JP0450022

JAERI－Conf 2003－019

Actinides Critical Masses and the Paxton Woodcock Rule

Dr. Jacques ANNO[1], Isabelle DUHAMEL[2], Caroline LAVARENNE[3], Matthieu DULUC[4]
[1,2,3] : *Institut de Radioprotection et de Sûreté Nucléaire*
IRSN, BP 17, 92262 Fontenay aux Roses Cédex, France.
[4]: *Nuclear Engineer Student at INSTN (Institut National des Sciences et Techniques Nucléaires) Saclay.*

This paper presents recent actinides (reflected or not, moderated or not) critical masses calculations performed by the French standard route (APOLLO2 Sn 8 P3, 20 energy groups cross-section collapsed from 172 energy groups CEA 93 library). Comparisons are also presented against more accurate routes of the French criticality package CRISTAL, showing the fair conservatism of the standard values. Checks of the Paxton Woodcock rule for transportation exemption limit were also made.

1. Introduction

For advanced fuels reprocessing or improvement in transport regulations, the critical masses of actinides are needed. Studies are being performed by many organizations or groups of experts, for example respectively JAERI [1,2] or ANS/ANSI 8.15 [3].

In ICNC'99, some results of the French contribution [4] to ANS/ANSI 8.15 were published, especially recalling characteristics of 34 Actinides and average nominal production (g/t) of 30 of them (in PWR, BWR, UOX or MOX fuels burnt up to 35 GWd/t, with a specific power of 35 W/g and 90 days of cooling time). It pointed out the very small production of some actinides, for example ^{232}U and ^{236}Pu (about or less than 1mg/t of initial U). Some proposals of exception limits for Transport were also given. Then, IRSN pursued extensive calculations to compare actinides critical mass obtained by various routes of the CRISTAL package [5] and to check the related results against critical experiments. Systematic comparisons were also made against the current transportation exemption rule of 15 g of fissile material [6,7] (i.e. Paxton & Woodcock Transportation Exemption Rule – PWTER) for comparison with former interesting study [8]. This paper present all these new results and comparisons.

2. Standard Route Results

IRSN standard route calculation for critical (especially minimum) values is APOLLO2 Sn 8 P3 using cross sections from the library CEA93 (V4) (X-mas) 172 energy groups derived from JEF2.2 collapsed in 20 energy groups. Results are given in Table 1.

Reflectors commonly used for criticality assessments are 20 cm of water (W), 30 cm of Stainless Steel (SS), the pair of 25 cm of lead plus 20 cm of water (LW), 60 cm of usual concrete (density ρ = 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).

Note that the concrete composition is the standard IRSN one, but more efficient concretes exist depending on the water amount [9].

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 efficient [10].

Deriving from mathematical fit of data displayed on Figure 1, relationships can be obtained between metallic reflected critical masses Y and bare ones X in kg, Y = aX + bX^2 (relationships are written on Figure 1).

3. Comparison with other routes

Other CRISTAL routes use 172 energy groups cross sections (with codes MORET4 or APOLLO2 Sn Keff) or point wise cross sections (with code TRIPOLI4.1). Results are given on Table 2 and 3 respectively for metallic or water moderated spheres. The standard route is conservative for metallic or moderated cases versus the other ones with 172 energy groups cross sections, but this is not always true versus TRIPOLI4.1 and point wise JEF 2.2 cross sections for some metallic cases.

Other comparisons are also being performed with other cross sections and codes in the frame of international study [11]. During this study, it was discovered for ^{236}Pu, that the minimum critical mass of moderated water case was smaller than the metallic case, which was not obtained with 20 or 172 energy groups cross sections. Thus, even if for many

---

[1] Corresponding author, Tel 33 1 58 35 81 15 Fax 33 1 46 57 29 98, E-Mail: jacques.anno@irsn.fr

actinides, production quantities are very small, the amounts to be transported are totally unknown, then one should be very careful when establishing sub-critical limits, even by using division factors (0.5 or 0.2 when no critical experiments are available or when cross sections are doubtful) on critical masses.

