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Criticality experiments for validation of cross sections: the ^{237}Np case

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Abstract. ^{237}Np is one of the actinides which is most abundantly produced in present reactors, and it could be a good candidate for incineration due to its long life time. However this requires a good knowledge of neutron cross sections. The n_TOF collaboration has carried out a fission cross section measurement at the n_TOF neutron facility at CERN, spanning a very broad energy range, from a fraction of eV to GeV. When compared to previous measurements the n_TOF fission cross section appears to be higher by 5-7% beyond the first fission threshold. We show however that several previous measurements, although consistent with each other, are dependent through re-normalization procedures. To check the relevance of n_TOF data, we simulate an experiment, performed at Los Alamos, with a sphere of ^{237}Np of 6 kg surrounded by enriched uranium ^{235}U, so as to reach criticity with fast neutrons. When introducing the n_TOF data for the fission cross section of ^{237}Np, the simulation predicts a multiplicating factor k_{eff} in better agreement with the experiment as the deviation of 750 pcm is reduced to 250 pcm. We explore also the hypothesis of possible deficiencies of the inelastic cross section in ^{235}U which has been invoked by some authors to explain the deviation of 750 pcm. However, the large distortion that should be applied to reconcile the critical experiment with its simulation discredits this option. These outcomes support the hypothesis of a higher fission cross section of ^{237}Np.

1 Introduction

^{237}Np is a long lived (2 My) radioactive isotope which is a abundantly produced in nuclear power plants. Therefore its incineration is of interest to reduce on the long term the 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 it 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 claimed accuracy, 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.

To get a clarification on this problem we adopted the method of checking the cross sections within a critical experiment. It consisted in 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

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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 high enough ($1.5\ 10^{-3}/\%$) to get a good test. Even if it cannot be considered as a definite 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. Finally 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.

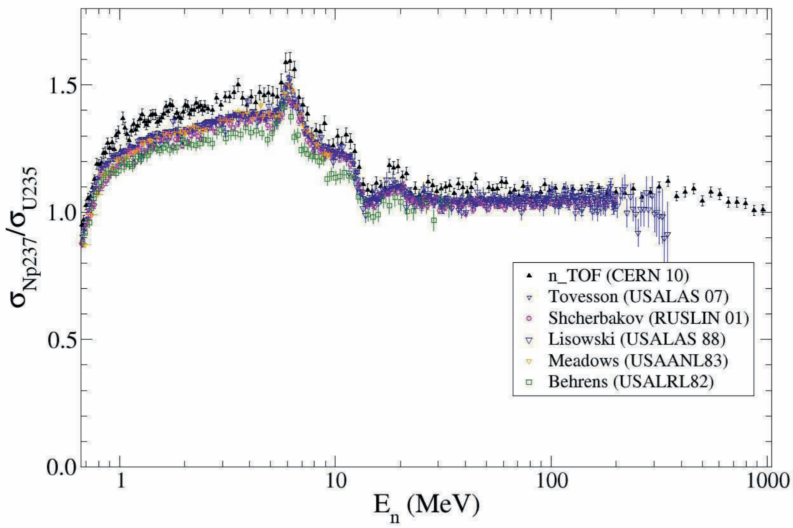

Fig. 1. Comparison of n_TOF ^{237}Np fission cross section, relative to ^{235}U

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. Those which cover a larger neutron energy range are listed below. Meadows [1] measured ^{237}Np(n,f) from 0 to 10 MeV in Argonne Fast Neutron Generator (FNG) laboratory. Later Lisowski [2] measured it in Los Alamos Meson Physics Facility (LAMPF) covering the broad neutron energy range from 1 to 400 MeV. Then Shcherbakov [3] performed another measurement at the Gneiss neutron source in Gatchina. In 2007 F. Tovesson[4] re-measured this cross section at Los Alamos Neutron Science Center, covering both subthreshold and above threshold fission from 100 keV to 200 MeV. All these data are consistent with each other (see figure 1).

Recently, an experiment has been performed at the n_TOF neutron facility at CERN with a neutron spectrum covering the most complete energy range 0.7eV - 1GeV[5][6].

The comparison of the results from the above-mentioned experiments is displayed on figure 1 as a ratio to the fission cross section of ^{235}U. Contrary to several measurements which are consistent within the experimental uncertainty (except the Behrens measurement), the n_TOF's data exhibit an excess of 6-7% above 1 MeV.

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 re-normalized to older ones. For example in Tovesson's measurement the cross section has been normalized to the ENDF/B-6.8 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 has been normalized to Meadows data over the 1 to 10 MeV energy range, for similar reasons concerning the targets. As a result the consistency of several experimental data sets, and of the derived evaluated libraries, is a consequence of this normalization procedure.

