Annular Cylinders Experimental Programme Containing Plutonium Solutions At Different [240]Pu Contents

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Abstract

From 1963 to 1976, 730 critical experiments dealing with annular cylinders containing plutonium nitrate solutions were conducted on Valduc critical facility, called "Apparatus B". They aimed at validating critical configurations encountered in the fuel cycle, especially in storage and also at validating the [240]Pu cross-sections in thermal neutron spectrum. It is to be noticed that these experiments validate criticality codes either in configurations with reactor-grade plutonium coming from the reprocessing cycle or with weapon-grade plutonium coming from the decommissioning of nuclear weapons.

KEYWORDS: critical experiments, annular cylinder, plutonium nitrate

1. Introduction

The use of annular cylinders was historically introduced with the aim to store a greater quantity of plutonium nitrate than in classical cylindrical tanks without using neutron absorbers (such as Raschig rings, borosilicate tubes (Pyrex trademark)).

Amongst the 730 critical experiments performed at Valduc critical facility, a selection of 83 ones representative of the range of the plutonium nitrate [240]Pu content, the annular cylinder diameter and the interacting material inside the cylinder has been done. The corresponding experiments have been evaluated. They are judged acceptable as benchmark experiments and have therefore been introduced in the I.C.S.B.E.P. Handbook [1] [1] (so called PU-SOL-THERM-022, 028, 029, 030, 032). Another selection of experiments has been planned and should be evaluated in PU-SOL-THERM-031. The selected 83 experiments involve 500×200 ($\phi o / \phi i^2$) and 500×300 annular cylinders reflected by 30 cm water on the side and at the bottom. The inner part contains air, water, paraffin and/or cadmium. The cylinders could be interacting (PU-SOL-THERM-029). The annular part of the cylinders contains a slightly acid plutonium nitrate solution. A wide range of plutonium concentration was examined (roughly from 20 g/l to 170 g/l). Four values of [240]Pu content were investigated: 1.5%, 3%, 10% or 19% and cover the whole range found in the fuel cycle. The experiments are based on the sub-critical approach technique, so that the final k_{eff} is approximately within -65.10^{-5} from the criticality, with critical conditions obtained by

extrapolation. Criticality is reached through the water and solution level increase.

The uncertainties associated with these experiments have been carefully determined. On the whole, the total uncertainties remain lower than 0.25% at the 1 σ level for the majority of configurations. They are mainly influenced by the uncertainties on acidity, plutonium concentration and radius of cylinder.

The benchmark experiment k_{eff} have been calculated with APOLLO2-MORET 4 codes (part of the CRISTAL French criticality safety package [2]).

A slight general tendency to overestimate k_{eff} (0.4% on average for a [240]Pu content between 3 and 19% and 1% for a [240]Pu content of 1.5%) has been highlighted running the benchmarks with APOLLO2-MORET 4 [3] codes with no visible trend either with plutonium concentration or with [240]Pu content. Further investigations are in progress. They deal with the comparison with experiments coming from other laboratories (to avoid a possible experimental bias) and the use of other cross-section libraries (JEFF3.1 and ENDF-BVI).

2. Description of the experiments

2.1 Experimental configuration

Apparatus B, used to carry out the experiments, comprised the 500×200 or a 500×300 annular cylinder containing the fissile solution placed inside the parallelepipedic reflector pool containing the water reflector. For interacting cylinders, they were two interacting 500×300 annular cylinders inside the reflector pool. This apparatus was inside a sealed cell, which was located inside a large room. The control room was adjacent, separated by 145-cm-thick concrete wall.

2.1.1 The room

Apparatus B was set in a sealed steel cell, 800 cm long, 600 cm wide and 600 cm high. Walls were made of large steel plates, 2×2 meters and 0.2 cm thick.
The cell was built and approximately centered in a larger room, $12.1 \times 8.8 \times 10$ meters high, with 1.45-meter-thick concrete walls. The thickness of the concrete floor was 40 cm. The thickness of the ceiling varied from 70 cm (side) to 110 cm (middle).