4. Experimental Validation

Only benchmarks with ^{233}U, ^{235}U, ^{239}Pu and ^{242}Pu are available in ICSBEP handbook [12, 13]. Related cases were used to qualify our codes and cross sections. For moderated cases, the agreement is quite fair, slightly conservative. For metallic cases, the general CRISTAL tendency is that bare or water reflected calculations are slightly (in average 500 – 700 10^{-5}) not conservative against experiments while calculations with APOLLO2 on stainless steel reflectors are over-conservative. In this latter case, qualification studies show a noticeable conservative margin depending on the reflector thickness [14]. In the case of ^{237}Np, US [15, 16] or French [16] replacement experiments let think that JEF2.2 sections are not ecact.

5. Checking the Paxton Woodcock Rule

Calculations were carried out to obtain the safe mass limit using (1), the Woodcock & Paxton Formula [7]. It gives the mass limit to be transported per package for an array of 250 packages (each one is 10x10x10 cm^3).

$$M_{\text{limit}} = \left(\left(\frac{Msafe}{N}\right)\rho^{s}\right)^{1/(s+1)} \qquad (1)$$

Msafe is the minimum of M (Keff = 0.95) and 0.7 Mc, N = 250, ρ = mass concentration (g/l).

Calculations results are given in table 4: in solution, where the minimal critical values are obtained, the exemption limit of 15 g was only obtained for ^{235}U and ^{247}Cm. Smaller limits are calculated for the others.

Preliminary calculations [11] also showed that arrays of 250 packages loaded of 15 g of material fissile in solution are not safe, as was also determined by N. Barton [8]. The presented results confirm those obtained with the Paxton Woodcock rule: When calculated mass limits are smaller than 15 g, such arrays are critical. New limits should be established.

6. Conclusion

As other organisations, for advanced fuels reprocessing or improvement in transportation regulations, IRSN is systematically studying the critical masses of actinides with the French criticality codes package CRISTAL. In previous ICNC'99 characteristics and production of 34 actinides were given and first proposal for transportation exemption made. Extended criticality data are now given, with various reflection conditions. The standard route (APOLLO2 SN 8 P3 20 energy cross-sections collapsed from the 172 energy groups CEA93 library) is generally conservative against other more accurate routes using 172 energy groups cross-sections from JEF2.2 or TRIPOLI4 with JEF2.2 point wise cross sections.

Between CRISTAL routes, systematic comparison shows the conservatism of the IRSN standard route calculations.

The results checking the Woodcock & Paxton Transportation Exemption also show that only ^{235}U and ^{247}Cm give a limit greater than 15 g. Thus new Transportation Exemption Limits are under study and will be next proposed to AIEA. For this purpose, even if some of these actinides are produced in very small (less than or equal to 1mg/t) amount in reactors, safety coefficients should be taking into account, considering the observed differences on minimum critical masses obtained with various codes and cross sections evaluations.

References

1) H. Okuno & H. Kawasaki, Critical and Subcritical Masses of Curium 245, 246 & 247 calculated with a Combination of MCNP4A Code and JENDL-3.2 Library, JAERI Research 2000-04 Sept. (2000).

2) H. Okuno & H. Kawasaki, Critical and subcritical Mass Calculations of Curium-243 to -247 Based on JENDL-3.2 for Revision of ANSI/ANS-8.15, Journal of Nuclear Science and Technology, Vol 39, N° 10, p 1072 Oct. (2002).

3) N. L. Pruvost, Current activities of the ANS-8.15 working group, Nuclear Standards News Vol 33/N°1, Jan.-Feb. (2002).

4) J. Anno & G. Sert, ''French Participation at ANS/ANSI 8.15 Working Group Updating Criticality Data on ^{237}Np. Criticality & Transportation Proposals'', Proc. Int. Conf. on Nuclear Criticality Safety, ICNC'99, Versailles, France, Sept.20-24, 1999, I, 447 (1999).

