Criticality experiments and benchmarks for validation of cross sections: the neptunium case


Institut de Physique Nucleaire, Orsay; Facultad de fisica, Universidade de Santiago de Compostela, Spain

Neptunium 237

1. Abundant waste produced in present thermal nuclear reactors.
2. $T_{1/2} = 2\ \text{My}.$
3. Candidate for incineration in fast neutron reactors.
4. Burning $^{237}\text{Np}$ needs a better knowledge of neutronic properties (neutron cross sections (XS)).
Motivation

Although most of the measurements are in agreement with each other, the last data obtained at the CERN n_TOF facility are about 5% to 6% higher than the others beyond 1 MeV.

Figure: Ref: C. Paradela et al, Phys. Rev. C82 (2010), 034601.
However! Several previous measurements are not independent.

1. ENDF-B7.0 based on Tovesson measurement (2008).
2. Tovesson’s one normalised to ENDF-B6.8 at 14 MeV.
3. ENDF-B6.8 based on Lisowski’s measurement (1988).
4. Lisowski normalized to Meadows (1983) between 1 and 10 MeV.
5. n_TOF measurement consistent with data at 14 MeV within the experimental uncertainty of 4%

Verification of $^{237}$Np cross section is necessary
Verification of $^{237}$Np Cross Section

1. $^{237}$Np+$^{235}$U Sphere critical model is critical experiment measurement performed in Los Alamos and proposed as a benchmark for neutron transport simulations.

2. $^{235}$U retains 86% of the mass, criticality is still sensitive to fission of $^{237}$Np. (0.3% uncertainty)

3. $K_{eff} = 1.0019 \pm 0.0036$ (experimental value)
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Los Alamos Experiment

Figure: a neutron source inside Np, ($K_{eff}$: Mutiplicatif factor)

$$N = 1 + K_{eff}^2 + K_{eff}^3 + ... = \frac{1}{1-K_{eff}}; N_d = \frac{\epsilon}{1-K_{eff}}$$

Final result: $K_{eff} = 1.0019 \pm 0.0036$
Simulation with MCNP5/MURE

Our work:

- Compute the same Benchmark with same conditions. \( K_{\text{eff}} = 0.9942 \text{ (exp-2}\sigma) \)
- Substitute nTOF Np XS in place of the evaluated data ENDF/B-7.0’s one.
- Result: criticality increased \( K_{\text{eff}} = 1.0043 \text{ (exp+0.8}\sigma) \).

\(^{237}\text{Np fission XS could be higher than previous measurements.}\)
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235U inelastic cross section
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Inelastic cross section
Random variation of XS for excitation of the $^{235}\text{U}$ levels. Criticality of $^{235}\text{U}$ sphere benchmark should remain invariant: (selection among the generated XS configurations)
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Figure: Criticality according to the continuum reduction

- Continuum (MT=91), bears most of the effect on criticality.
- Variation of criticality by the modification of MT=91, to get closer to the experimental value
Is 40% reduction of the continuum inelastic compatible with existed measurements (Knitter and Batchelor)?
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Figure: Comparison between experimental inelastic cross section measured for $^{235}$U and ENDF/B-7.0, versus the outgoing neutron energy.
3% increase on $\bar{\nu}$ of $^{237}$Np gives 0.76% variation of criticality.
The $^{237}$Np criticality experiment seems to support the n_TOF data. Now, we compare the $^{237}$Np fission rate under different neutron fields.
Reaction Rate: GODIVA, MASURCA

\[ R_f(E) = \frac{\sum_i \Phi(E_i) \times \sigma(E_i)_{237Np}}{\sum_i \Phi(E_i) \times \sigma(E_i)_{235U}} \]

<table>
<thead>
<tr>
<th>Set-up</th>
<th>( R_f(\text{exp}) )</th>
<th>( R_f(\text{calc-ENDF/B-7}) )</th>
<th>( R_f(\text{calc-nTOF}) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>GODIVA(HMF001-002)</td>
<td>0.85</td>
<td>0.83</td>
<td>0.88</td>
</tr>
<tr>
<td>MASURKA(COSMO)</td>
<td>0.285</td>
<td>0.284</td>
<td>0.299</td>
</tr>
</tbody>
</table>

**Table**: The Experimental measurement lies between n_TOF and ENDF/B-7.0 data for GODIVA. However, ENDF/B-7.0 seems to be more consistent with MASURCA reaction rate.
Calculated spectrum-averaged cross section is compared to measured integral benchmarks in $^{252}$Cf spontaneous fission neutron field.

$$<\sigma> = \frac{\int W(E)\sigma(E)dE}{\int W(E)dE}$$

<table>
<thead>
<tr>
<th></th>
<th>calc $&lt;\sigma&gt;$ ($b$)</th>
<th>exp $&lt;\sigma&gt;$ ($b$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{235}$U($n, f$)</td>
<td>1.225</td>
<td>1.21 ± 0.014</td>
</tr>
<tr>
<td>$^{237}$Np(ENDFB7)</td>
<td>1.357</td>
<td>1.361 ± 0.022</td>
</tr>
<tr>
<td>$^{237}$Np(n_TOF)</td>
<td>1.431</td>
<td>1.361 ± 0.022</td>
</tr>
</tbody>
</table>

The table shows that n_TOF fission cross section is 5% higher than the experimental value.
1. We used the $^{237}$Np critical benchmark to test the validity of the $^{237}$Np fission cross section.

2. The $K_{eff}$ predicted using the $n_{\text{TOF}}$ cross section, slightly exceeds the experimental value, it is much closer to the benchmark value.

3. $(n,n')$ cross section in $^{235}$U doesn’t explain the discrepancy. Because the -40% configuration is strongly discrepant with experimental data.

4. The discrepancy can’t be ascribed to the $^{237}$Np $\bar{\nu}$

5. Integral fission rate experiments do not agree completely with $n_{\text{TOF}}$ experiment data.

6. New measurements for confirmation of $^{237}$Np fission cross section are desired.
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THANK YOU!
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ANNEXE
## Criticality experiments and benchmarks for validation of cross sections: the neptunium case


### Introduction

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### Inelastic cross section

<table>
<thead>
<tr>
<th>$E_n$-$E_{n'}$ (MeV)</th>
<th>$E_n$=1.9 MeV</th>
<th>$E_n$=2.3 MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Exp</td>
<td>ENDF/B-7</td>
</tr>
<tr>
<td></td>
<td>$\sigma_{inel} \pm \Delta \sigma_{inel}$</td>
<td></td>
</tr>
<tr>
<td>0.5 - 0.7</td>
<td>0.046±0.022</td>
<td>0.087</td>
</tr>
<tr>
<td>0.7 - 0.9</td>
<td>0.113±0.022</td>
<td>0.147</td>
</tr>
<tr>
<td>0.9 - 1.1</td>
<td>0.213±0.022</td>
<td>0.205</td>
</tr>
<tr>
<td>1.1 - 1.3</td>
<td>0.294±0.022</td>
<td>0.290</td>
</tr>
<tr>
<td>1.3 - 1.5</td>
<td>0.267±0.022</td>
<td>0.320</td>
</tr>
<tr>
<td>1.5 - 1.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.7 - 1.9</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>