2.1.2 The reflector pool

The reflector pool, parallelepiped-shaped, was made of stainless steel and contained the water reflector. Its internal dimensions were 320 cm $\times$ 210 cm and 150 cm high. Walls and bottom were 0.4 cm thick. Sidewalls and bottom were reinforced with I-shaped girders, which were respectively 8×4.2 cm on the sides and 12×5.8 cm below the bottom. The pool was equipped with a 'needle of measurement,' which followed the free upper level of the water.
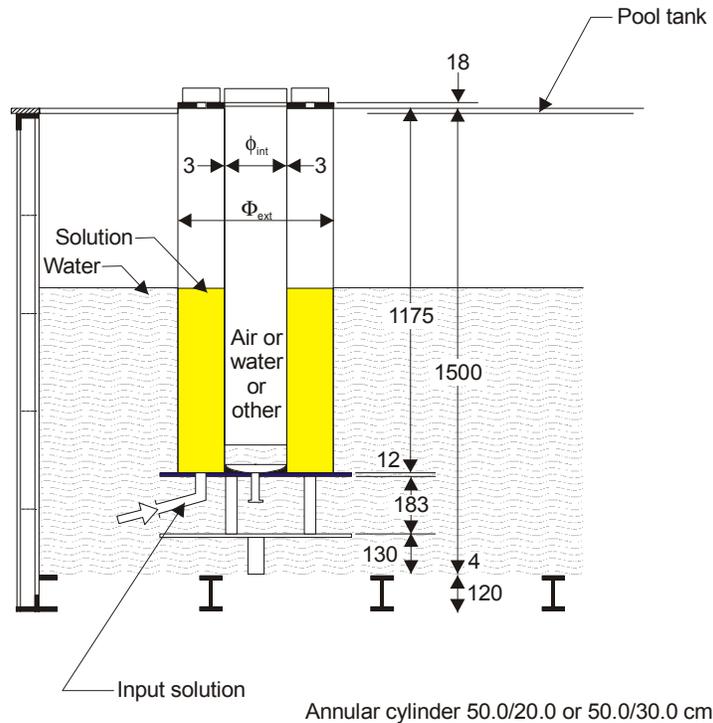
2.1.3 The annular cylinder

The experimental configuration described in Fig.1 consisted of an annular cylinder containing the nitrate solution. It was made of two concentric tubes having inside diameters of 50 cm and 19.4 cm for 500×200 annular cylinder and 50 cm and 29.4 cm for 500×300

annular cylinders, 120.1-cm external height, and a 0.3-cm wall thickness. These tubes were welded to a bottom plate (thickness varying from 1.8 cm to 1.2 cm) and to a 1.8-cm-thick top plate. The top plate had 8 holes of 6 cm diameter, at 45°, centered on a circle 40 cm in diameter. The inner part was either empty (air), or contained water, paraffin and/or cadmium. The cadmium sheet (0.07 or 0.08 cm thick) was flatten against the inner surface of the annular cylinder. The annular cylinder was supported with support feet; the total height of the feet was 31.3 cm. The tubes, plates, and support feet were made of stainless steel. The annular cylinder was equipped with a needle of measurement.

The nitrate solution was pumped in at the bottom of the annular cylinder, and the water reflector was pumped in at the bottom of the pool. The top surfaces of the water (outer reflector and inner part) and of the nitrate solution were at the same level. The varying thickness of the bottom plate of the annular cylinder, from 1.8 cm thick at one side to 1.2 cm thick at the other, provided a small slope to direct the draining solution toward the inlet tube. The "zero" level of the critical measurements was 1.40 cm above the outer annular cylinder bottom face.

For all configurations, both nitrate-solution and water-reflector temperatures were about 21°C.

Figure 1: Experimental Configuration of an Annular Cylinder (500×200 or 500×300 cm).