Therefore the mismatch between the fission cross sections should be investigated properly. We did 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.

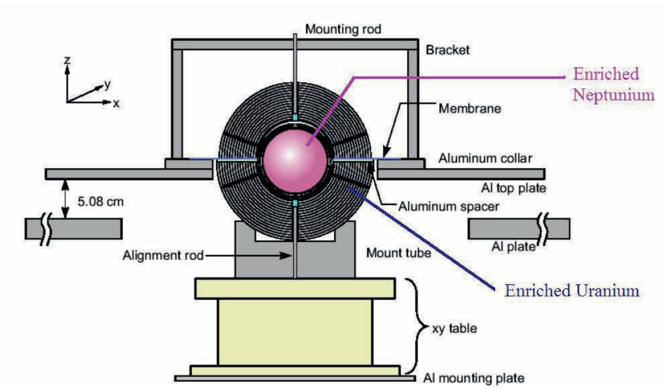

Fig. 2. Detailed scheme of the composite (^{237}Np and HEU) assemby in compact configuration

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 scope of better defining the critical mass of ^{237}Np[7]. This experiment is based on a ^{237}Np sphere of 6 kg surrounded by nested hemispherical shells of highly enriched uranium (HEU), as sketched in figure 2, so as to reach criticity in compact configuration of the system.

Although ^{235}U contributes to 85% of the fissile mass and ^{237}Np is less fissile than ^{235}U due to its threshold, still 13 % of the fissions occur in the neptunium because the neutron flux is higher in the central part. Therefore the k_{eff} is significanlty sensitive to the ^{237}Np cross sections and mostly the fission. The configuration where the criticity is reached (k_{eff}=1) is determined by extrapolating linearly the inverse of the number of detected neutrons. When all components are in contact, criticity was determined to be k_{eff}=1.0026 $\pm$ 0.0036, that is 360 pcm uncertainty[8][9].

3.2 The critical neptunium benchmark

To promote this experiment as a benchmark much easier to simulate, Mosteller and co-workers[8][9], 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

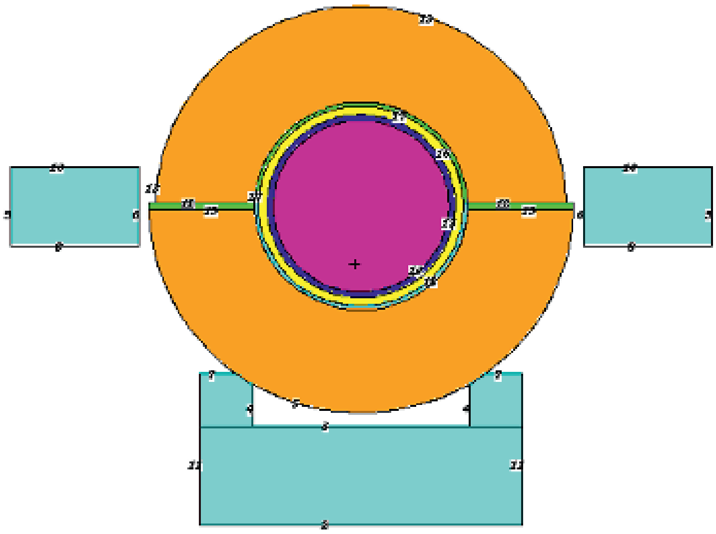

Fig. 3. Neptunium's benchmark geometry

very close to the real experiment, the variation of k_{eff} which is expected from the simplifications can be quantified by simulation[8][9] and the criticity that would be found if they would be really applied would be k_{eff}=1.0019 $\pm$ 0.0036. It 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 figure 3 and we used it in all the simulations.

We computed the benchmark with MCNP5 [10] (Monte Carlo code for neutron transport) driven by MURE [11] (MCNP Utility for Reactor Evolution). The default cross sections were those from the evaluated library ENDF/B-7.0[12], and we used 5750000 active neutrons distributed over 1200 generations of 5000 neutrons and the 50 first generations were discarded, so that the starting generation be representative of the average distribution.

In such conditions the computed criticity is k_{eff}=0.99418$\pm$0.00006, very close to the value computed by the Los Alamos group[7]. Unfortunately the deviation from the benchmark value is -770 pcm more than twice the experimental uncertainty, arousing suspicions about the ^{237}Np cross sections.

We recall that the ^{237}Np fission cross section in ENDF/B-7.0 is essentially based on Tovesson's measurement. As the n_TOF cross section is higher by about 6% above 1 MeV it could help increase k_{eff} and bring it closer to the benchmark value. To check this hypothesis we replaced the ENDF/B-7.0 ^{237}Np fission cross section by the tabulated n_TOF data, leaving all the other cross sections unchanged, therefore the total reaction cross section scaled accordingly to accomodate the variation of the fission cross section.

