

# Preliminary (Rev.2) Analysis of the Plutonium Thermal Solution Benchmarks from the ICSBEP Handbook

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## Introduction

Plutonium data in practically all recently released evaluated data libraries were tuned to some extent to achieve reasonable performance in real applications. In some cases, this led to departure from measured differential data, particularly the PFNS and the thermal constants that were evaluated with the Standards-2017. The libraries under consideration are:

- ENDF/B-VII.1 library (as the reference case),
- ENDF/B-VIII.0 library,
- JEFF-3.3 library,
- pu239e80p5 Trial <sup>239</sup>Pu evaluation (taking ENDF/B-VIII.0 for the rest).

The focus is on the known anomalies in the ENDF/B-VIII.0 library compared to Standards-2017, which are summarized as follows:

	Standards-2017	ENDF/B-VII.1	JEFF-3.3	ENDF/B-VIII.0	pu239e80p5
E_av(PFNS)		2.111 MeV	2.097 MeV	2.117 MeV	2.074 MeV
Sig_f	752.4+/-2.2	747.92	749.35	747.39	751.41
Sig_g	269.8+/-2.5	270.73	271.43	270.14	270.91
nu-bar_tot	2.878+/-0.0060	2.8789	2.8659	2.8745	2.8776
K1	1175.2	1172.1	1159.2	1164.0	1167.1

In order to assess the cross-section data of <sup>239</sup>Pu near thermal energies the PU-SOL-THERM benchmarks (PST for short) from the ICSBEP Handbook were analysed. The computational models for the MCNP code obtained from A. Kahler were used, whenever available. In some cases, the models from the ICSBEP Handbook were used. The aim was to perform a broad scale analysis on as many benchmarks as possible.

The purpose of the present analysis is to:

- identify benchmarks that might exhibit anomalous behaviour that should be excluded from the analysis
- mark benchmarks that might be sensitive to other materials like various absorber materials, reflectors, etc.

- select a short list of benchmarks that are representative of the broader list, which could be used for testing intermediate trial evaluations of <sup>239</sup>Pu.
- Check the adequacy of the short list proposed by the OECD/NEA SG-34.

The ICSBEP collection includes 38 benchmark groups PST001 through PST038. The list of benchmarks that were NOT included in the present work are:

- PST008 Concrete-reflected 14-inch-diameter spheres of plutonium nitrate solutions.
- PST013 Interacting cylinders of 256-mm diameter with plutonium nitrate solution (115.1gpu/l) in air (inputs available in ICSBEP).
- PST014 Interacting cylinders of 300-mm diameter with plutonium nitrate solution (115.1gpu/l) in air (inputs available in ICSBEP).
- PST015 Interacting cylinders of 300-mm diameter with plutonium nitrate solution (152.5gpu/l) in air (Inputs available in ICSBEP).
- PST016 Interacting cylinders of 300-mm and 256-mm diameters with plutonium nitrate solution (152.5 and 115.1gpu/l) and nitric acid (2n) in air (Inputs available in ICSBEP).
- PST017 Interacting cylinders of 256-mm and 300-mm diameters with plutonium nitrate solution (115.1gpu/l) in air (inputs available in ICSBEP).
- PST019 Plutonium sulfate solutions reflected by beryllium oxide and graphite PROSERPINE reactor – Saclay.
- PST020 Water-reflected and water-cadmium-reflected 14-inch diameter spheres of plutonium nitrate solutions (inputs available in ICSBEP).
- PST023 Plutonium (33.89% and 4.23% <sup>240</sup>Pu) nitrate solutions in two water-reflected cylindrical concentric tanks (inputs available in ICSBEP).
- ~~PST024 Slabs of plutonium nitrate solutions reflected by 1-inch thick plexiglas (inputs available in ICSBEP).~~
- ~~PST025 Water-reflected slabs of plutonium nitrate solutions (inputs available in ICSBEP).~~
- PST026 Unreflected slabs of plutonium nitrate solutions for which no input models were available in ICSBEP.
- PST029 Interacting (water-reflected or not) annular cylinders (50/30 cm diam.) Containing plutonium (3% <sup>240</sup>Pu) nitrate solution.
- PST030 Water-reflected annular cylinders (50/20 cm diam.) containing plutonium (1.5% <sup>240</sup>Pu) nitrate solutions.
- PST031 Plutonium (19% <sup>240</sup>Pu) nitrate solution in a water-reflected parallelepiped tank (50 × 50 cm side) poisoned by an array of hafnium plates.

Benchmarks PST027 are included in MCT011, Benchmarks PST035, PST036 and PST037 are marked as “Rejected” in the ICSBEP Handbook. Some of the above benchmarks, particularly those that have MCNP input in the ICSBEP as plain text files, will be analysed later.

## Broad assessment of the groups of benchmarks

Multiplication factors calculated from the various libraries were compared to the benchmark values and are shown in the plots in the Appendix. A summary of cases is given in Table I, indicating benchmarks that could be excluded from the list of “clean cases” with justification in the Comments column, where applicable.

Table I: List of benchmarks included in the analysis.

Benchmark	Cases	Comment
PST001	6	Water-reflected 11.5-inch diameter spheres of plutonium nitrate solutions
PST002	7	Water-reflected 12-inch diameter spheres of plutonium nitrate solutions
PST003	8	Water-reflected 13-inch diameter spheres of plutonium nitrate solutions
PST004	13	Water-reflected 14-inch diameter spheres of plutonium nitrate solutions 0.54% to 3.43% <sup>240</sup> Pu
PST005	9	Water-reflected 14-inch diameter spheres of plutonium nitrate solutions 4.05% and 4.40% <sup>240</sup> Pu
PST006	3	Water-reflected 15-inch diameter spheres of plutonium nitrate solutions
PST007	8	Water-reflected 11.5-inch-diameter spheres partly filled with plutonium nitrate solutions
PST009	3	Unreflected 48-inch-diameter sphere of plutonium nitrate solution ( <b>large over-prediction of reactivity</b> )
PST010	14	Water-reflected 9-, 10-, 11-, and 12-inch-diameter cylinders of plutonium nitrate solutions
PST011	12	Bare 16- and 18-inch-diameter spheres of plutonium nitrate solutions
PST012	23	Criticality of plutonium nitrate solution in a large water-reflected cubic tank (130 x 130 x 100 cm) (19% <sup>240</sup> Pu) ( <b>large <sup>240</sup>Pu content</b> )
PST018	9	Bare 16- and 18-inch-diameter spheres of plutonium nitrate solutions
PST021	10	Water-reflected and bare 15.2-inch-diameter spheres of plutonium nitrate solutions ( <b>only cases 1 and 3 analysed</b> )
PST022	17	Plutonium (19% <sup>240</sup> Pu) nitrate solution in a water-reflected annular cylinder tank (50/20 cm dia.) ( <b>large <sup>240</sup>Pu content</b> )
PST024	23	Slabs of plutonium nitrate solutions reflected by 1-inch-thick plexiglas ( <b>large <sup>240</sup>Pu content, correlated with PST025, 026</b> )
PST025	44	Water-reflected slabs of plutonium nitrate solutions ( <b>large <sup>240</sup>Pu content, correlated with PST024, 026</b> )
PST026	7	Unreflected slabs of plutonium nitrate solutions (cases 1, 2, 3, 4, 9, 15 and 17) ( <b>large <sup>240</sup>Pu content, correlated with PST024, 025</b> )
PST028	9	Water-reflected annular cylinders (50/30 cm diam.) Containing plutonium (3% <sup>240</sup> Pu) nitrate solutions
PST032	17	Water-reflected annular cylinders (50/20 cm diam.) Containing plutonium (9.95% <sup>240</sup> Pu) nitrate solutions ( <b>large <sup>240</sup>Pu content</b> )
PST033	63	Water-reflected cylinders of plutonium (3.13 or 4.23% <sup>240</sup> Pu) nitrate solutions, poisoned with borated pyrex tubes or raschig rings and not poisoned ( <b>very large differences from benchmark values that exceed expected deviations due to nuclear data</b> )
PST034	15	Plutonium (8.3 wt.% <sup>240</sup> Pu) nitrate solution with gadolinium in water- reflected 24-inch diameter cylinder ( <b>the presence of Gd masks the performance of Pu</b> )
PST038	5	Plutonium temperature effect program - low concentrated (20, 15 or 14.3 g/l) plutonium nitrate solutions at room temperature ( <b>large <sup>240</sup>Pu content</b> )

A plot of the differences from the calculated multiplication factors and the benchmark values is shown in Figure 1 as a function of the energy of the average lethargy causing fission. Benchmark groups PST024, PST025, PST026 and PST033 were excluded because they introduce a significant bias.

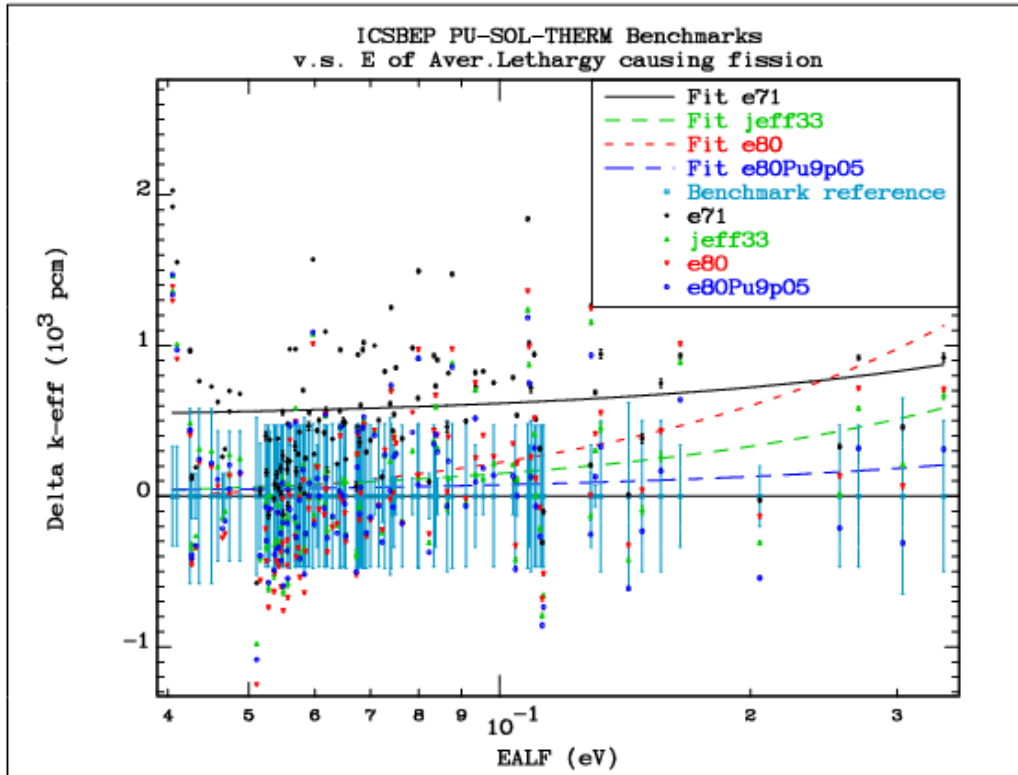


Figure 1: Broad assessment of the library performance on the plutonium thermal solution benchmarks from the ICSBEP Handbook (excluding PST033), displayed as a function of the energy corresponding to the average lethargy causing fission (because the “above thermal fission fraction” ATLF is not available for all benchmarks).

