LA-UR- 04-7959

Approved for public release; distribution is unlimited.

Title: Analysis of Np-237 ENDF for the Theoretical Interpretation of Critical Assembly Experiments

Author(s): Bogdan Mihaila
Mark Chadwick
Robert MacFarlane
Toshihiko Kawano

Submitted to: DNP Fall '04 Meeting

Los Alamos
NATIONAL LABORATORY

Analysis of the Np-237 ENDF for the theoretical interpretation of critical assembly experiments

Bogdan Mihaila, Mark B. Chadwick, Robert E. MacFarlane, and Toshihiko Kawano
Theoretical Division, Los Alamos National Laboratory, Los Alamos, NM 87545

We report on the present status of our effort toward an improved Np-237 evaluated nuclear data file (ENDF). The aim here is to bridge the gap between calculated and observed k-eff values, as measured at the Np-U critical assembly at LANL, TA-18. As such, we perform a critical analysis of the existing body of experimental data and recommended evaluations. We are targeting in principal the fission nu-bar and cross section in Np-237, as well as the inelastic scattering which is particularly important since Np-237 is a threshold fissioner. This analysis will be employed in a future sensitivity study of the calculated k-eff with respect to variations of the afore mentioned nuclear data.

I. INTRODUCTION

Power nuclear reactors produce substantial quantities of Neptunium isotopes, ^{237}Np and ^{239}Np. While ^{239}N decays quickly ($t_{1/2}$=2.355 days, the ^{237}Np has a very long half-life (2.14×10^6 years). Therefore, the critical mass of ^{237}Np is a quantity of interest, in particular for non-proliferation concerns.

^{237}Np has an effective threshold for fission at $\approx$ 550 keV. In nuclear reactors, ^{237}Np is produced by (n,2n) reactions on ^{238}U, and also by (n,γ) reactions in ^{236}U, followed by the β^- decay of ^{237}U. As the ^{237}Np nuclear cross sections are not very well known, neither is its critical mass (see Tab. I for predictions based on the existing nuclear data evaluations). Previous estimates of the critical mass have been based primarily on so-called "replacement" measurements, where only very small quantities (tens of grams) of ^{237}Np were involved.

The 2000 Np-Composite experiment [1] performed on the Planet vertical assembly machine at the Los Alamos Critical Experimental Facility (TA-18), aimed to changed this situation, and provide a more realistic experimental setup from which a reliable determination of the Np critical mass can be inferred. The 2000 Np-Composite experiment (see Fig. 1) involved a 6.0704 kg sphere of nearly pure ^{237}Np surrounded by nested hemispherical shells of highly enriched uranium (HEU) to produce a critical configuration. The radius of the Np sphere was 1.6335 inches, leading to a a density of 20.289 g/cm^3. The sphere was enclosed in tungsten and two layers of nickel in order to reduce the radiological dose due to 310 keV gamma-rays from ^{233}Pa. The estimated total mass of the HEU shells was estimated to be 62.555 kg. Most importantly, the expected value of k_{eff} was 1.0026 ± 0.0034.

TABLE I: MCNP results for k_{eff} corresponding to the Np-composite calculation

Evaluation	^{237}Np critical mass estimate [kg]
CENDL-2	52.4
JEF-2	82.3
JENDL-3.2	80.2
JENDL-3.3	65.3
ENDF/B-VI	64.9

The analysis of the experimental data leads to the conclusion that the criticality is substantially underpredicted: $k_{eff} = 0.9896 \pm 0.0003$. The corresponding underprediction (Δk_{eff} = -0.0130 $\pm$ 0.0034), can be accounted for in part by noting that only about 1 fission in for 8 actually occurs in the ^{237}Np sphere, and therefore the evaluation of k_{eff} is dominated by uncertainties in the ^{235}U nuclear data. However, this source of possible errors is estimated to reduce the k_{eff} underprediction by only 0.009 Δk_{eff}. A second possible source of errors, namely the so-called "missing-mass" problem is estimated to have an even smaller effect. The "missing-mass" problem is related to the fact that 0.946 wt. % of the mass of the sphere is unaccounted for, because the sample assay did not dissolve completely. This in turn results in an uncertainty of the underprediction of $\approx \pm 0.0012\ \Delta k_{eff}$.

