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Minimum Critical Values of Uranyl and Plutonium Nitrate Solutions Calculated by Various Routes of the French Criticality Codes System CRISTAL using the New Isopiestic Nitrate Density Law.

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This paper provides for various cases of ^{235}U enrichment or Pu isotopic vectors, and different reflectors, new minimum critical values of uranyl nitrate and plutonium nitrate solutions ($H^+ = 0$) obtained by the standard IRSN calculation route and the new isopiestic density laws. Comparisons are also made with other more accurate routes showing that the standard one's results are most often conservative and usable for criticality safety assessments.

1. Introduction

This paper presents summarized studies related to the updating of the French Guide [1)] and to the French contribution of the OECD Minimum Critical Values (MCV) group [2)].
It shortly gives new MCV of Uranyl Nitrate (UNH) and Plutonium nitrate (PuNH) solutions. These values are calculated by the standard route of the French criticality code system CRISTAL [3)] using the new isopiestic nitrate density law [4)]. Moreover, in some specific cases, comparisons are also made with results obtained by other more accurate routes, showing a slight safety margin.

2. MCV Calculation

2.1 Calculation Cases

For UNH, six uranium enrichments are taken into account: 3, 4, 5, 10, 20, 100, ^{235}U wt%.
For PuNH, eight isotopic vectors are taken into account, respectively $^{239}Pu/^{240}Pu/^{241}Pu/^{242}Pu$ in wt%: 100/0/0/0/ - 71/17/11/1 – 95/5/0/0 – 90/5/5/0 – 90/10/0/0 – 80/10/10/0 – 80/20/0/0 – 80/15/5/0.
These solutions are pure aqueous ones, i.e., water moderated, without acidity (H+ = 0). These cases are unrealistic from a chemical point of view, but are convenient for the purpose of criticality. They are more reactive than the real existing cases, i.e., solution of the uranyl nitrate or plutonium nitrate crystals, respectively $UO_2(NO_3)_2$-$6H_2O$ and $Pu(NO_3)_4$-$5H_2O$, in nitric acid.
Three types of close reflectors, convenient in criticality assessments, in contact with the fissile solutions are taken into account: 20 cm of water, the pair of 25 cm of lead and 20 cm of water, and 60 cm of usual concrete (density $\rho = 2.3$ g/cm^3, H = 1.3740 10^{22}, O = 4.5908 10^{22}, Na = 2.7780 10^{21}, Al = 1.7380 10^{21}, Si = 1.6608 10^{22}, Ca= 1.4989 10^{21}, at/cm^3).
All cases but the Pu vector 71/17/11/1 one and the two latter reflexion ones were proposed by OECD, for international comparison on MCV.
Note also that the concrete composition is the standard IRSN one, but there are other more efficient concretes especially depending on water amount [5)].
The lead/water pair reflexion (25cm/20cm lead/water) is a standard IRSN one, but some arrangements of these two materials can also be more effective [6)].

2.2 Theory and Experimental Validation of Isopiestic Nitrate Density Law

Former usual nitrate density laws (for example, Leroy & Jouan or ARH-600 laws, currently previously used by IRSN, named 'Old' further) are fitted mathematical expressions based on measurements in a restricted range of concentrations. They are often used outside of their determination range, without any scientific certainty[7)].
One can derive new isopiestic nitrate density laws from thermodynamic theory based on equality of water activity in electrolytes mixtures. Detailed theory and validation of these new density laws are presented in another paper of ICNC'03 meeting [4)]. Here, note only that validation is made against solution ICSBEP benchmarks [8)], being a new original benchmark use.

The improvements obtained by the new isopiestic law are quite impressive, especially for Pu nitrate solutions, which results are now closer to the

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experimental ones while they could formerly lead to unsafe (-3400 10^{-5}) results near the geometry moderation optimum.