Annular cylinder 50.0/20.0 or 50.0/30.0 cm

other: Paraffin + Cd, Cd + air or Cd + water or water + Cd + air

2.2 Chemical data

2.2.1 Plutonium isotopic vectors and concentrations

Different solutions are contained in the annular cylinders. Their isotopic vectors are provided in Table 1. The ranges of plutonium concentrations are the followings:

  ➢ from 28.7 to 165 g/l for the 500×200 annular cylinder with 19% ^{240}Pu,
  ➢ from 23.95 to 62.1 g/l for the 500×200 annular cylinder with 9.95% ^{240}Pu,
  ➢ from 34.9 to 80.6 g/l for the 500×300 annular cylinder with 3% ^{240}Pu,
  ➢ 40.6 g/l for the two 500×300 interacting annular cylinders with 3% ^{240}Pu,
  ➢ from 21.87 to 51.21 g/l for the 500×200 annular cylinder with 1.5% ^{240}Pu.

The inner part of the annular cylinders is occupied by:

  ➢ air or paraffin surrounded by a cadmium sheet (0.08 cm thick) for the 500×200 annular cylinder with 19% ^{240}Pu,
  ➢ air or water for the 500×200 annular cylinder with 9.95% ^{240}Pu,
  ➢ air, water or a sandwich of water + cadmium sheet (0.07 cm thick) + air for the 500×300 annular cylinder with 3% ^{240}Pu,
  ➢ air or water or air and a cadmium sheet (0.07 cm thick) for the two 500×300 interacting annular cylinders with 3% ^{240}Pu,
  ➢ air or water or air and a cadmium sheet (0.08 cm thick) for the 500×200 annular cylinder with 1.5% ^{240}Pu,

Table 1: Plutonium Isotopic Vectors [1]

Cylinders ^{240}Pu %	Analysis date	Sample	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
500×200, 19% ^{240}Pu	October 01, 1973	Atom %		74.2	18.88	5.78[3]	1.14	0.61
500×200, 9.95% ^{240}Pu	March 03, 1966	Weight %	0.05	88.6	9.95	1.28	0.09	
500×300, 3% ^{240}Pu	March 03, 1966	Weight %		96.64	3.22	0.14		
500×300 – interaction 3% ^{240}Pu		Weight %		97.02	2.98			
500×200, 1.5% ^{240}Pu	April 16, 1965	Weight %		98.47	1.50	0.03		

2.2.2 Impurities and material density

Solutions impurities were also determined. Chemical analyses performed before sub-critical approaches gave the impurity contents of the solutions in ppm (impurity weight per plutonium weight in units of 10^{-6}). The main impurity is iron (from 47 to 3400 mg/l).

The free acidity of the solutions were measured and found near 2N (2 moles H$^+$/l).
The cadmium (8.65 g/cm^3) is natural and possible impurities are neglected.

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[3] As the ^{241}Pu amount is not negligible, the decay of ^{241}Pu in ^{241}Am is considered.

Water impurities are not provided.

The paraffin composition $C_{25}H_{52}$ and the density equal to 0.90 g/cm^3 were provided by the contacted experimentalist, J. M. Honoré (private communication, June 1995).

3. Results

The 83 benchmark experiments k_{eff} were calculated using CRISTAL V1.0 criticality safety package [2]. The validation of CRISTAL V1.0 with these 83 benchmark experiments is based on the difference between the benchmark k_{eff} (1.0000) and the calculated k_{eff}.

The validation work consists in calculating the C-E value [4], which is given by calculated k_{eff} minus benchmark k_{eff}, and its combined standard deviation ($\sigma = \sqrt{\sigma_{calculation}^2 + \sigma_{benchmark}^2}$).

Calculated k_{eff} are considered in good agreement with the benchmark k_{eff} when the discrepancies are in the uncertainties ranges (depending on the combined standard deviation and on the confidence interval).

The calculations were performed using the standard route APOLLO2-MORET 4 associated with the CEA93 V6 172-energy group cross-section library based on JEF2.2 evaluation of the CRISTAL criticality safety package.