5) J. M. Gomit, P. Cousinou, A. Duprey, C. Diop, J. P. Grouiller, & L. Leyval, ''The new CRISTAL Criticality-Safety Package'', Proc. Int. Conf. on Nuclear Criticality Safety, ICNC'99, Versailles, France, Sept.20-24, 1999, I, 308 (1999).

6) Safety Standards Series N° TS-G-1.1 (ST-2) Advisory Material for the AIEA Regulations for the Safe Transport of Radioactive Material. Safety Guide IAEA, Vienna (2002).

7) R.E. Woodcock & H.C. Paxton "The Criticality Aspects of Transportation of Fissile Materials' p. 401 in Progress in Nuclear Energy Series IV Technology Engineering & Safety Vol 4 C. M. Nichols Pergamon Press (1961)

8) N. J. Barton & C. K. Wilson,'' Review of Fissile Exception Criteria in IAEA Regulation", Proc. Int. Conf. on Nuclear Criticality Safety, ICNC'95, Albuquerque, USA, Sept. 17-21, 1995, **II**, 9-15 (1995).

9) J. Anno & G. Kyriazidis "Variation against distance of concretes or steel thicknesses equivalent to a nominal close water reflection" Proc. Int. Conf. on Nuclear Criticality Safety, ICNC'99, Versailles France, Sept.20-24 1999, **I**, 115 (1999).

10) D. Clayton Gmelin Handbook of Inorganic Chemistry – Supplement Vol A6 "General Properties Criticality", 244 (1983).

11) C. Lavarenne, P. Cousinou, V. Rouyer, D. Mennerdahl, C. Dean, N. Barton, M.T. Lizot, G. Sert, ''Selections of Exception Limits for all Actinide Nuclides based on revised Criteria for Safe International Transport'', Proc. Int. Conf. on Nuclear Criticality Safety, ICNC'03, Tokai Mura, Japan, Oct 20-24 (2003).

12) AEN/NEA/OECD International Handbook of Evaluated Critical Safety Benchmark. Experiments. NEANSC DOC (95) 03 Sept. (2002).

13) R.W. Brewer "242Pu Critical Mass", Proc. Int. Conf. on Nuclear Criticality Safety, ICNC'99, Versailles, France, Sept.20-24, 1999, I, 456 (1999).

14) I. Duhamel, J. M. Gomit, Y. K. Lee & C. Venard, "Synthesis on the Validation of the CRISTAL V0 Package'', Proc. Int. Conf. on Nuclear Criticality Safety, ICNC'03, Tokai Mura, Japan, Oct 20-24 (2003).

15) R.G. Sanchez, "Critical Mass of ^{237}Np", Proc. Int. Conf. on Nuclear Criticality Safety, Albuquerque, NM, Sept. 17-21, pp. 182-189 (1995).

16) R.G. Sanchez, D.J. Loaiza & R.H. Kimpland ''Criticality of a Neptinium-237 Sphere", LA-UR-03-0379, ANS Conference, San Diego, June (2003).

17) P. Humbert & B. Méchitoua " Numerical Simulation of Caliban Reactivity Perturbation Experiments using Neptunium-237 Samples''. Proc. Nuclear Mathematical and Computational Sciences: A Century in Review, a Century Anew, Gattlinburg, Tennessee, April 6-10, (2003).