2.3 Calculations Routes, Codes and Cross-sections

Two routes are used:
Standard route: for IRSN MCV standard route is APOLLO2 Sn, n = 8, P3, 20 energy groups cross-sections collapsed from 172 energy groups CEA93 ones deriving from JEF2.2. The convergence accuracy is 10^{-5}. The calculation process and the energy groups collapsing were studied and purposely defined to be efficient, fast and slightly conservative aiming more at safety than at very exact results.
Note also that the concrete composition is the standard IRSN one, but there are other more efficient concretes especially depending on water amount [5].
The lead/water pair reflexion (25cm/20cm lead/water) is a standard IRSN one, but some arrangements of these two materials can also be more effective [6].
Accurate routes: used for the conservatism evaluation of the former standard route results, they are:
- APOLLO2 Sn Keff with 172 energy groups cross sections coming from JEF2.2.
- TRIPOLI 4.1 with point wise JEF2.2 or ENDF B VI cross-sections (standard deviation $\sigma \leq 100\ 10^{-5}$)

For standard MCV, described later, the very slight improvement of accuracy of these latter routes is not interesting considering their large time consuming. Moreover, for criticality assessment, IRSN uses safe values defined as the minimum of the two following values:
- C x MCV, with, C = 0.7 for mass (0.43 if possible double batching), C = 0.75 for volume, C = 0.85 for cylinder diameter, C = 0.75 for slab thickness.
- Values calculated for Keff = 0.95.

Differences obtained between critical and such safe values give a large safety margin $\geq 5000\ 10^{-5}$, and much larger than possible uncertainties reactivity weight.

3. Standard Results

Many calculations are performed, giving Kinfinite, material bucklings (B2m), and critical values against the moderation ratio H/X. The following Tables 1 and 1bis on next page, present only the 145 MCV results obtained by the standard route (corresponding to 1500 calculation cases of various H/U or H/Pu, ^{235}U enrichments and Pu isotopic vectors). For example, Figures 1 and 2 present minimum critical volume variations against ^{235}U enrichment and ^{239}Pu equivalent **9)**.

Table 1 MCV of Uranyl Nitrate Solution ($H^+ = 0$)

Enrichment ^{235}U (wt%)	**3**	**4**	**5**	**10**	**20**	**100**	Reflector Thickness (cm)
Volume (litre)	365.5	134.92	80.48	29.20	16.03	6.54	Water 20 cm
Mass U (kg)	464.9	144.2	75.38	17.38	5.92	0.81	
Cylinder Diameter (cm)	64.62	45.18	37.82	26.18	20.92	14.88	
Slab Thickness (cm)	37.4	24.98	20.04	12.53	9.18	5.40	
Volume (litre)	312.15	113.23	67.38	24.78	13.82	5.77	Concrete 60 cm
Mass U (kg)	399.88	122.69	64.09	14.96	5.17	0.71	
Cylinder Diameter (cm)	59.56	41.20	34.04	23.32	18.58	13.10	
Slab Thickness (cm)	31.42	19.58	15.08	8.52	5.74	2.70	
Volume (litre)	244.1	88.61	53.25	20.23	11.56	5.09	Lead/Water 25 /20cm
Mass U (kg)	313.20	95.37	49.97	11.91	4.17	0.58	
Cylinder Diameter (cm)	52.28	35.88	29.64	20.52	16.54	11.98	
Slab Thickness (cm)	23.32	13.80	10.46	5.94	4.14	2.30	
Critical limit concentration (g/l)	624	415	314	137	65	12	

Table 1bis MCV of Plutonium Nitrate Solution (H^+ =0)