3.1 500 × 200 annular cylinder with 19% ^{240}Pu

The k_{eff} results are given in Table 2. The cases with air in the inner part of the cylinder should be differentiated from the cases with cadmium + paraffin inside it. In the first case, the k_{eff} is overestimated on average by 368.10^{-5}. In the second case, it is underestimated by 284.10^{-5}. Given the experimental uncertainty values (1σ), the k_{eff} difference is lower than three times the combined standard deviation. The results are found satisfactory.

Table 2: k_{eff} results for 500 × 200 annular cylinder with 19% ^{240}Pu

Case N°	Benchmark k_{eff}	Experimental uncertainties in 10^{-5} (1 σ)	C(Pu), g/liter	Critical height Hc (cm)	Material inside the annular cylinder	Average calculated k_{eff} (standard deviation in pcm^4)
1 to 9	1.0000	240	28.7 to 152.0	34.31 to 93.34	Air	1.00368 (137)
10 to 17	1.0000	240	53.7 to 165.0	51.46 to 92.09	Cd + Paraffin	0.99716 (178)

3.2 500 × 200 annular cylinder with 9.95% ^{240}Pu

The k_{eff} results are given in Table 3. The cases with air in the inner part of the cylinder should be differentiated from the cases with water inside it. In the first case, the k_{eff} is overestimated on average by 521.10^{-5}. In the second case, it is overestimated by 286.10^{-5}.

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[4] 1 pcm = 10^{-5}

Given the experimental uncertainty values (1σ), the k_{eff} difference is lower than three times the combined standard deviation. The results are found satisfactory.

Table 3: k_{eff} results for 500×200 annular cylinder with 9.95% ^{240}Pu

Case N°	Benchmark k_{eff}	Experimental uncertainties in 10^{-5} (1 σ)	C(Pu), g/liter	Critical height Hc (cm)	Material inside the annular cylinder	Average calculated k_{eff} (standard deviation in pcm)
2 to 12	1.0000	193	23.95 to 57.35	29.5 to 72.4	Air	1.00521 (102)
13 to 17	1.0000	193	33.3 to 50.55	34.0 to 54	Water	1.00286 (119)

3.3 500×300 annular cylinder with 3% ^{240}Pu

The k_{eff} results are given in Table 4. The cases with air in the inner part of the cylinder should be differentiated from the cases with water or air + water and air + cadmium + water inside it. In the first case, the k_{eff} is overestimated on average by 846.10^{-5}. In the second case, it is overestimated by 591.10^{-5}. In the third case, it is overestimated by 488.10^{-5}. Given the experimental uncertainty values (1σ), the k_{eff} difference is higher than three times the combined standard deviation. A tendency to overestimation is then observed.

Table 4: k_{eff} results for 500×300 annular cylinder with 3% ^{240}Pu.

Case N°	Benchmark k_{eff}	Experimental uncertainties in 10^{-5} (1 σ)	C(Pu), g/liter	Critical height Hc (cm)	Material inside the annular cylinder	Average calculated k_{eff} (standard deviation in pcm)
1 to 6	1.0000	120	34.9 to 80.6	42.60 to 97.95	Air	1.00846 (149)
7 to 9	1.0000	120	60.5 to 80.6	55.37 to 91.94	Water	1.00591 (98)
10 to 15	1.0000	120	80.6	45.01 to 69.74	A+W, W+A, A+Cd+W W+Cd+A W+Cd+W	1.00488 (342)

A+W: air + water, W+A: water + air, A+Cd+W: air + cadmium + water, W+A+Cd: water + air + cadmium, W+Cd+A: water + cadmium + air, W+Cd+W: water + cadmium + water

3.4 Two 500×300 interacting annular cylinders with 3% ^{240}Pu

The k_{eff} results are given in Table 5. The cases with air in the inner part of a cylinder should be differentiated from the cases with air/water, water or air + cadmium inside it. In the first case, the k_{eff} is overestimated on average by 395.10^{-5}. In the second case, it is overestimated by 400.10^{-5}. In the third case, it is underestimated by 302.10^{-5}. In the fourth case, it is overestimated by 632.10^{-5}. Given the experimental uncertainty values (1σ), the k_{eff} difference

is higher than three times the combined standard deviation only for annular cylinders with air + cadmium in their inner parts. Globally, good results are obtained. However, a tendency to overestimation is observed.