## Selection of a short list of representative benchmarks

In the OECD/WPEC SG-34 report in Table 2 a short list of thermal solution benchmarks was identified to be representative for the testing of the PFNS of plutonium data. The list is given in Table II. It was noted that in Table 1 of the OECD/WPEC SG-34 report an alternative short list of thermal solution benchmarks for general testing of  $^{239}\text{Pu}$  data is given, which does not coincide with the present list. The combined list is given in Table IIa, with the addition of two cases from more recent Valduc experiments (namely PST038-003 and PST038-004) to complement the older Valduc experiments (PST012-005 and PST012-13), which had much larger quoted uncertainties; both Valduc benchmarks contain a significant fraction (~20%) of  $^{240}\text{Pu}$ . The differences between the calculated multiplication factors and benchmark values for the benchmarks from Table IIa are shown in Figure 2. The choice of the PST009 benchmark might require a more detailed investigation due to the large discrepancy with the trend from the other benchmarks.

Table II: Short list of PU-SOL-THERM benchmarks proposed by WPEC-SG34 Table 2.

No.	ICSBEP Label	Short name	Common name
1	PU-SOL-THERM-001	pst001-001	PNL (11.5) 2210-1
2	PU-SOL-THERM-001	pst001-004	PNL (11.5) 2164-4
3	PU-SOL-THERM-001	pst001-006	PNL (11.5) 2218-6
4	PU-SOL-THERM-004	pst004-005	P11 (14) -05
5	PU-SOL-THERM-005	pst005-001	P11 (14) Pu0-1
6	PU-SOL-THERM-005	pst005-007	P11 (14) Pu0-7
7	PU-SOL-THERM-006	pst006-002	P11 (15) -2
8	PU-SOL-THERM-007	pst007-003	PNL (11.5) p-03
9	PU-SOL-THERM-007	pst007-010	PNL (11.5) p-10
10	PU-SOL-THERM-012	pst012-005	Valduc-05
11	PU-SOL-THERM-012	pst012-013	Valduc-13

Table IIa: Combined Short list of PU-SOL-THERM benchmarks proposed by WPEC-SG34 Table 1 and 2.

No.	ICSBEP Label	Short name	Common name
1	PU-SOL-THERM-001	pst001-001	PNL (11.5) 2210-1
2	PU-SOL-THERM-001	pst001-004	PNL (11.5) 2164-4
3	PU-SOL-THERM-001	pst001-006	PNL (11.5) 2218-6
4	PU-SOL-THERM-004	pst004-001	P11 (14) -01
5	PU-SOL-THERM-004	pst004-005	P11 (14) -05
6	PU-SOL-THERM-005	pst005-001	P11 (14) Pu0-1
7	PU-SOL-THERM-005	pst005-007	P11 (14) Pu0-7
8	PU-SOL-THERM-006	pst006-002	P11 (15) -2
9	PU-SOL-THERM-007	pst007-003	PNL (11.5) p-03
10	PU-SOL-THERM-007	pst007-010	PNL (11.5) p-10
11	PU-SOL-THERM-009	pst009-003a	PNL-48R-3a
12	PU-SOL-THERM-012	pst012-005	Valduc-05
13	PU-SOL-THERM-012	pst012-010	Valduc-10
14	PU-SOL-THERM-012	pst012-013	Valduc-13
15	PU-SOL-THERM-018	pst018-006	PNL-11-6
16	PU-SOL-THERM-034	pst034-003	PNL-10-Gd-03
17	PU-SOL-THERM-034	pst034-014	PNL-10-Gd-14
18	PU-SOL-THERM-038	pst038-003	Valduc_loPu-03
19	PU-SOL-THERM-038	pst038-004	Valduc_loPu-04

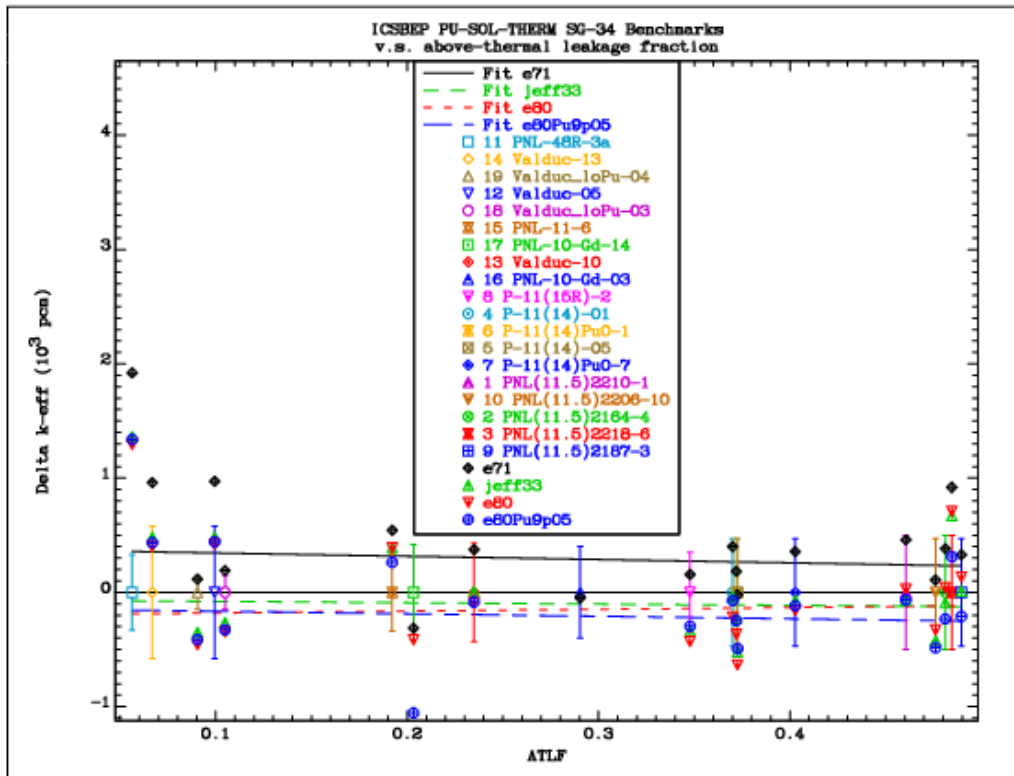


Figure 2: Assessment of the library performance on the WPEC-SG34 selection of representative plutonium thermal solution benchmarks from the ICSBEP Handbook.

## Conclusions

The broad assessment of the plutonium thermal solution benchmarks with new libraries (excluding PST033) shows an overall decrease in the predicted reactivity compared to the ENDF/B-VII.1, without any significant trend with increasing spectrum hardness (EALF) or with the above-thermal leakage fraction (ATLF).

The choice of representative benchmarks by WPEC-SG34 seems reasonable, but there is some ambiguity in the benchmarks with very soft spectra, considering the older Valduc benchmarks PST012 (labelled Valduc-05 and Valduc-13 in Figure 2; see also the Appendix) that have rather large uncertainties and the more recent Valduc measurements PST038 (labelled Valduc\_loPu-03 and Valduc\_loPu-04 in Figure 2) with much smaller uncertainties, for which the predicted reactivity with all libraries is much smaller.

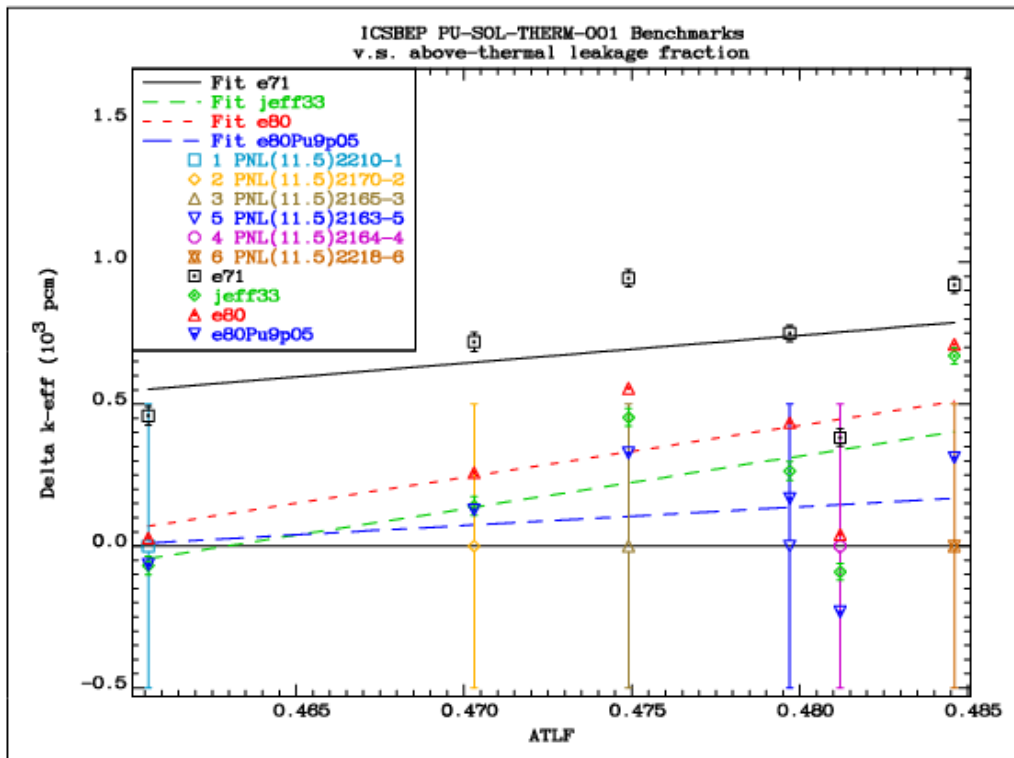
The softer PFNS of  $^{239}\text{Pu}$ , evaluated with the Standards-2017 (which is not a Standard itself) results in a significant increase of reactivity, particularly in the high-leakage systems). In the trial evaluation “pu239e80p5” this increase was effectively compensated by adjusting the ENDF/B-VIII.0  $^{239}\text{Pu}$  evaluation to agree with the thermal constants evaluated together with Standards-2017 and an increase of the capture-to-fission ratio in the first resonance of  $^{239}\text{Pu}$ .

## Appendix

Assessment of the benchmarks by group is shown below.

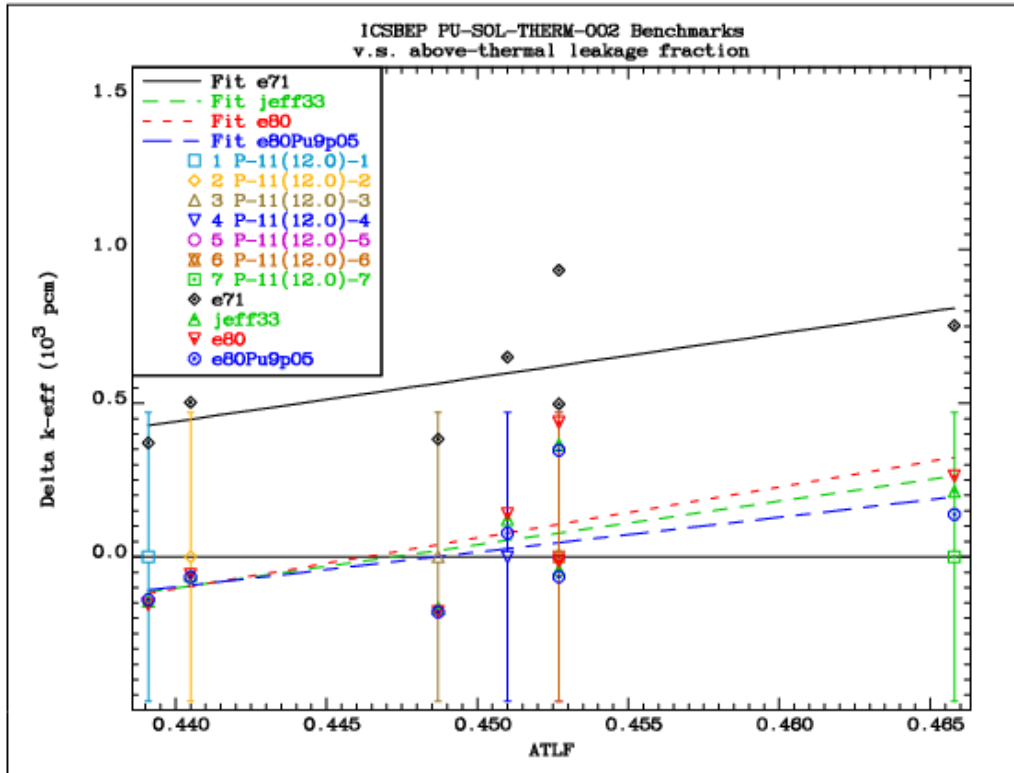
### PU-SOL-THERM-001

The benchmarks represent “water-reflected 11.5-inch diameter spheres of plutonium nitrate solutions” from the Battelle Pacific Northwest Laboratories. The over-prediction of reactivity with ENDF/B-VII.1 data is eliminated with ENDF/B-VIII.0 and JEFF-3.3 data to practically within the uncertainty band and almost no gradient as a function of ATLF.