The purpose of this report is to perform an analysis of the existing body of experimental data and recommended evaluations. This analysis will be employed in a future sensitivity study of the calculated k-eff with respect to variations of the afore mentioned nuclear data.

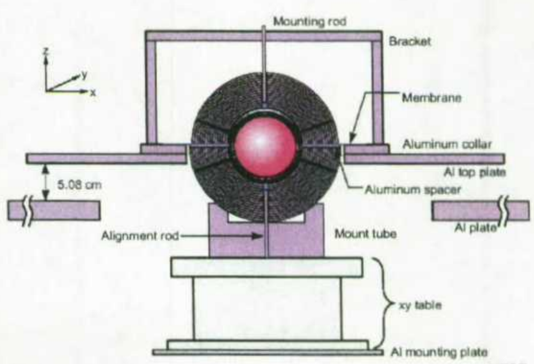

FIG. 1: TA-18 Np-composite experimental setup.

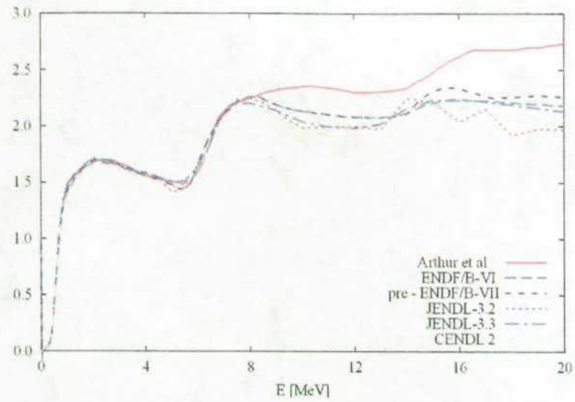

FIG. 2: ^{237}Np fission cross section.

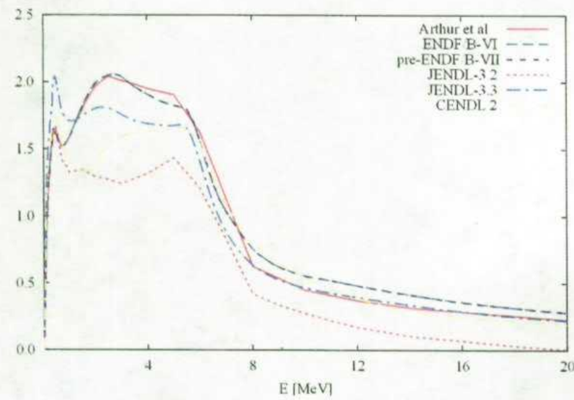

FIG. 3: ^{237}Np inelastic cross section.

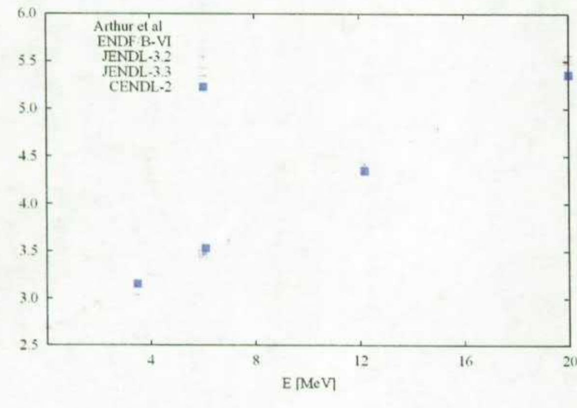

FIG. 4: Total fission $\bar{\nu}$ in ^{237}Np

II. NUCLEAR DATA EVALUATIONS.

Figs. 2-6 depict the energy dependence of the fission and inelastic cross sections of ^{237}Np, the total, delayed and prompt fission $\bar{\nu}$ (the average number of neutrons resulting from one fission process), in ^{237}Np. Due to its importance for the k$_{eff}$ simulations, we detail in Fig. 7 the energy dependence of the prompt fission $\bar{\nu}$ corresponding to an energy range of the incident neutron (E) between thermal and fast regimes, respectively. Actual values of the various $\bar{\nu}$'s are shown in table II. We note here that

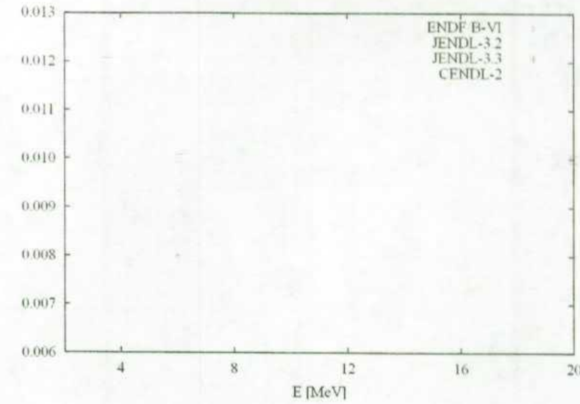

FIG. 5: Delayed fission $\bar{\nu}$ in ^{237}Np.