Table 5: k_{eff} results for the two 500×300 interacting annular cylinders with 3% ^{240}Pu

Case N°	benchmark k_{eff}	Experimental uncertainties in 10^{-5} (1 σ)	C(Pu), g/liter	Critical height Hc (cm)	Material inside the annular cylinder	Interaction distance D in mm	Average calculated k_{eff} (standard deviation in pcm)
1 to 5	0.9989	250	40.6	48.60 to 71.72	Air	10 to 300	1.00395 (79)
13 to 17	0.9989	250	40.6	50.06 to 72.17	Air/water	10 to 300	1.00400 (164)
6 to 7	0.9989	250	40.6	54.00 to 91.80	Water	10 to 35	0.99698 (217)
8 to 10	0.9989	250	40.6	57.53 to 89.5	Air + Cd	10 to 300	1.00632 (82)

Air/water: Air inside one of the two annular cylinders and water inside the second annular cylinder

3.5 500×200 annular cylinder with 1.5% ^{240}Pu

The k_{eff} results are given in Table 6. The cases with air in the inner part of the cylinder should be differentiated from the cases with water, or air + cadmium inside it. In the first case, the k_{eff} is overestimated on average by 1109.10^{-5}. In the second case, it is overestimated by 873.10^{-5}. In the third case, it is overestimated by 1058.10^{-5}. Given the uncertainty values (1σ), the k_{eff} difference is higher than three times the combined standard deviation only for annular cylinders with air + cadmium in their inner parts. Globally, a tendency to overestimation is observed.

Table 6: k_{eff} results for 500×200 annular cylinder with 1.5% ^{240}Pu

Case N°	Benchmark k_{eff}	Experimental uncertainties in pcm	C(Pu), g/liter	Critical height Hc (cm)	Material inside the annular cylinder	Average calculated k_{eff} (standard deviation in pcm)
1, 2, 9, 10, 11, 12	1.0000	173 to 379	21.87 to 45.10	26.34 to 53.07	Air	1.01109 (78)
4 to 7	1.0000	173 to 388	29.79 to 45.10	27.60 to 39.99	Water	1.00873 (34)
13 to 16	1.0000	173	24.75 to 39.85	29.60 to 50.04	Air + Cd	1.01058 (274)

3.6 Analysis of the overestimation

Three concentrations have been chosen (30 g/l, 40 g/l and 60 g/l) to show the variation of the k_{eff} difference against ^{240}Pu content (Fig.2). It appears that the k_{eff} difference increases when the plutonium content in ^{240}Pu decreases. The observed overestimation is coherent with APOLLO2-MORET 4 validation database results. For plutonium nitrate solution reflected or not by water, k_{eff} is overestimated by around 0.5-0.6% [4]. CRISTAL validation database comprises plutonium solution with a ^{240}Pu wt. % of 23% down to 0.54%. In the present paper, it is shown that for a lower proportion of ^{240}Pu, the overestimation is larger (1%). This tendency is confirmed by TRIPOLI4 [2] calculations using point-wise cross-sections from JEF2.2 evaluation. On Fig. 2, given the experimental uncertainties, the tendency is not clear. Therefore, given that a bias associated with the laboratory cannot be excluded, it is necessary to find additional cases to definitely conclude. It can be done using other cases from the I.C.S.B.E.P. Handbook [1] with a ^{240}Pu content varying from 0.54% to 23%.

In the I.C.S.B.E.P. database, PU-SOL-THERM-001, 002, 003, 004, 005, 006, 009, 010, 011, 012 and 024 are benchmark experiments corresponding to the previous criteria. When comparing the Δk_{eff} values obtained for the annular cylinders with the ones obtained for CRISTAL validation database experiments (Fig.3), it appears that these values are in good agreement. No particular tendency to overestimation with ^{240}Pu content can be highlighted. However, since these experiments have large experimental uncertainties, an experimental bias cannot be excluded. As a consequence, it is difficult to conclude to a tendency with ^{240}Pu % in nitrate solutions.

