.0 underestimates the result by 2.4% whereas the n_TOF data is higher by 3.5%.

We also looked at the COSMO benchmark which is a well defined critical configuration of MASURCA (180 cm × 180 cm × 164.2 cm) (MASURCA-Benchmark, 2005) made of 3 parts, which compositions are homogenized: MOX core, reflector and shielding. The fission rates of ^{237}Np and ^{235}U have been measured at several points but we refer here to the central measurement where the flux has a maximum. Again Table 2 shows the experimental ratio ^{237}Np/^{235}U and the results of the computations. From this comparison it turns out that the n_TOF cross section is too high by 5% whereas the ENDF/B-VII.0 is very close to the experimental value.

The last situation we will discuss is the case of a neutron field generated by the spontaneous fission of a ^{252}Cf source. The fission rates from ^{237}Np and ^{235}U deposits in ionization chambers are recorded when they are placed at different distances from a stainless steel spherical shell containing a ^{252}Cf source, as described in the FUND-NIST-CF-MULT-FISS-001 benchmark (Kim et al., 2010). The manufacturing of ionization chambers and the fissile deposits is described in (Grundle et al., 1975) and their absolute calibration has been obtained by α-spectroscopy and α-counting. The spherical shell can be filled with water but we just consider the dry case when the sphere is empty. In such conditions the neutrons are only slightly scattered and moderated by the shell and other mechanical pieces as holders and cases, as emphasized in (Kim et al., 2010). Instead of carrying a full simulation of the experiment we just performed a simple calculation of the fission averaged fission cross section by assuming a pure Watt spectrum coming from the spontaneous fission of ^{252}Cf, with $a = 0.84746\ \mathrm{MeV}^{-1}$ and $b = 1.03419\ \mathrm{MeV}^{-1}$ (Verbeke et al., 2010), which is also the parametrization adopted in ENDF/B-VII. We applied a correction for the tiny moderation of the neutrons. This correction has been estimated from the difference of the unperturbated and moderated fluxes as computed in Kim et al. (2010), and it amounts to 1.2%.

Table 2
Comparison of experimental and simulated fission rates and averaged fission cross section.

Observable	Set-up	Exp.	Calc. (ENDF/B-VII.0)	Calc. (n_TOF)
R_f	GODIVA (HMF001–002)	0.8516 (12)	0.83	0.88
	MASURCA (COSMO)	0.285	0.284	0.299
$\langle\sigma\rangle_{Np237}$ (barn)	FUND-NIST-CF-MULT-FISS-001	1.420 (25)	1.34	1.41
	Adamov et al.	1.442 (23)	1.364	1.431

The comparison between the measured averaged fission cross section of ^{237}Np and the simulation using the ENDF/B-VII.0 ^{237}Np cross section and the n_TOF data is shown in the third line of Table 2. The latter gives a value much closer to the experimental one. It's interesting to note that when using ENDF/B-V the agreement was very good (Verbeke et al., 2010) and ENDF/B-VII does worse because the parametrization of Watt's spectrum for ^{252}Cf has been changed between ENDF/B-V and ENDF/B-VI. Nevertheless the combination of the ENDF/B-VII.0 ^{252}Cf neutron energy spectrum with the n_TOF fission cross section of ^{237}Np allows to recover a very good agreement.

Another absolute fission cross section, under the neutron flux generated by a ^{252}Cf source, has been measured by (Adamov et al., 1977). The comparison with the simulation displayed on the last line of Table 2 shows again a good agreement with the n_TOF data.

This comparison of fission rates seems to be puzzling because it leads to contradictory conclusions according to the different experiments. However we would like to stress that some of them are not calibrated in absolute value. As an example, it is the case of the COSMO experiment where the amount of fissile deposit has not been calibrated in absolute value by α-counting, but from fission rates in a reference neutron flux. Therefore the calibration depends on the fission cross section itself, and the agreement obtained in other neutrons fields like the COSMO measurement merely indicates that the cross section has the right energy dependence but does not say anything on its absolute value. In that respect the shown COSMO comparison is not really significant as the cross section data set used for the calibration and the reactor measurement is not the same.

In the GODIVA case we do not have any information on the calibration procedure of the samples which have been used, but in the case of FUND-NIST-CF-MULT-FISS-001 we know that the samples have been absolutely calibrated either by α counting or by weighing. Overall the fission rates experiment are not contradictory with an increase of the fission cross section of ^{237}Np by 4–5%.

7. Conclusion

We used the ^{237}Np critical benchmark to test the validity of the ^{237}Np fission cross section measured at n_TOF, which appeared to be larger than previous measurements. The k_{eff} predicted using the n_TOF cross section, although slightly exceeding the experimental value, is much closer to the benchmark value and falls inside the uncertainty range whereas this was not the case for older ^{237}Np fission cross sections. As some authors invoked a possible deficiency of the (n,n') cross section in ^{235}U to explain the mismatch we investigated this hypothesis. It turns out that the experimental range can be reached only by depressing the continuum contribution of the inelastic cross section by at least 40%. Such a modification is hardly consistent with existing measurements of the ^{235}U inelastic cross section. Beside, we showed that the discrepancy cannot be ascribed to the ^{237}Np $\bar{\nu}$. Although other effects and uncertainties can affect the criticality, our prediction of the criticality of the ^{237}Np benchmark may indicate that the ^{237}Np fission cross section is higher than expected from previous measurements. When testing the ^{237}Np fission cross section with fission rate experiments the outcomes are not univoque, but we showed that the calibration procedure of the detectors is of crucial importance and its description is often lacking. In two cases where all the information is available and the detectors have been calibrated in absolute value, the n_TOF cross section gives a much better agreement with the measured value. Although this cannot be considered as definitely conclusive, it indicates that a global increase of the ^{237}Np fission cross section by 4–5%, against the data libraries and more peculiarly ENDF/B-VII.0, would reconcile all the comparisons we have shown.

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