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

4 ^{235}U inelastic cross section

The disagreement between the experimental criticity and its simulated value when using the ENDF/B-7.0 library, and even larger when ENDF/B-6.8 was involved, has already been noticed[7][8][12]. The authors ascribed this effect to a possible deficiency of the inelastic cross section in ^{235}U.

The (n,n') reaction is treated as a set of individual cross sections for excited levels in the target nucleus at the expense of the incident neutron energy. If the cross section for the highest levels is decreased whereas it is increased for the lowest, the outgoing neutron is expected to have a higher energy in average and this hardens the neutron spectrum. This is shown in figure 4 where the bell-shaped curves are for the energy spectrum of the neutron flux. They do not represent reliably the simulated flux and they are drawn only for sake of illustration. The dashed curve, labeled *correct inelastic* results from a modification of the inelastic cross sections as described above and it is shifted toward higher energies. The ^{237}Np and ^{235}U fission cross sections are also displayed and it can be seen that the fission rate in ^{235}U is rather insensitive to the shift on the flux because the cross section is flat, whereas for ^{237}Np the cutting effect of the fission threshold is strong and leads to a higher fission rate for the shifted flux (dashed curve). This possible deficiency of the ^{235}U inelastic cross section was also suspected from the under-prediction of fission rate ratios ^{237}Np/^{235}U at the center of several critical experiments[12].

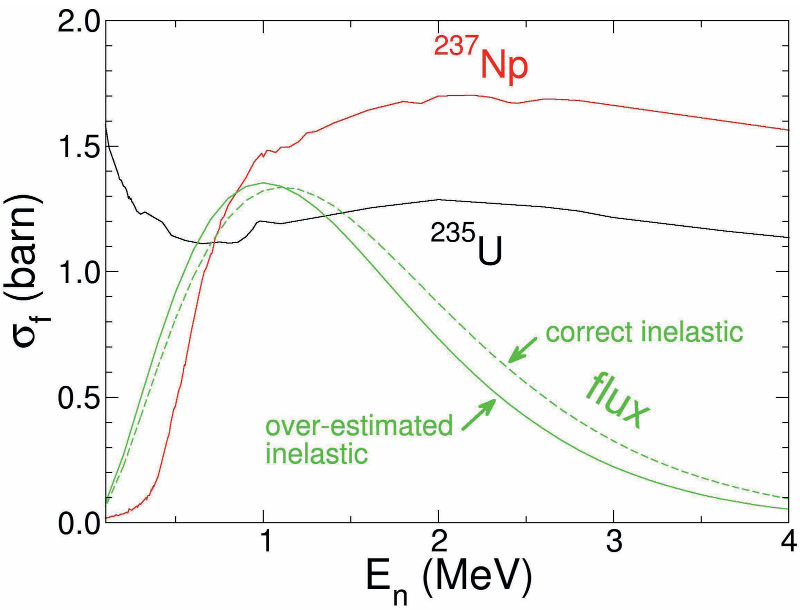

Fig. 4. Effect of a distortion of the inelastic cross section on the mean energy of the neutron flux and its impact on fission rates

We tried to investigate this problem and to quantify the modification of the inelastic cross sections needed to reconcile the criticity predicted by the simulation with the ^{237}Np benchmark value. In the following the ^{237}Np fission cross section remains untouched and equal to the ENDF/B-7.0 one. All cross sections other than inelastic are also kept constant so that the total reaction cross section is affected by the variation of the inelastic cross section.

The inelastic channel involves many cross sections corresponding to the different levels in ^{235}U: in MCNP, MT=51 to 84 for discrete levels and MT=91 for higher levels and the continuum. 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.

However 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 criticity of a HEU sphere. We checked this feature by running a MCNP calculation with the proposed cross section set over the GODIVA benchmark describing a bare HEU sphere reaching criticity[13]. 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. Otherwise the set was retained and applied to the computation of the ^{237}Np benchmark. As a result the application of 551 acceptable inelastic cross section sets needed to run more than 8000 sets over the GODIVA benchmark.

Figure 5 shows how k_{eff} distributes when the acceptable random cross sections are applied. As expected it peaks at the unperturbated value for the ^{237}Np benchmark and spreads rather narrowly around its average, however a few values reach the experimental range. It means that it is possible to find some specific modifications of the inelastic cross section, complying with the conservation of the ^{235}U critical mass, and giving a good agreement with the measured criticity for the ^{237}Np benchmark.