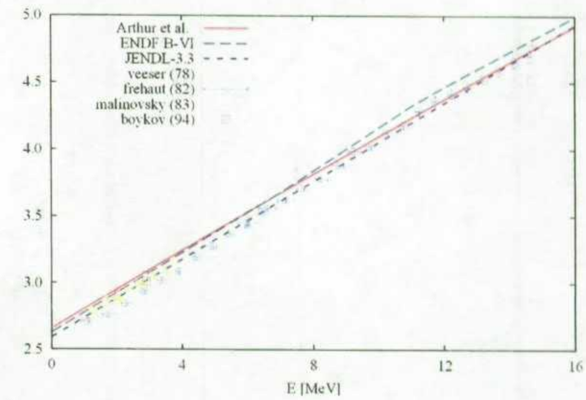

FIG. 6: Prompt fission $\bar{\nu}$ in ^{237}Np, compared with the available experimental data.

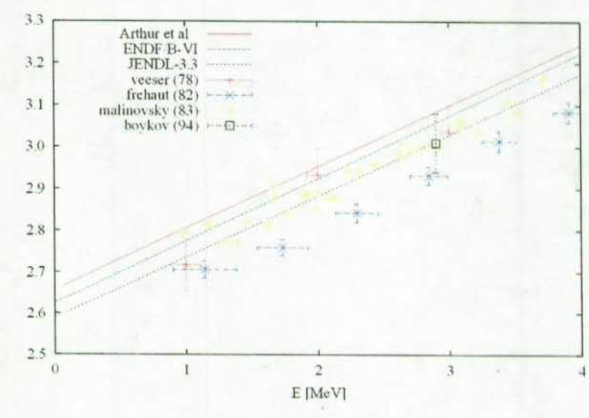

FIG. 7: Same as Fig. 6 (detail).

TABLE II: Values of fission $\bar{\nu}$ corresponding to thermal and fast (1 MeV) neutron energies, in various commonly used evaluations

	$\bar{\nu}_{\text{total}}$		$\bar{\nu}_{\text{delayed}}$		$\bar{\nu}_{\text{prompt}}$	
	@ 10^{-5} eV	@ 1 MeV	@ 10^{-5} eV	@ 1 MeV	@ 10^{-5} eV	@ 1 MeV
Arthur et al	2.67120E+00	2.81900			2.65820E+00	2.80620
ENDF/B-VI	2.63581E+00	2.78562	1.08100E-02		2.62500E+00	2.77481
JENDL-3.2	2.54060E+00	2.68189	1.22000E-02		2.52840E+00	2.66968
JENDL-3.3	2.60140E+00	2.74780	1.20000E-02		2.58940E+00	2.73580
CENDL-2	2.52840E+00	2.70603	1.22000E-02		2.51620E+00	2.69389

TABLE III: MCNP results for k_{eff} corresponding to the Godiva calculation

	ENDF/B-VI	pre-ENDF/B-VII
w/ ^{234}U	0.9971 ± 0.0006	0.9997 ± 0.0006
w/o ^{234}U	0.9968 ± 0.0006	1.0001 ± 0.0006

TABLE IV: MCNP results for k_{eff} corresponding to the Np-composite calculation

ENDF/B-VI	pre-ENDF/B-VII
0.9893 ± 0.0003	0.9926 ± 0.0002

the delayed fission $\bar{\nu}$ has only a small contribution to the neutron multiplicity calculation, and therefore k_{eff} is not particularly sensitive to this quantity.

The various evaluations for the prompt fission $\bar{\nu}$ are compared in Figs.6 and 7 with the available experimental data of Refs. [2–6].