## PU-SOL-THERM-002

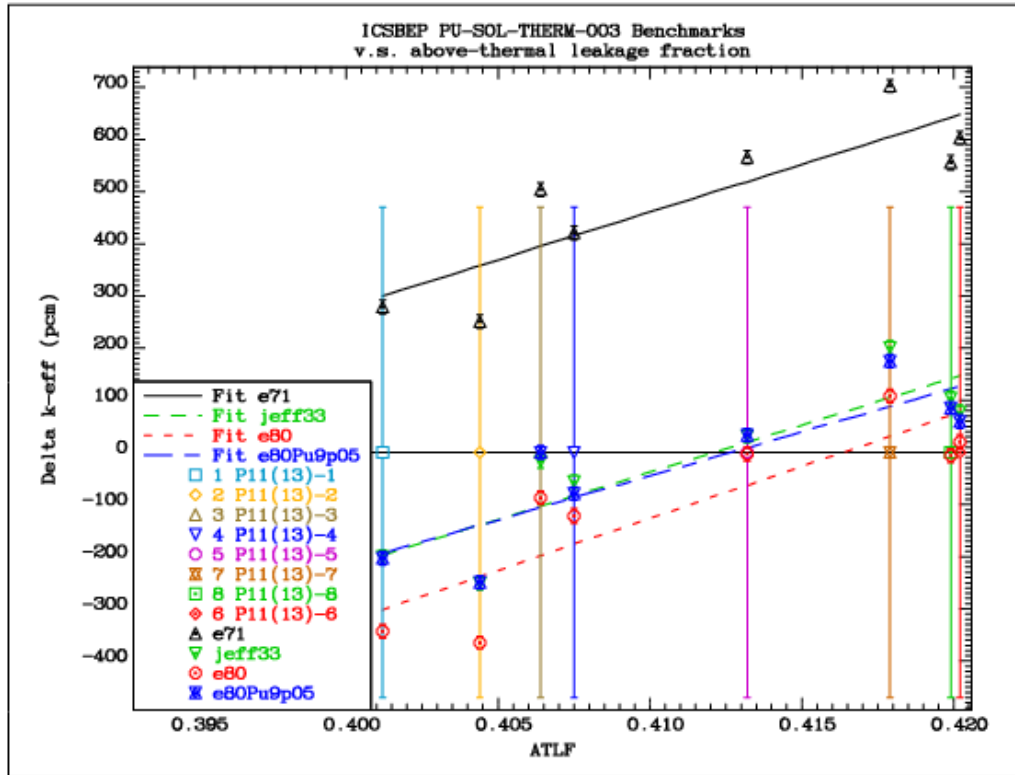
The benchmarks represent “water-reflected 12-inch diameter spheres of plutonium nitrate solutions” from the P-11 area of the Hanford Reservation. There is a marginally small positive gradient with ATLF. The results are very similar to the PST001 results.





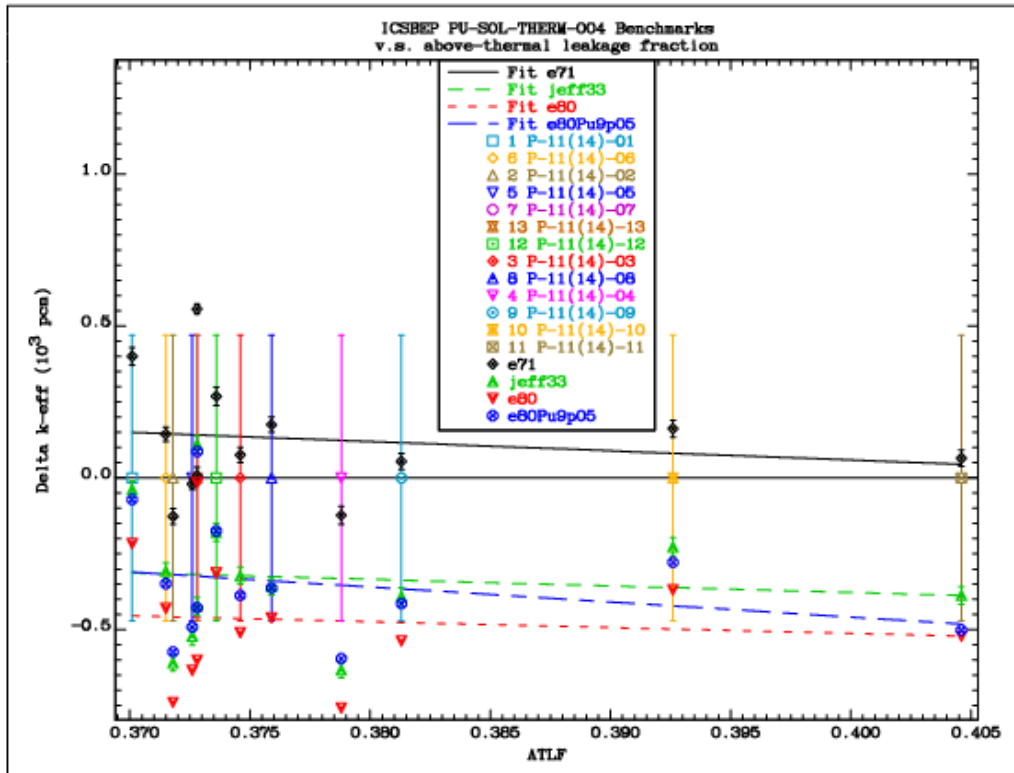
## PU-SOL-THERM-003

The benchmarks represent “water-reflected 13-inch diameter spheres of plutonium nitrate solutions” from the P-11 area of the Hanford Reservation. The benchmarks are similar to PST002, except for the larger sphere diameter. The results are very similar to the PST002 results.



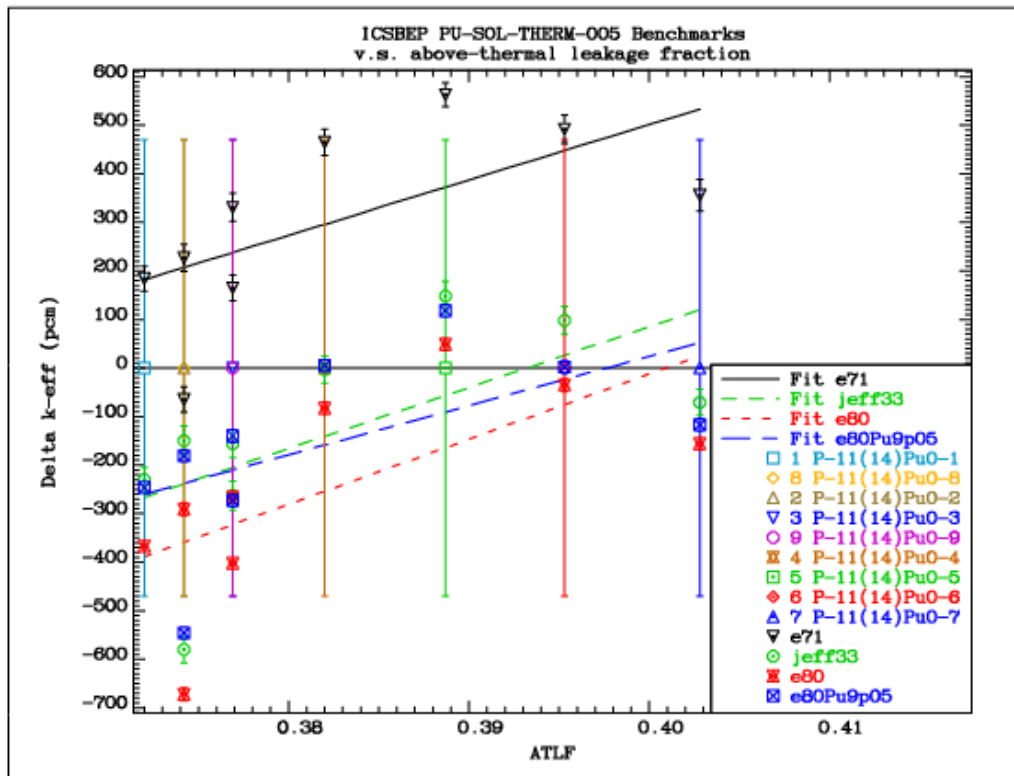
## PU-SOL-THERM-004

The benchmarks represent “water-reflected 14-inch diameter spheres of plutonium nitrate solutions with 0.54% to 3.43%  $^{240}\text{Pu}$ ” from the P-11 area of the Hanford Reservation. The benchmarks are similar to PST002, except for the larger sphere diameter and different  $^{240}\text{Pu}$  concentration. The predicted reactivity is systematically lower, but remains very close to the uncertainty band with all libraries. There is practically no gradient with ATLF.



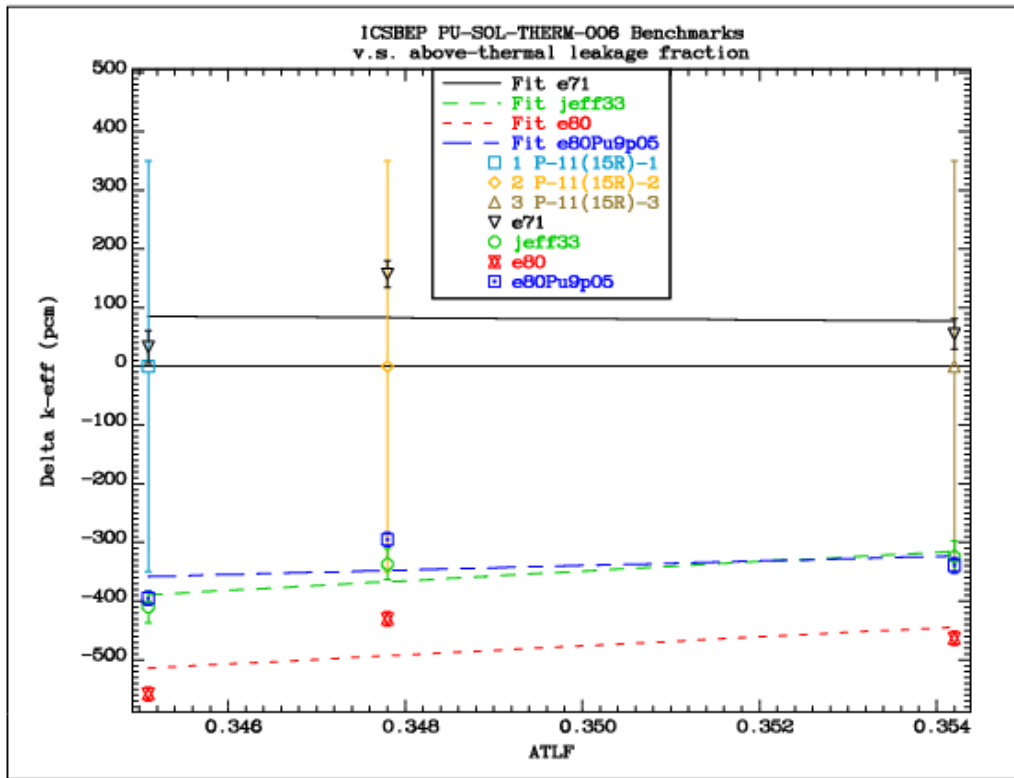
## PU-SOL-THERM-005

The benchmarks represent “water-reflected 14-inch diameter spheres of plutonium nitrate solutions with 4.05% and 4.40%  $^{240}\text{Pu}$ ” from the P-11 area of the Hanford Reservation. The main difference from PST004 is the higher  $^{240}\text{Pu}$  content. There is a small positive gradient with ATLF.