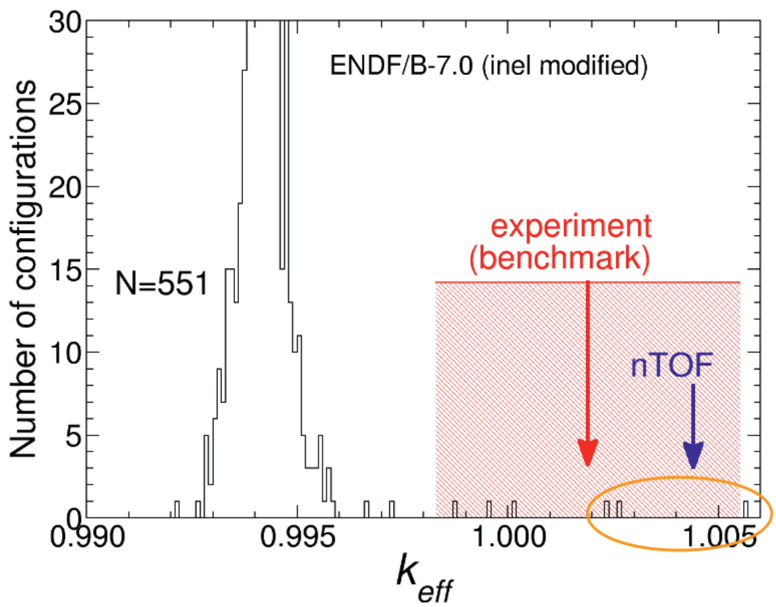

Fig. 5. Spectrum of k_{eff} for the ^{237}Np benchmark computed with acceptable random variations of the inelastic cross section of ^{235}U. The red arrow indicates the benchmark value and the red hatched area the associated uncertainty.

But looking at the pattern of cross section modifications for those configurations one notices they are always generated by highly depressing the continuum contribution (MT=91). In fact, the continuum part contains more than 100 excited levels beyond the 40's first ones, and is a significant part of the inelastic cross section. Therefore the needed shift of the energy spectrum can only be obtained by reducing strongly its contribution, in favor of lower lying levels to conserve the criticity of ^{235}U. This is illustrated in figure 6, similar to figure 5, where the histograms show the k_{eff} distributions when the continuum part is reduced by given factors. The statistics drops steeply when the level of reduction is enhanced because it becomes more difficult to find cross section configurations conserving the criticity of ^{235}U.

As expected the increase in k_{eff} shows up clearly when the continuum fraction is depressed. However it appears that at least a 40% reduction is needed to reach the zone covered by the experimental benchmark.

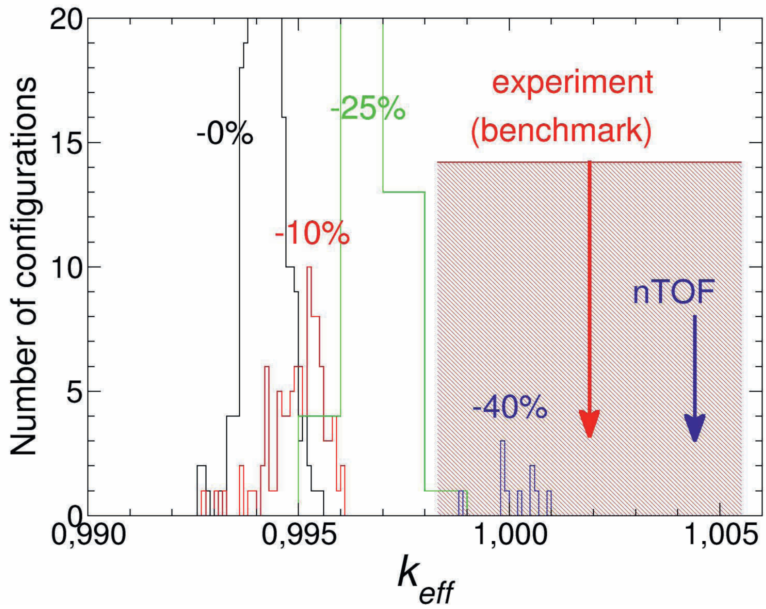

Fig. 6. Same as figure 5 but the histograms are associated to different levels of reduction of the continuum part of the inelastic cross section. The reduction factor is indicated by labels on the histograms.

5 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 higher than previous measurements. The predicted k_{eff}, although slightly exceeding the experimental value, is much closer to the benchmark value and falls inside the uncertainty range whereas it 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 % which does not seem realistic presently. Although a definite conclusion on the validity of the n_TOF cross section cannot be stated, because other effects and uncertainties can affect the criticity, the good prediction of the criticity of the ^{237}Np benchmark may indicate that the ^{237}Np fission cross section is higher than expected from previous measurements.

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