	Isotopic Vector								
^{239}Pu ^{240}Pu ^{241}Pu ^{242}Pu (wt%)	100 0 0 0	71 17 11 1	95 5 0 0	90 5 5 0	90 10 0 0	80 10 10 0	80 15 5 0	80 20 0 0	Reflector (cm)
Volume (litre)	7.57	15.60	10.74	10.27	13.29	12.06	15.24	19.56	Water 20 cm
Mass Pu (kg)	0.51	0.92	0.63	0.60	0.77	0.71	0.89	1.16	
Cylinder Diameter (cm)	15.66	20.66	17.94	17.64	19.46	18.74	20.48	22.46	
Slab Thickness (cm)	5.84	8.90	7.24	7.04	8.18	7.7	8.83	10.06	
Volume (litre)	6.42	13.00	Not calculated						Concrete 60 cm
Mass Pu (kg)	0.45	0.78							
Cylinder Diameter (cm)	13.6	18.02							
Slab Thickness (cm)	2.96	5.16							
Volume (litre)	5.60	10.73							Lead/Water 25/20 cm
Mass Pu (kg)	0.36	0.62							
Cylinder Diameter (cm)	12.32	15.90							
Slab Thickness (cm)	2.35	3.68							
Critical limit concentration (g/l)	7.2	9.2	7.8	7.64	8.47	8.07	9.1	10.22	

4. Comparison and Comments

Standard MCV are compared taking into account the old and the new isopiestic density laws, standard route, and more accurate routes (these routes are given in § 2.3). Note that calculation with Sn 8 is more accurate than with Sn 4 formerly used by IRSN for the MCV data in [1].

4.1 Comparison using Previous and New Isopiestic Density Laws

As can be forecast from validation or B^2m results [4], differences are not important for optimum moderation (mass) but can be noticeable for geometry, and larger for Pu nitrate solution than for uranyl nitrate one. For example, comparison results on critical volume (litre) are given for some representative water reflected cases (^{235}U Enrichment = 100%, 5% & 3% and Pu vectors $^{239}Pu/^{240}Pu/^{241}Pu/^{242}Pu$ in % = 100/0/0/0/ & 71/17/11/1) in the following Table 2.

Table 2 Critical Volume V (litre) obtained by Standard Calculation Route using new Isopiestic (Iso) Density Laws or previous (Old) Density Laws.

Law	Iso	Old	Iso	Old	Iso	Old
^{235}U	100 %		5 %		3 %	
V (litre)	6.54	6.50	80.48	80.40	365.53	369.26
^{239}Pu	100 %		71 %			
V(litre)	7.57	8.46	15.6	16.70		

4.2 Comparison against Calculations using 172 Energy Groups Cross-sections

20 energy groups cross-sections are routinely used because they are better than the old 16 Hansen & Roach ones and faster (calculation time reduced by a factor 10) than the 172 energy groups ones. The following Table 3 presents the effect of the energy groups' number for critical geometries also determined by Apollo 2 Sn 8 but with 172 energy groups.

Table 3 Keff calculated by APOLLO2 Sn 8 172 energy groups CEA 93 cross-sections for Standard Route Critical Geometries using Isopiestic Density Laws.

	^{235}U		^{239}Pu	
^{235}U or ^{239}Pu	100 %	3%	100 %	71 %
Sphere	0.99877	0.9946	0.99932	0.99857
Slab	0.99755	0.99948	1.00129	0.99577
Cylinder	0.99901	0.99937	0.99935	0.99874

Excepted for one case (slab, ^{239}Pu = 100%), all standard calculations are conservative.

4.3 Comparison against TRIPOLI 4.1 calculations using pointwise Cross-sections

The two following Tables 4 & 5 present Keff values calculated by TRIPOLI4.1for critical geometries obtained by the standard route (sphere and infinite slab water reflected). Standard deviations are in the range between 50 10^{-5} and 100 10^{-5}.

Table 4 Keff of Critical Standard Spheres by TRIPOLI 4.1.