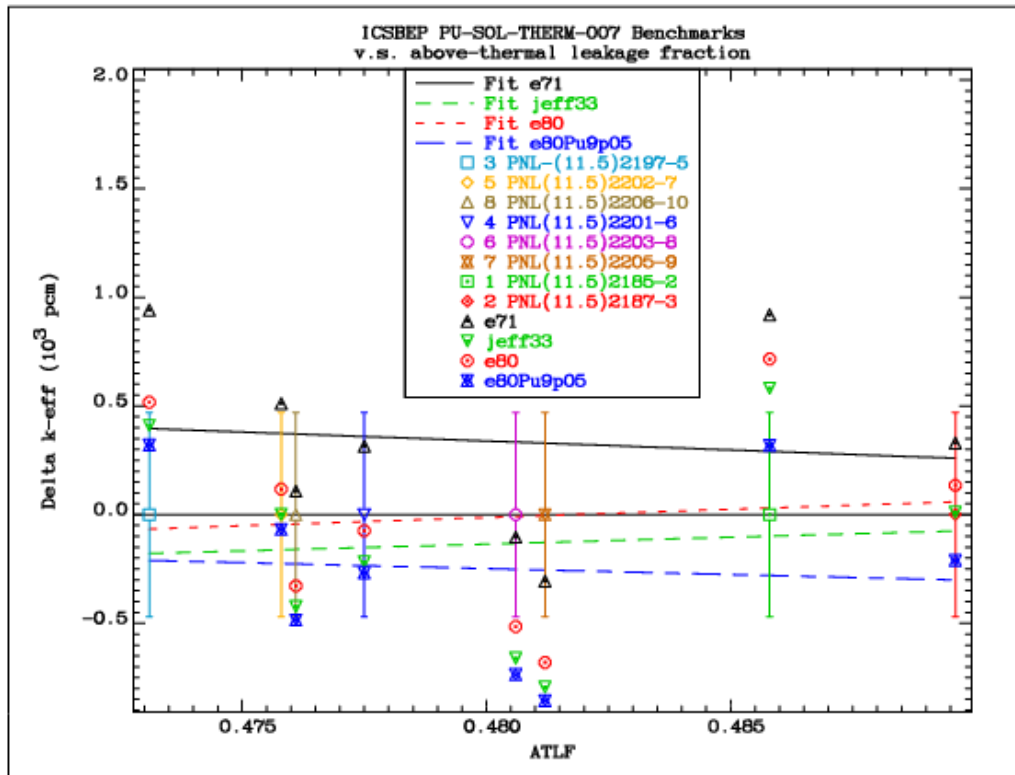
## PU-SOL-THERM-006

The benchmarks represent “water-reflected 15-inch diameter spheres of plutonium nitrate solutions” from the P-11 area of the Hanford Reservation. The benchmarks are similar to PST002, except for the larger sphere diameter. Only three cases are included in this group. There is practically no gradient with ATLF.



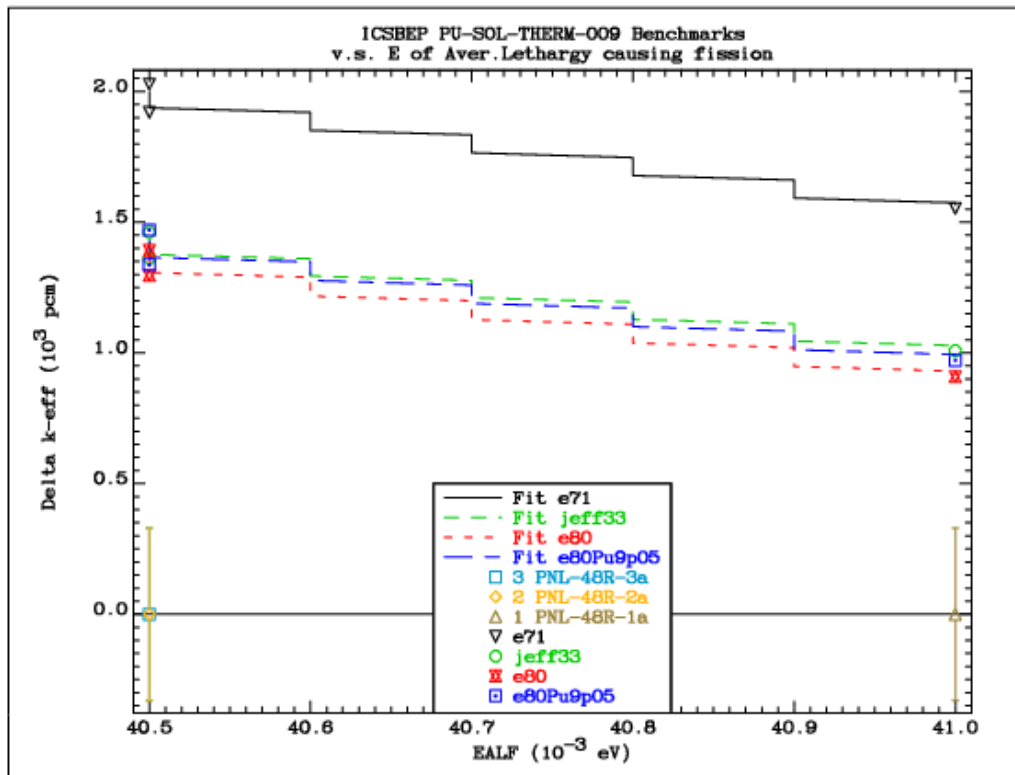
## PU-SOL-THERM-007

The benchmarks represent “water-reflected 11.5-inch-diameter spheres partly filled with plutonium nitrate solutions” from the Battelle Pacific Northwest Laboratories. The scattering in the calculated results is slightly higher. There is practically no gradient with ATLF. Other characteristics are similar to those observed in the other benchmarks listed above.



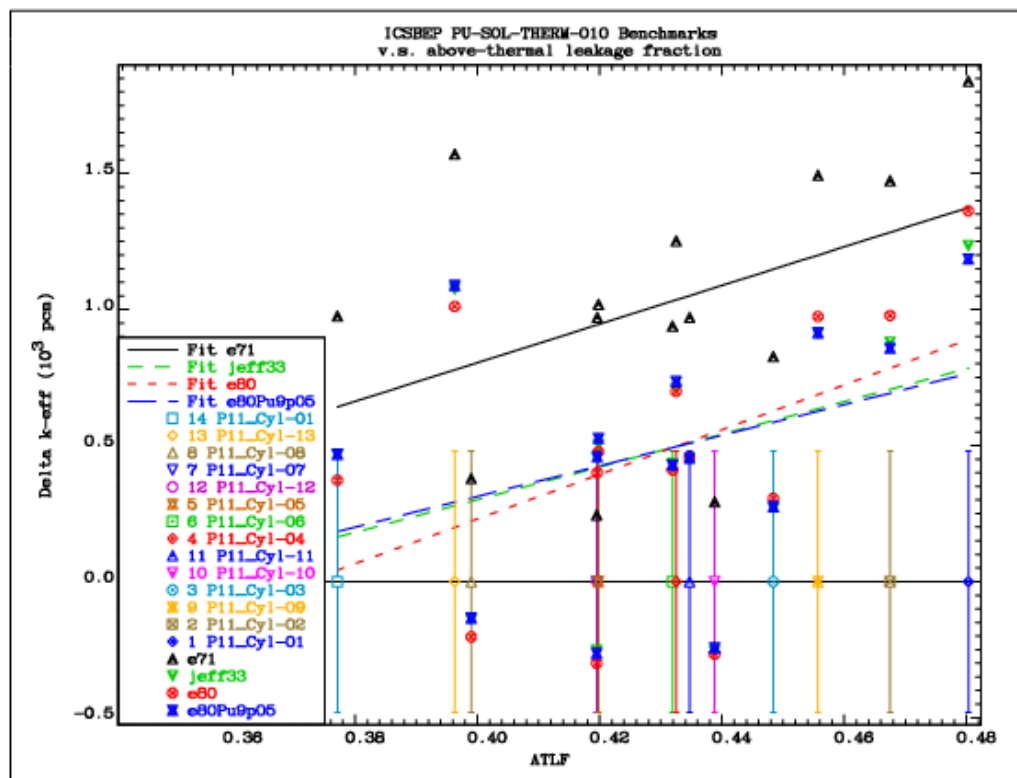
## PU-SOL-THERM-009

The benchmarks represent “unreflected 48-inch-diameter sphere of plutonium nitrate solution” from the Battelle Pacific Northwest Laboratories. Three cases are available. The benchmark involved nearly one cubic metre of solution. The calculated reactivity of all three cases is overpredicted by 1000 pcm to 2000 pcm. In view of the results with other benchmarks, the specifications for this group of benchmarks should be checked.



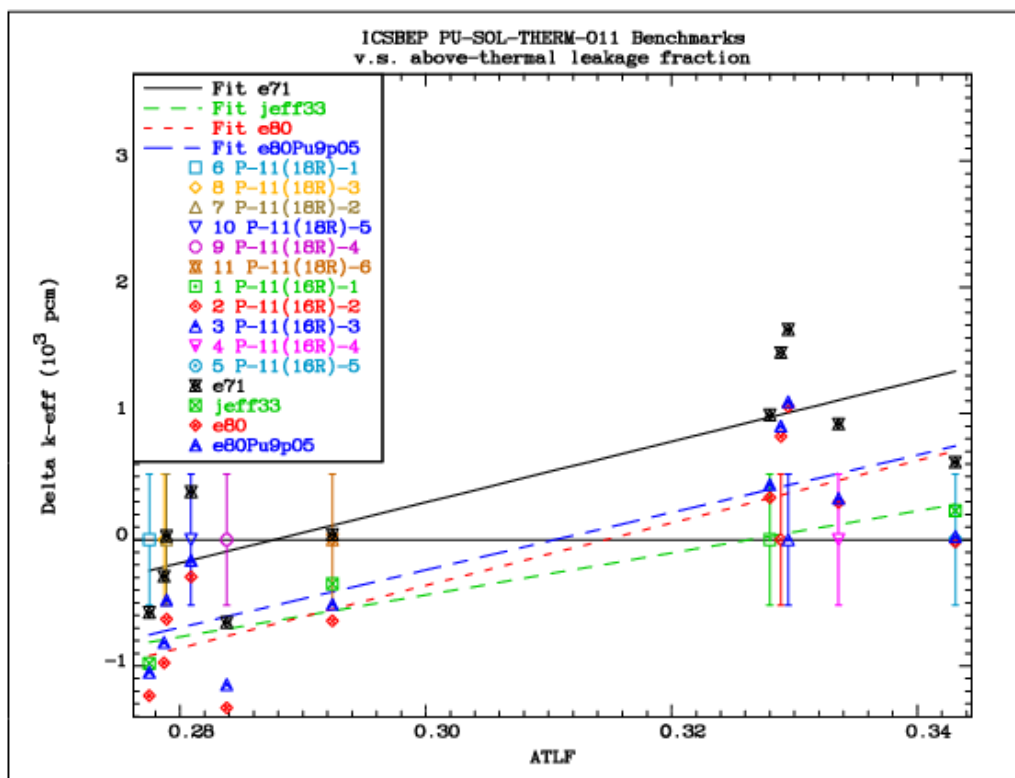
## PU-SOL-THERM-010

The benchmarks represent “water-reflected 9-, 10-, 11-, and 12-inch-diameter cylinders of plutonium nitrate solutions” from the P-11 area of the Hanford Reservation. The scattering in the calculated results compared to the benchmark values is very large and exceeds 1500 pcm in some cases, while a number of other cases are predicted within the quoted experimental uncertainties. This observation is surprising, especially since several other benchmarks from the same facility discussed above do not show such anomalies. The measurements were conducted in the early 1950’s; the benchmark specifications might need a review.