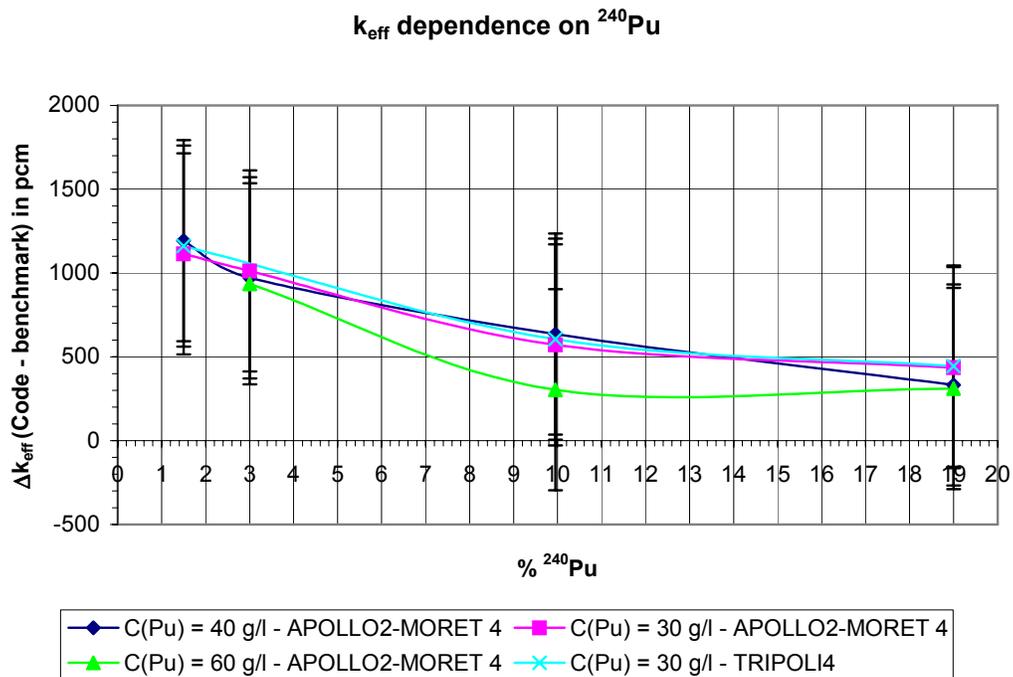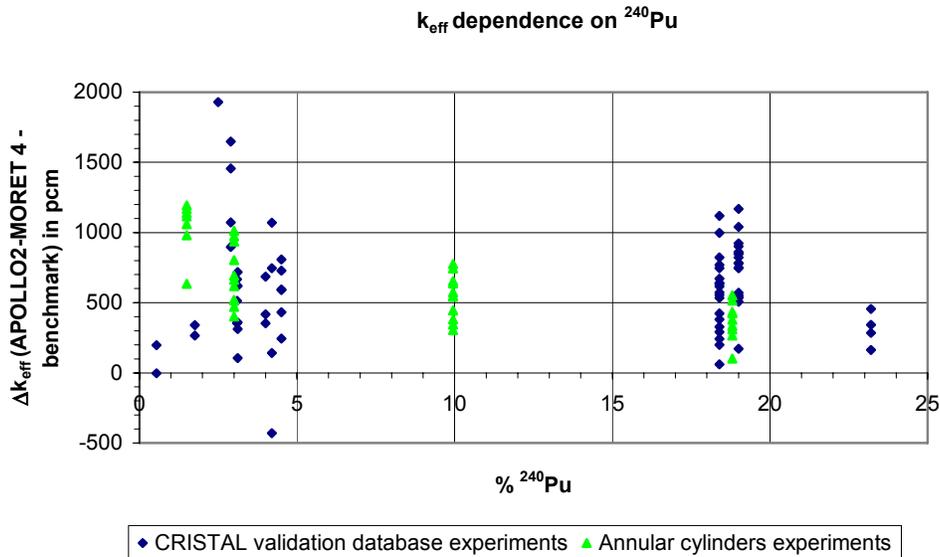**Figure 2**: Variation of the k_{eff} difference (APOLLO2-MORET 4 – benchmark) against ^{240}Pu content