Isotopic (%)		Critical Data		Keff Results	
^{235}U	^{239}Pu	H/X	R (cm)	JEF2.2	ENDF BVI
100	/	70	11.60	0.99240	0.99296
3	/	12	44.36	0.99942	0.99573
/	100	100	12.18	0.99702	0.99678
/	71	200	15.50	0.99483	0.99792

Table 5 Keff of Critical Standard Slabs by TRIPOLI 4.1

Isotopic (%)		Critical Data		Keff Results	
^{235}U	^{239}Pu	H/X	Ep (cm)	JEF2.2	ENDF BVI
100	/	30	5.40	1.00368	1.00364
3	/	12	37.40	0.99443	0.99371
/	100	50	5.84	0.99967	0.99907
/	71	200	8.90	0.99238	0.99305

Examples on Tables 4 & 5 show that calculations by standard route are always conservative but in one case of the 100% uranyl nitrate solution slab.

5. Conclusion

New MCV values, calculated by IRSN standard route for updating the French criticality guide and for the OECD international comparison, are presented. They are based on a new isopiestic density law derived from thermodynamic theory and not from empirical measurements mathematical fits. They are calculated by the recent French criticality codes package CRISTAL by APOLLO2 Sn 8 using CEA-93 172 energy groups cross-sections collapsed in 20 energy groups.

Standard results are conservative against more accurate results obtained by other routes:

- 200 10^{-5} against calculations carried out by APOLLO2 Sn Keff with 172 energy groups cross sections,
- 700 10^{-5} against calculations carried out by IRSN reference code TRIPOLI4.1 and JEF2.2 or ENDF B VI point wise cross sections.

Presented MCV are also conservative for criticality assessments, for two reasons:

- criticality assessments are based on smaller safe values (at less for $\Delta Keff = 5000\ 10^{-5}$),
- the studied solutions in water without acidity exist only for the purpose of criticality safety, while in real nitrate solutions MCV values are higher when taking into account the poisoning effect of NO_3^-.

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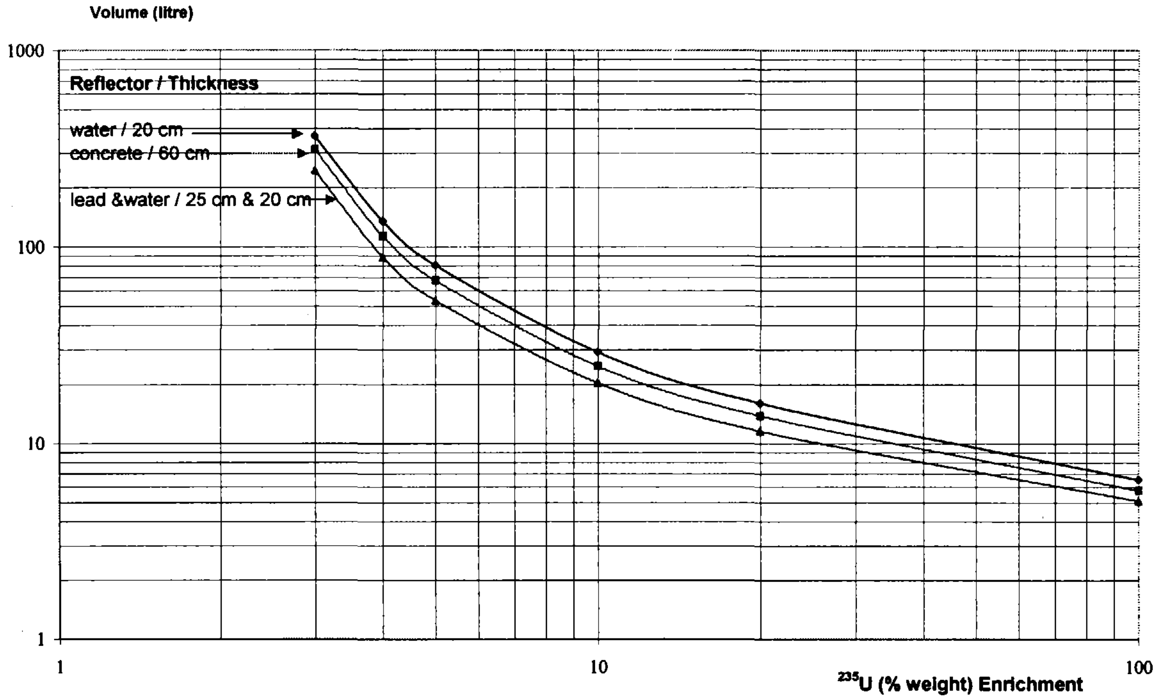