## PU-SOL-THERM-011

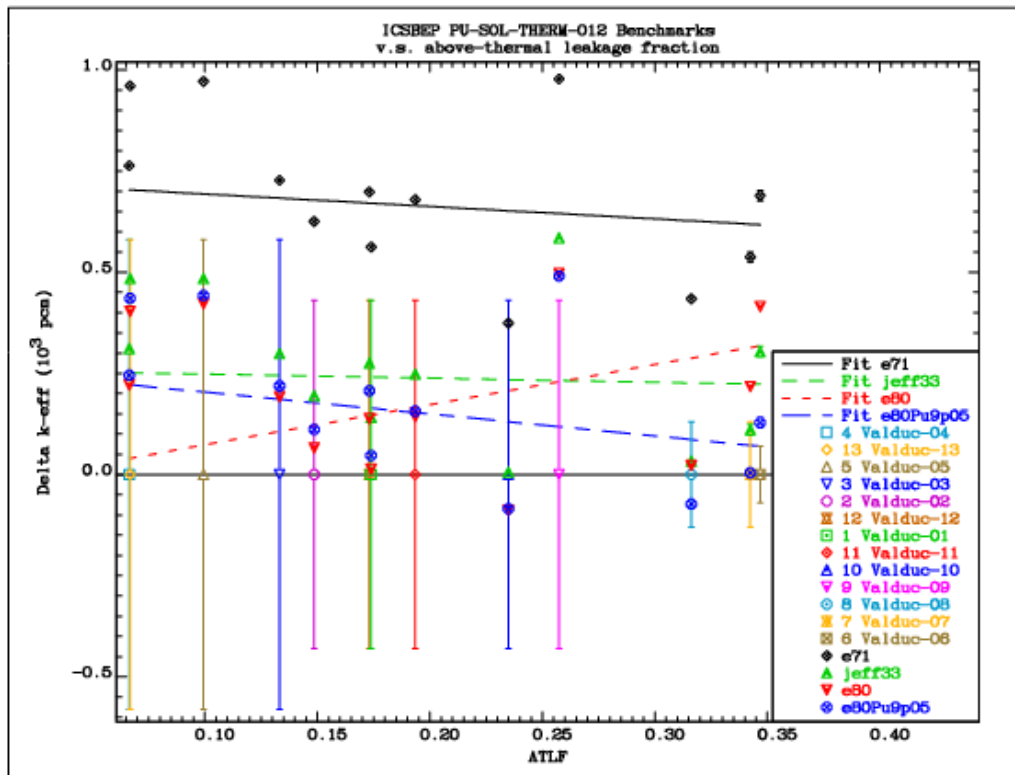
The benchmarks represent “bare 16- and 18-inch-diameter spheres of plutonium nitrate solutions” from the P-11 area of the Hanford Reservation. The larger spheres have a lower ATLF. The calculated results underpredict reactivity with all libraries (except ENDF/B-VII.1); on the contrary, the calculations generally overpredict reactivity with all libraries (more so with ENDF/B-VII.1 data). The measurements were conducted in the early 1950’s; the benchmark specifications might need a review.





## PU-SOL-THERM-012

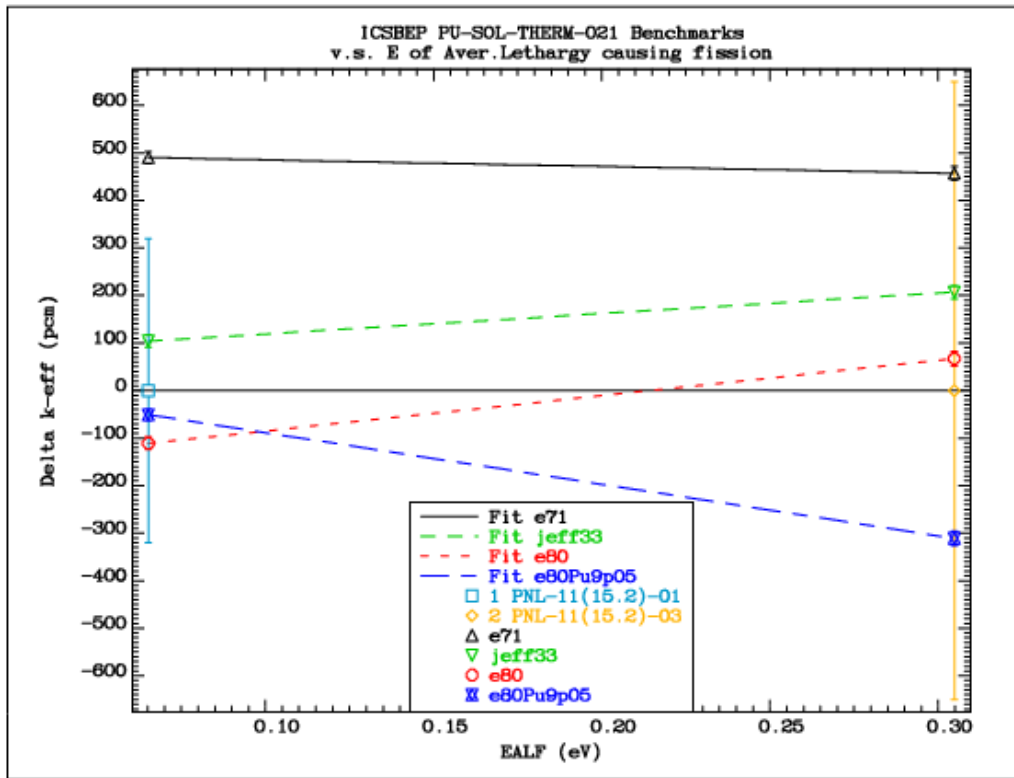
The benchmarks represent “criticality of plutonium nitrate solution in a large water-reflected cubic tank (130 x 130 x 100 cm) (19% 240pu)” from the VALDUC facility (CEA-FRANCE), conducted in 1974. Cases 4 and 13 exhibit a very low ATLF. Cases 6, 7 and 8 have the highest ATLF and very low quoted uncertainties. The results with all libraries are in reasonable agreement with the benchmark values, except ENDF/B-VII.1, which systematically overpredicts reactivity of these benchmarks.





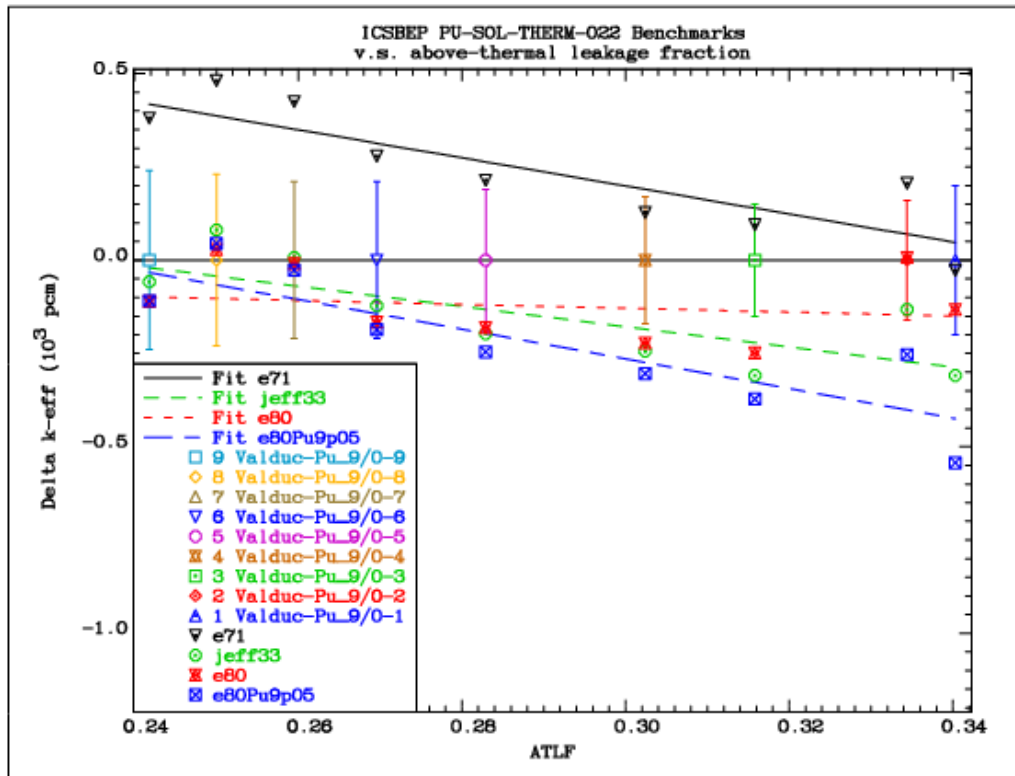
## PU-SOL-THERM-021

The benchmarks represent “water-reflected and bare 15.2-inch-diameter spheres of plutonium nitrate solutions” from the Battelle Pacific Northwest Laboratories. Input models are available for Cases 1 and 3 only. The results with all libraries are in reasonable agreement with the benchmark values.



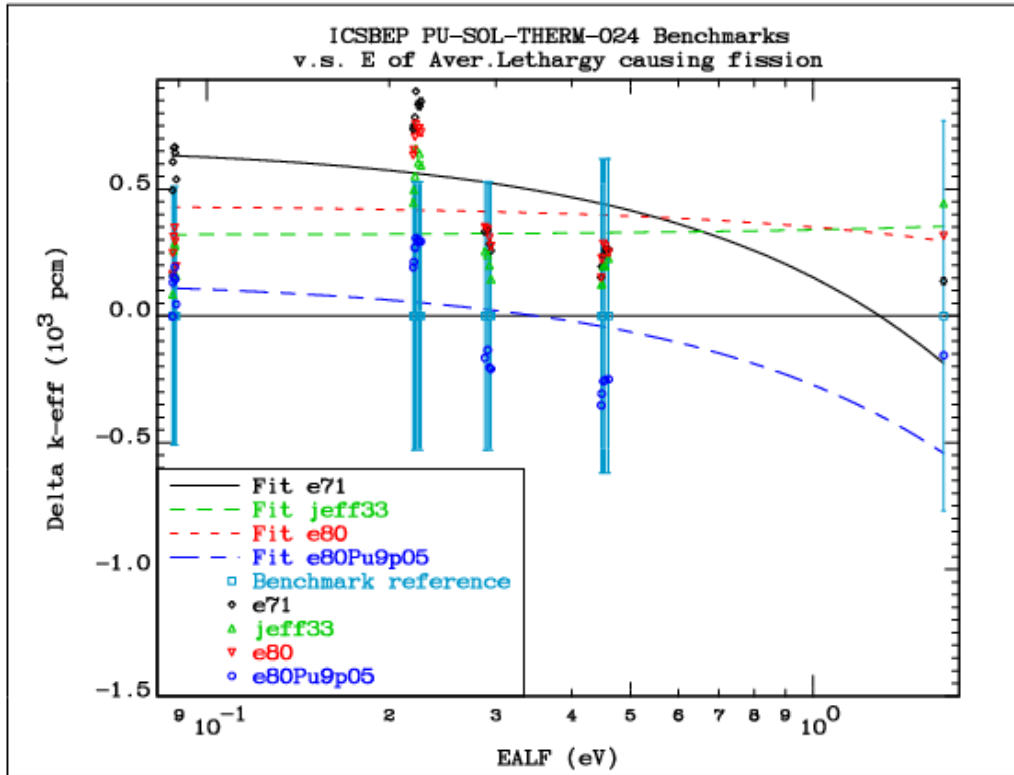
## PU-SOL-THERM-022

The benchmarks represent “plutonium (19%  $^{240}\text{Pu}$ ) nitrate solution in a water-reflected annular cylinder tank (50/20 cm dia.)” from the Valduc facility (CEA-FRANCE), conducted in 1973. The assemblies contain a central void and inserts of paraffin and cadmium. The same plutonium nitrate was also used to perform experiments in a large cubic tank (PU-SOL-THERM-012). Similar experiments performed in the Valduc facility with 50/30 and 50/20 annular cylinders (with diameters 50 and 30, 20 cm) and different  $^{240}\text{Pu}$  contents are presented in PU-SOL-THERM-028, 029, 030, 031 and 032. The results with all libraries show a negative gradient with ATLF.