Primarily included in this review are the pre-ENDF/B-VI evaluations of Arthur et al, the ENDF/B-VI evaluation, and the pre-ENDF/B-VII evaluation. We compare the above nuclear data evaluations with the Japanese standards, JENDL-3.2 and JENDL-3.1, and the Chinese evaluation, CENDL-2.

The $\bar{\nu}$ values in the pre-ENDF/B-VII and ENDF/B-VI are identical at this time. These two evaluations differ mainly through a revised evaluation of the fission cross section in ^{235}U. Since all other cross sections are reported as ratios with respect to ^{235}U, the ^{235}U cross section turns out to be the reason for the change in the MCNP estimate of k_{eff}, when comparing the pre-ENDF/B-VII and ENDF/B-VI nuclear sets of data.

III. MCNP CALCULATIONS

We have performed a detailed analysis of the k_{eff} estimates using nuclear data libraries based on the various ENDF/B evaluations. We have used the MCNP5 Monte-Carlo code [7] to simulate the neutron transport and derive theoretical values for k_{eff} in both Godiva and Np-composite geometries. Each calculation employed 5000 generations of 1250 neutrons each. First 50 generations were excluded from the statistics in each case. Therefore the results for k_{eff} reported here were based on more than 6,000,000 active neutron histories, and are accurate to about ± 0.0005 (two sigmas). The numerical values for the k_{eff} estimates are shown in Tables III and IV for Godiva and Np-composite experiments, respectively. For Godiva, we perform two calculations, with and without ^{234}U added in the material. Our ENDF/B-VI results are statistically equivalent with previously reported k_{eff} estimates, available in the literature (0.9965 ± 0.0002 for Godiva, and 0.9896 ± 0.0002 for the Np-composite experiment).

We observe that the re-evaluation of the ^{235}U fission cross section has resulted in a 0.04% increase of k_{eff} for Godiva, and a better agreement with the experimental value. This in turn also has a sensible effect on the k_{eff} estimate for the Np-composite experiment, which increases by 0.33%. According to our calculations, the initial estimate of the k_{eff} underprediction in the Np-Composite experiment, $\Delta k_{eff} = 0.0133 \pm 0.0034$, is decreased by 0.248 Δk by using the pre-ENDF/B-VII evaluation, due to the adjustment of the ^{235}U fission cross section. To date this is the largest improvement toward agreement with the experimental value, and the uncertainty in the ^{235}U fission cross section results in a much larger change in k_{eff} than previously discussed geometry and material effects.

IV. PRELIMINARY CONCLUSIONS

Based on the above collection of information we can already draw some preliminary conclusions. It is clear at this point that indeed the uncertainties in the ^{235}U data limit our ability to constrain the nuclear cross sections in ^{237}Np. Two observables in particular need further work. First, the inelastic cross section, as in the fast region we observe as much as 50% scatter between the various evaluations, see Fig. 3. Secondly, the prompt fission $\bar{\nu}$ may also be a problem, as it also uncertainties of the order of 4-5%, see Fig. 7. It is important to note that based on previous similar situation, one expects that 1% error in the prompt fission $\bar{\nu}$ reflects linearly in a 1% change in k_{eff}, but the same 1% change may result in a a up to 5% change in the critical mass. Therefore the uncertainty on the prompt fission $\bar{\nu}$ must be better controlled in future ENDF/B evaluations. In addition, since ^{237}Np is a threshold fissioner, it is possible that if the ^{235}U neutron spectrum is too soft, that may have adverse effects on k_{eff} in the Np-composite experiment, while the net effect on the Godiva result is negligible.

[1] Mosteller, R.D., Loaiza, D.J. and Sanchez, R.G., *Creation of a Simplified Benchmark Model for the Neptunium Sphere experiment*, LA-UR-03-8321 (2003)

[2] Veeser L.R., Phys. Rev. C **17**, 385 (1978).

[3] Frehaut J. et al., 1982 Antwerp, 78 (1982).

[4] Malinovskii V.V. et al., Sov. At. Energy **54**, 226 (1983).

[5] Boykov G.S. et al., Phys. At. Energy **57**, 2047 (1994).

[6] Mughabghab S.F. et al., *Neutron Cross Sections*, Vol.1, Part B., Academic Press, INC. (1984).

[7] MCNP is a trademark of the Regents of the University of California, Los Alamos National Laboratory.