Fig.1 Minimum Critical Volume of Uranyl Nitrate Solutions ($H^+ = 0$)

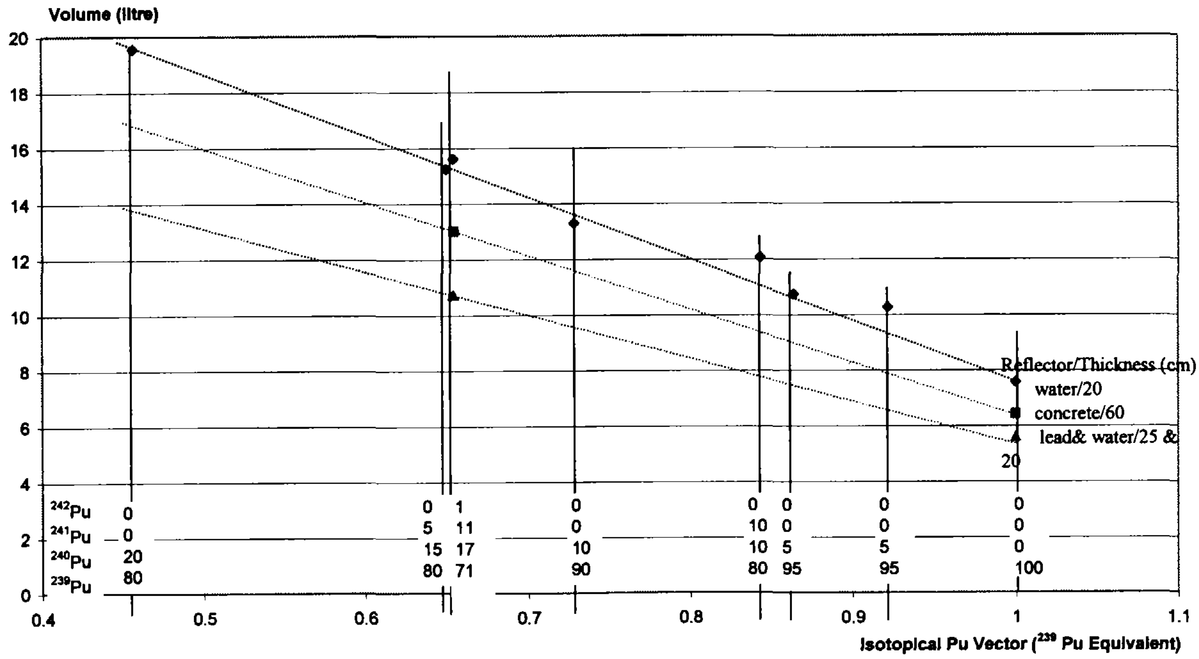

Fig.2 Minimum Critical Volume of Plutonium Nitrate Solution ($H^+ = 0$)

Pu Equivalent = Pu Eq = $[^{239}Pu] - 1.7177[^{240}Pu] + 2.15[^{241}Pu] - 0.0115[^{238}Pu + ^{242}Pu]$ see [9]

Isotopical Vector					Isotopical Vector				
^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	Pu Eq	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	Pu Eq
100	0	0	0	1	90	10	0	0	0.72823
71	17	11	1	0.654376	80	10	10	0	0.84323
95	5	0	0	0.864115	80	15	5	0	0.649845
90	5	5	0	0.921615	80	20	0	0	0.45646