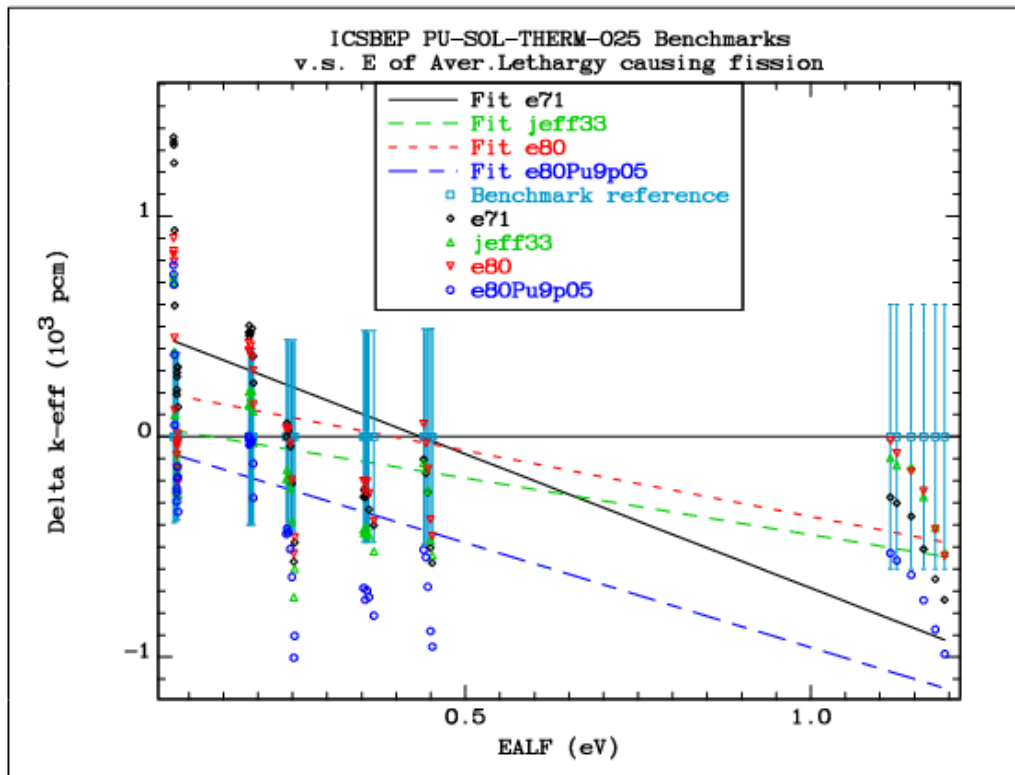
## PU-SOL-THERM-024

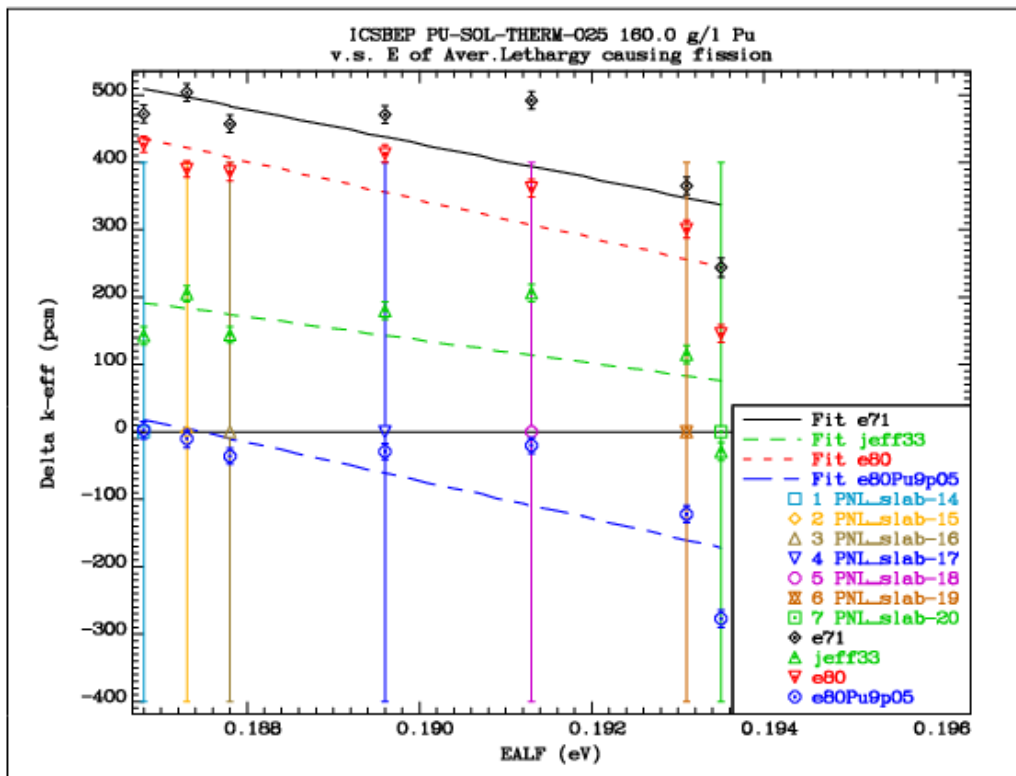
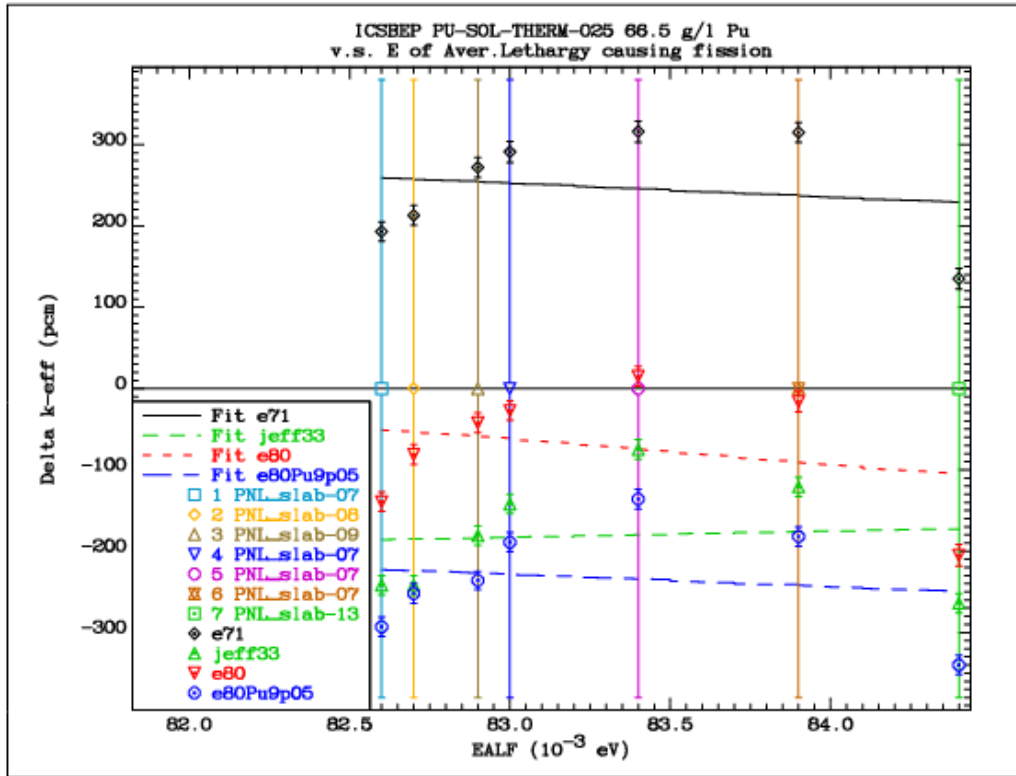
The benchmarks represent “slabs of plutonium nitrate solutions reflected by 1-inch-thick plexiglas” from the Battelle-Pacific Northwest Laboratory. The results with all libraries are in reasonable agreement with the benchmark values.