k_{eff} dependence on ^{240}Pu

Figure 3: Comparison between annular cylinder experiments and CRISTAL validation database experiments

k$_{eff}$ dependence on ^{240}Pu

◆ CRISTAL validation database experiments ▲ Annular cylinders experiments

4 Evaluation of experimental uncertainties

4.1 Evaluation method

The experimental uncertainties were evaluated for the different parts of the Annular Cylinders experimental programme.

When measurements were performed (chemical data for example), the experimenters provide the level of confidence at which the measurement is known. The 1σ uncertainty is directly derived.

When only tolerances are available, the evaluator must do assumptions on the distribution of measurements. Most of the time, the equiprobable distribution is assumed and the tolerance must be divided by square root of three to obtain the 1σ uncertainty.

A Monte-Carlo perturbation or S$_n$ calculation is then performed to weigh the reactivity worth of the 1σ uncertainty, the parameter being varied from 1σ. A Δk$_{eff}$ (k$_{eff}$ difference) corresponding to the parameter variation is obtained.

These Δk$_{eff}$ are combined quadratically (making the assumptions that the uncertainties are uncorrelated) to obtain the total experimental uncertainty.

4.2 Results

For each type of sub-programme (type of annular cylinder and ^{240}Pu content), the maximum total uncertainty value is provided. It can be noted that the total uncertainty is lower than 0.4%, which is reasonable. For each type of cylinder, the maximum total uncertainty has been retained. The difference with the average one is particularly important for 500 × 200 annular cylinders with 1.5% ^{240}Pu. In that case, large uncertainties on critical

height (last reached height far from critical height) are noticed. But generally, the range of variation of the total uncertainties is the following:

> from 0.153% to 0.237% for 500×200 annular cylinder with 19% ^{240}Pu,
> from 0.120% to 0.193% for 500×200 annular cylinder with 9.95% ^{240}Pu,
> from 0.122% to 0.388% for 500×200 annular cylinder with 1.5% ^{240}Pu,
> from 0.110% to 0.121% for 500×300 annular cylinder with 3% ^{240}Pu,
> from 0.171% to 0.249% for 500×300 interacting annular cylinders with 3% ^{240}Pu.

Therefore, it can be concluded that the experimenters carried out the experiments precisely enough. The main uncertainties are those on plutonium concentration, acidity, radius of the annular tank and critical height for experiment n° 487 making use of plutonium nitrate solution with 1.5% ^{240}Pu. The large uncertainty on critical height is associated with the extrapolation of the last reached fissile solution height to the critical level.

The total uncertainty value is approximately the same for the different parts of the programme. The low values of total uncertainties enable to use the experiments for validation purpose of the APOLLO2-MORET 4 calculation route.

5. Conclusion

From 1963 to 1976, 730 critical experiments with Annular Cylinders containing plutonium nitrate solutions with various ^{240}Pu contents have been carried out and some were evaluated. These experiments aimed at validating criticality codes either in configurations with reactor-grade plutonium coming from the reprocessing cycle or with weapon-grade plutonium coming from the decommissioning of nuclear weapons and also validate this way of storing nitrate solutions. The 83 selected experiments are all the more valuable as they show small uncertainties and as their calculated k_{eff} is close to the benchmark k_{eff} (except for solutions at a 1.5% ^{240}Pu content). Nevertheless a small tendency to overestimation ($\sim$ 1%) has been noticed for 500×200 Annular Cylinders with 1.5% ^{240}Pu. This tendency does not seem to be correlated with the ^{240}Pu content in plutonium solutions. Further investigations are in progress to confirm this tendency. They deal with the comparison with experiments coming from other laboratories (to avoid a possible experimental bias) and the use of other cross-sections libraries (JEFF3.1 and ENDF-BVI).

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