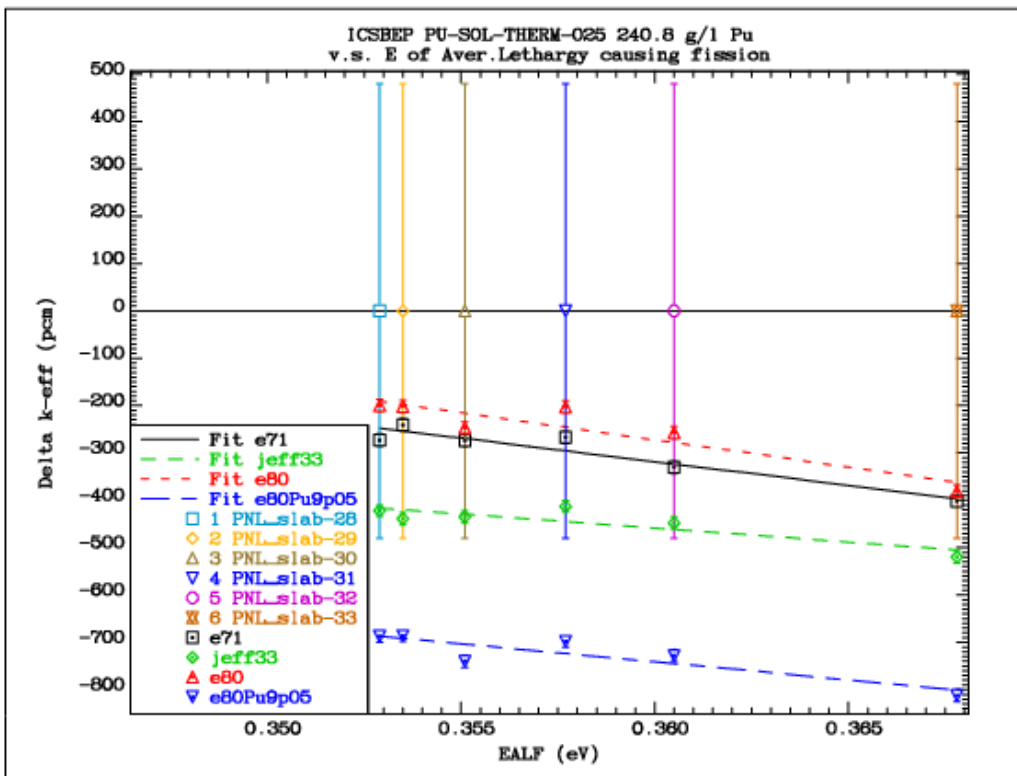
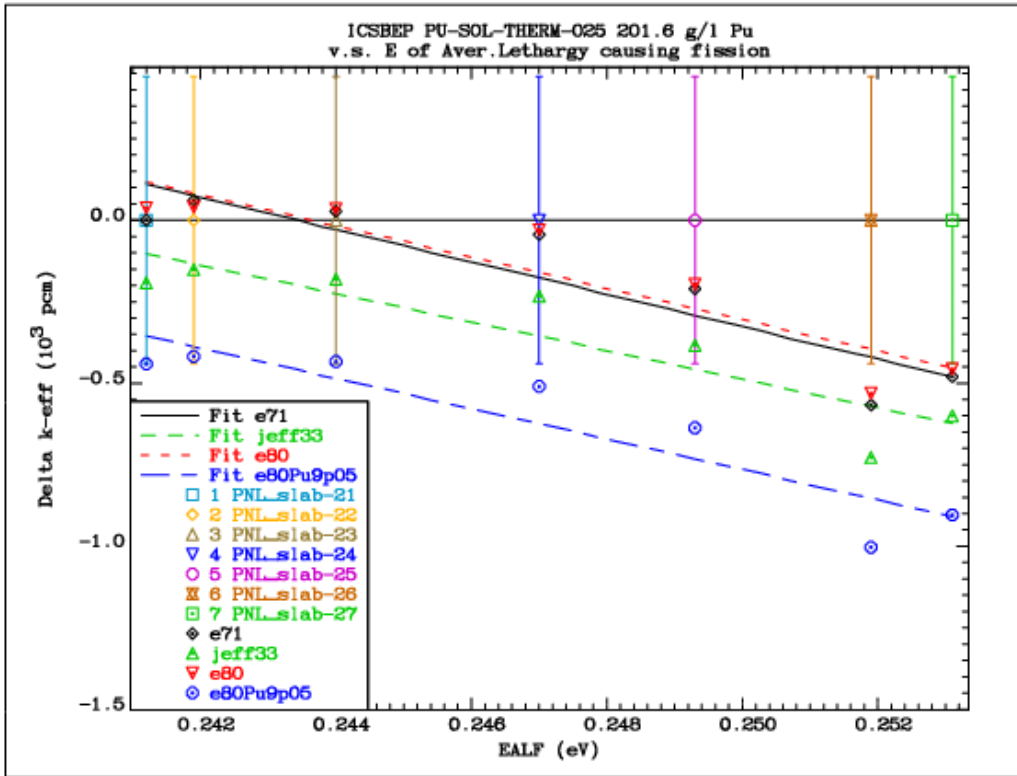


## PU-SOL-THERM-025

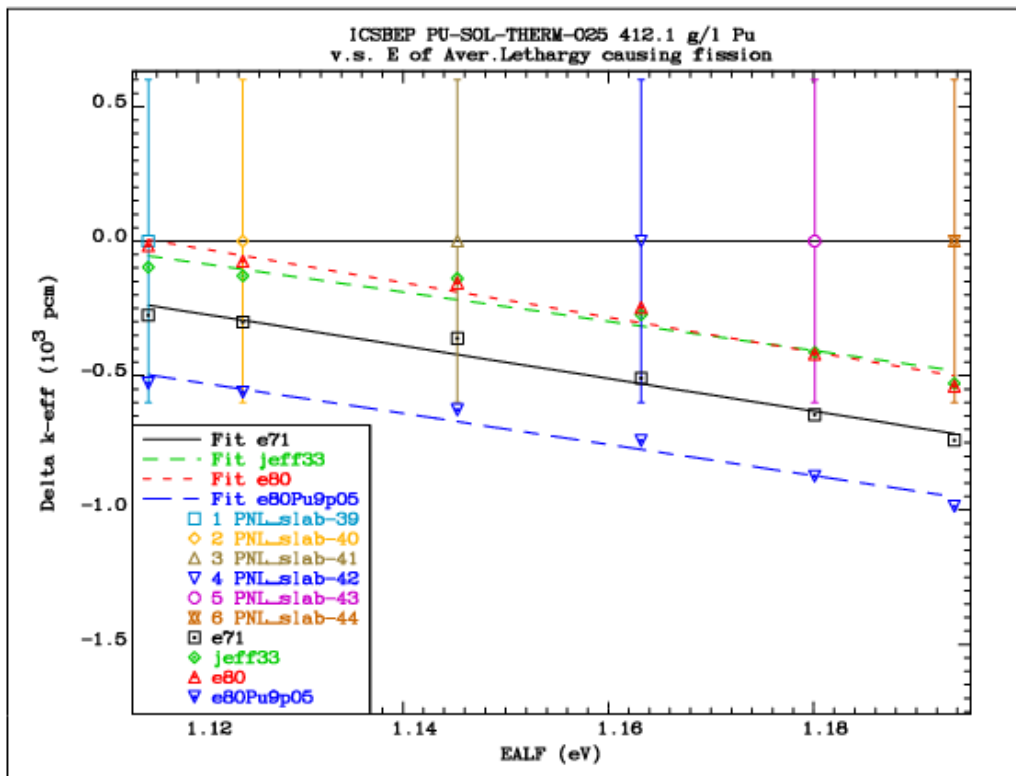
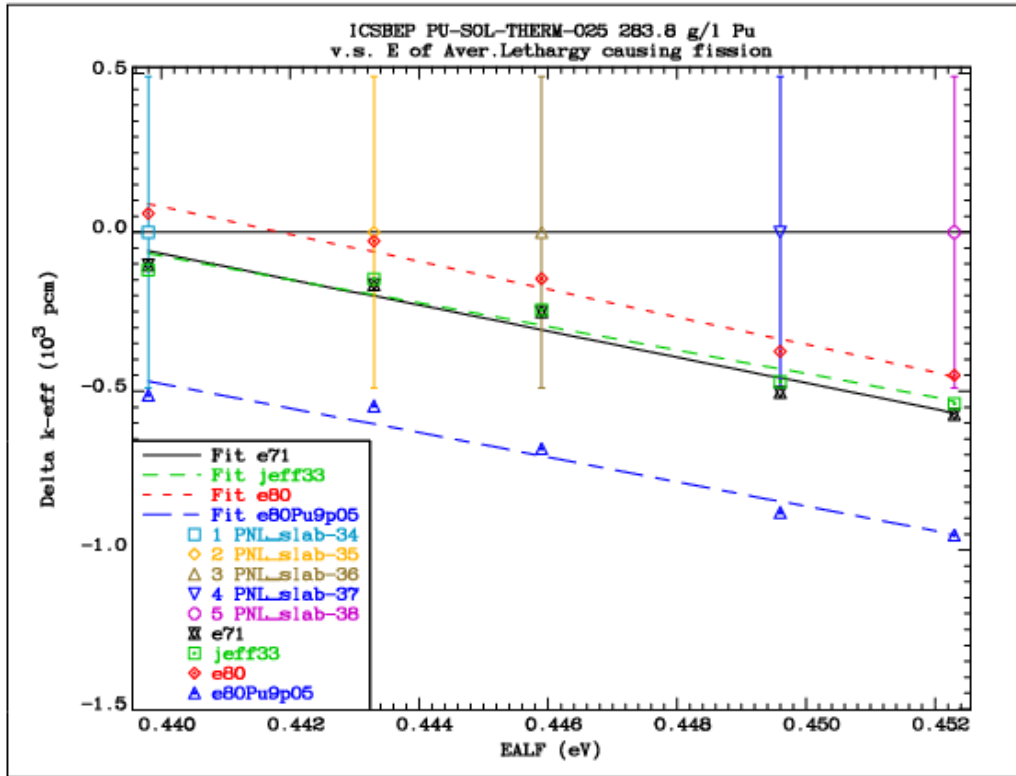
The benchmarks represent “water-reflected slabs of plutonium nitrate solutions” from the Battelle-Pacific Northwest Laboratory. The 44 cases in this benchmark group can be split into sub-groups, differing by the Pu concentration and  $^{240}\text{Pu}$  content. Separate plots usually exhibit a negative gradient with EALF, but in the overall results for the benchmarks it is clear that the EALF spectral parameter is not the only factor that characterises this group of benchmarks. The results are not understood, especially considering the similar benchmarks from the same facility in PST024 and PST026.





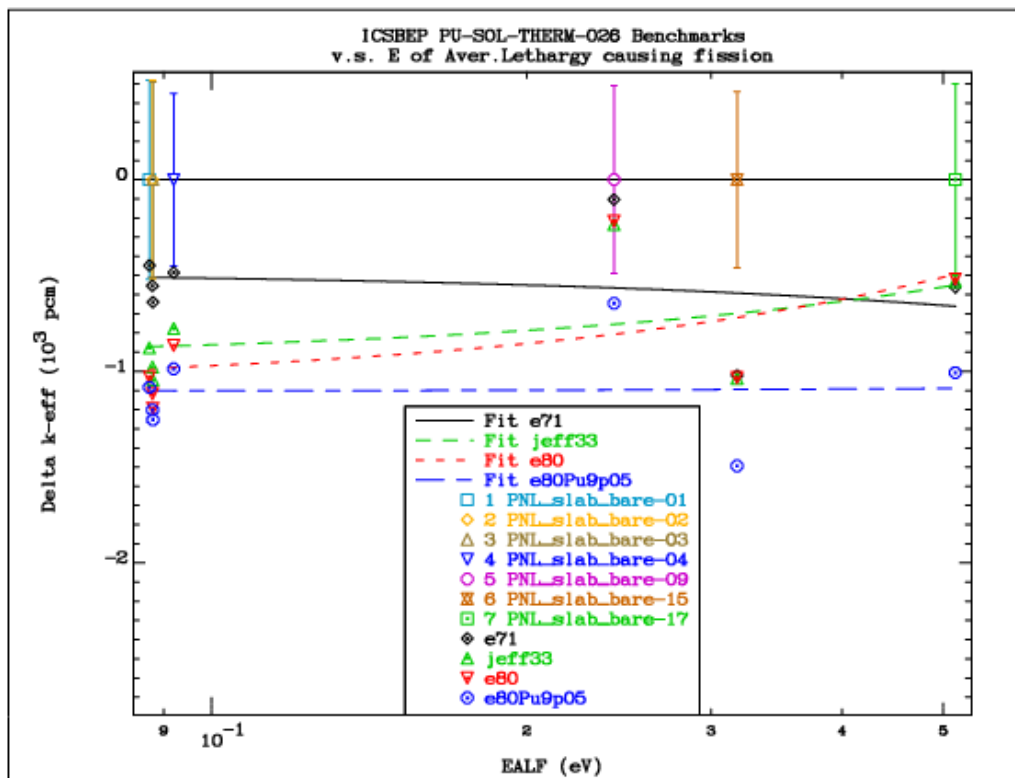






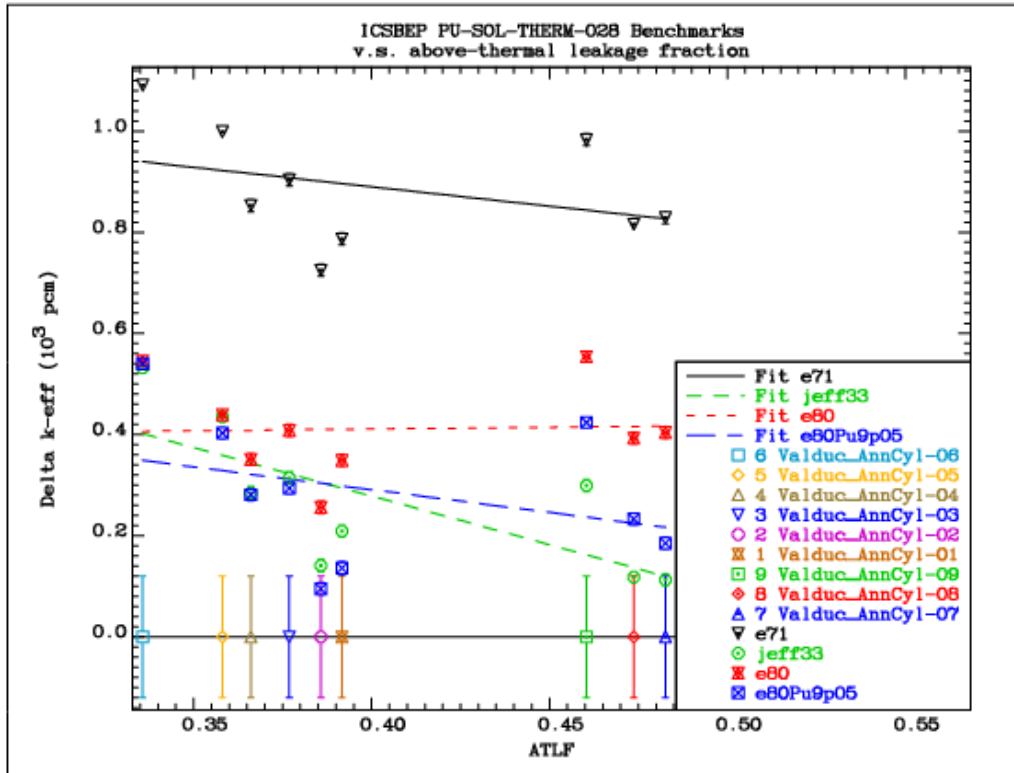
## PU-SOL-THERM-026

The benchmarks represent “unreflected slabs of plutonium nitrate solutions” from the Battelle-Pacific Northwest Laboratory. Reactivity is strongly underpredicted with all libraries, which might indicate a deficiency in the modelling of the benchmark, since it is very likely highly sensitive to room-return.



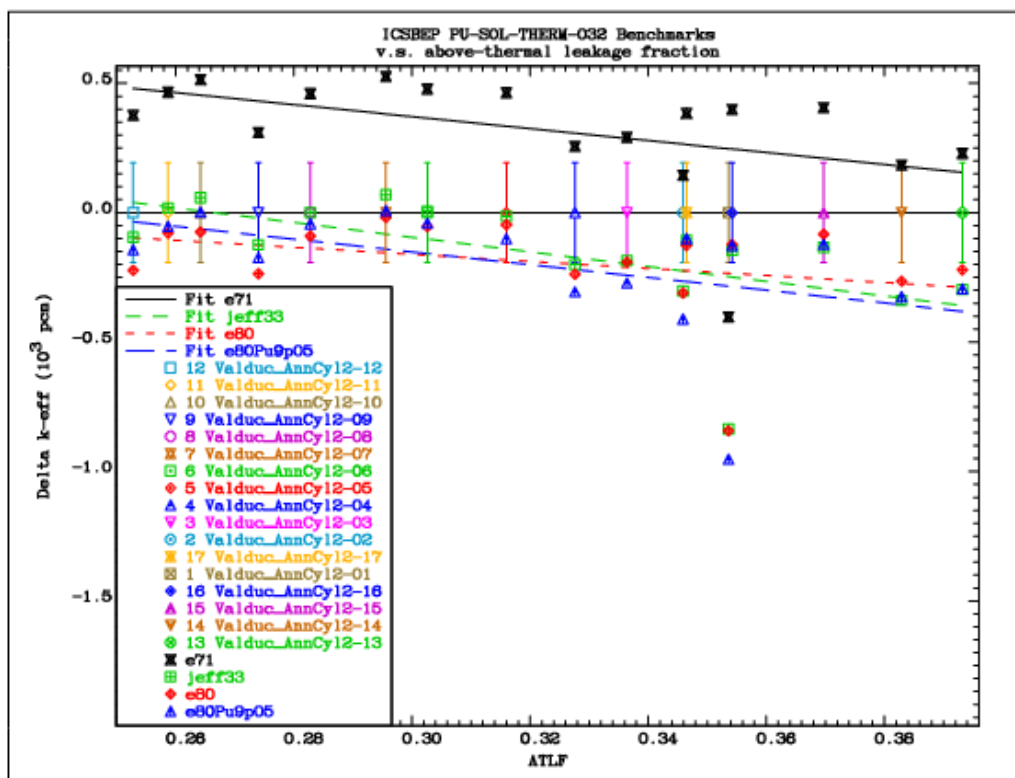
## PU-SOL-THERM-028

The benchmarks represent “water-reflected annular cylinders (50/30 cm diam.) containing plutonium (3% 240pu) nitrate solutions” from the Valduc critical facility, conducted between 1963 and 1976. Reactivity is strongly overpredicted with all libraries. The reason is not understood.



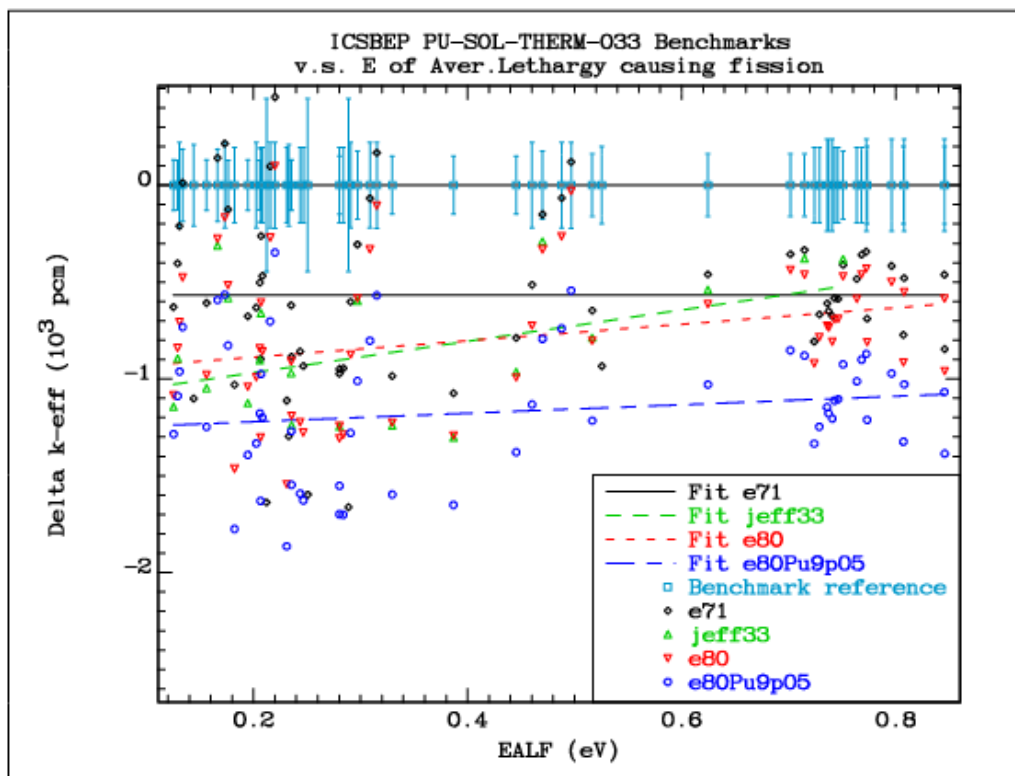
## PU-SOL-THERM-032

The benchmarks represent “water-reflected annular cylinders (50/20 cm diam.) containing plutonium (9.95% 240pu) nitrate solutions” from the Valduc critical facility conducted between 1963 and 1976. Contrary to the observation in similar PST028 benchmarks on the same facility, the predicted reactivity is much lower with all libraries; the negative gradient with ATLF is similar.



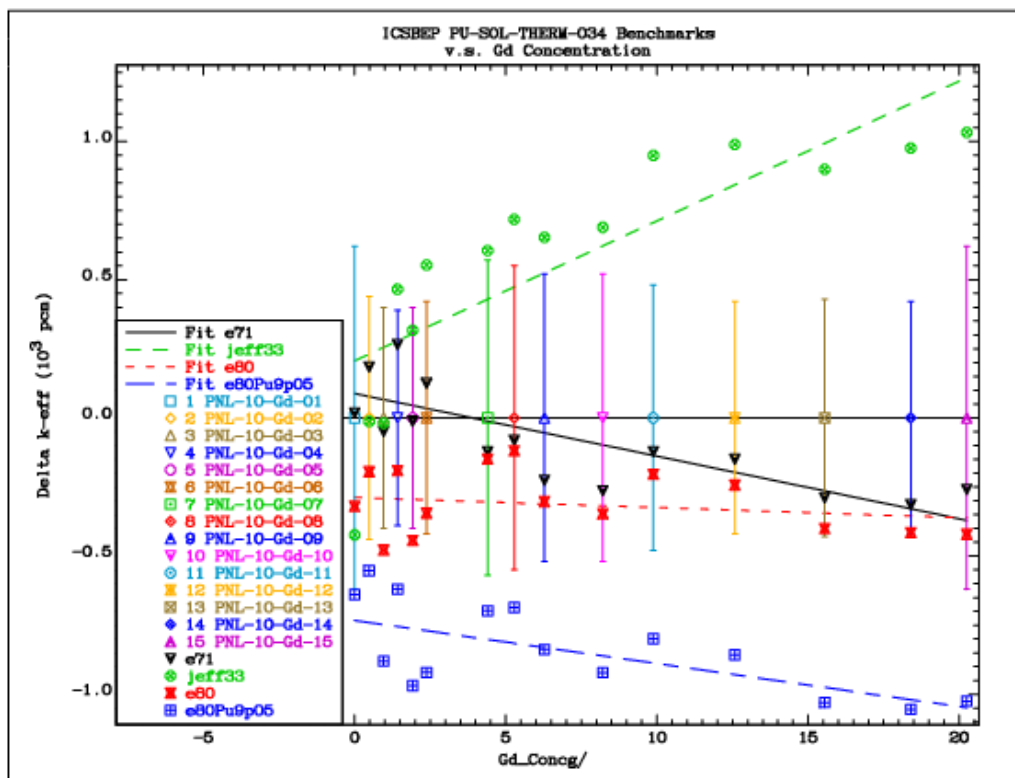
### PU-SOL-THERM-033

The benchmarks represent “water-reflected cylinders of plutonium (3.13 or 4.23% 240pu) nitrate solutions, poisoned with borated pyrex tubes or raschig rings and not poisoned” from the Valduc facility (C.E.A. France) conducted in 1966-1968. The group includes 63 cases and the quoted uncertainties are very small, but the scattering of the results is enormous. No sensible conclusions can be extracted from these benchmarks.



## PU-SOL-THERM-034

The benchmarks represent “plutonium (8.3 wt.% 240pu) nitrate solution with gadolinium in water-reflected 24-inch diameter cylinder” from the Battelle Pacific Northwest Laboratories. This group of benchmarks tests the dependence on gadolinium concentration. Note the distinct difference in the trends using ENDF/B-VIII.0 and JEFF-3.3 libraries. The changes in the trial “e80Pu9p05” library increase the sensitivity to gadolinium, which results in a decrease of reactivity with increasing gadolinium concentration. Note that all libraries predict reactivity within the experimental uncertainties in the first case that contains no gadolinium.



## PU-SOL-THERM-038

The benchmarks represent “plutonium temperature effect program - low concentrated (20, 15 or 14.3 g/l) plutonium nitrate solutions at room temperature” from the Valduc facility. Only the benchmarks at room temperature are included in this group. The benchmark is similar to PST012, except that it spans a lower range of ATLF and has a much lower assigned uncertainty.