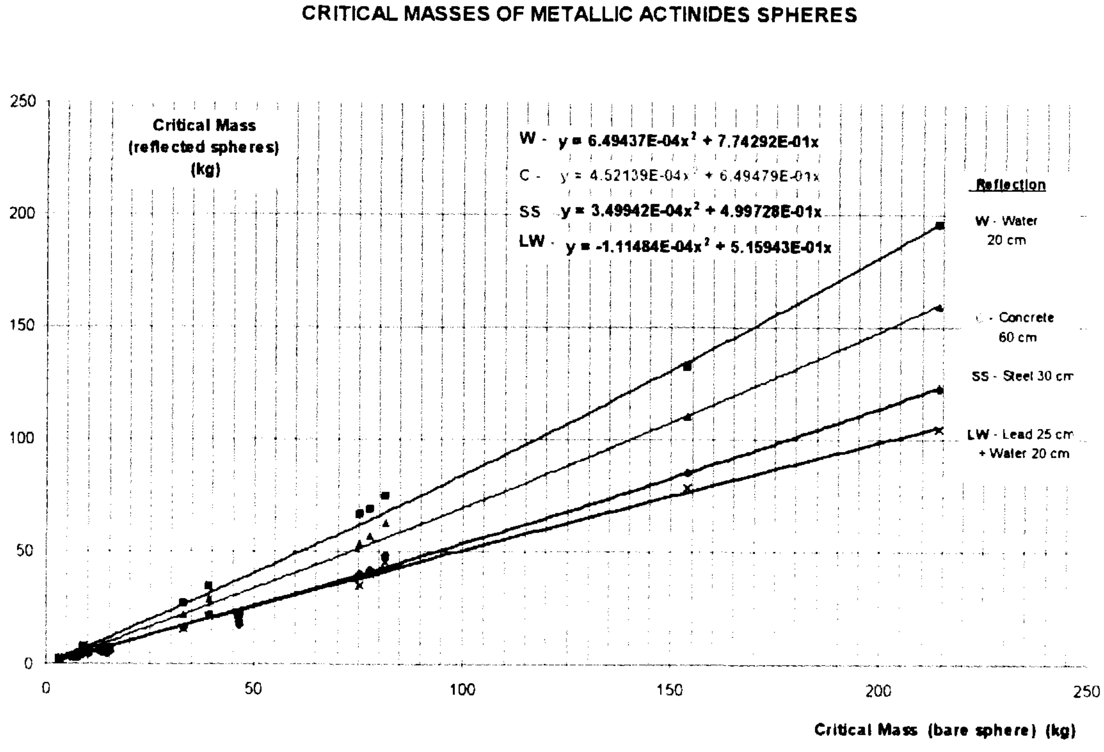

Figure 1 Actinides Metallic Critical Spheres
Relationship between Reflected Masses and Bare Masses

Table 1 Actinides Critical Masses - Results of the standard route CRISTAL

State	Metal					Solution
Reflector Thickness	Bare	SS 30 cm	Water 20 cm	Concrete 60 cm	Lead/Water 25 cm/30cm	Water 20cm
^{232}U	3.477	1.835	2.048	2.057	2.065	
^{233}U	15.505	6.032	7.097	6.763	7.026	0.553
^{234}U	153.78	85.183	132.087	110.374	78.453	
^{235}U	46.563	17.079 *	21.347	18.726	20.109	0.779
^{236}**Pu**	8.156	3.747	4.840	4.450	4.092	
^{238}**Pu**	9.115	4.421	7.280	5.964	5.055	
^{239}**Pu**	10.225	4.655 *	5.989	5.526	5.364	0.498
^{240}**Pu**	39.306	21.694	34.681	28.653	21.389	
^{241}**Pu**	13.143	5.309	6.531	6.074	6.476	0.267
^{242}**Pu**	77.660	42.164	68.852	56.623	40.436	
^{237}**Np**	81.655	48.073	74.704	62.526	45.245	
^{241}**Am**	75.495	40.066	66.807	53.197	35.264	
^{242m}**Am**	14.375	4.505	6.368	5.411	5.828	0.023
^{243}**Am**	214.3	122.54	195.440	159.036	104.413	
^{242}**Cm**	25.152	**12. 8**	**19.9**	**16.6**	**12.9**	
^{243}**Cm**	7.336	2.758	2.829	2.939	3.390	0.264
^{244}**Cm**	32.965	16.007	26.871	21.605	15.479	
^{245}**Cm**	6.809	2.657	2.607	2.620	3.206	0.047
^{246}**Cm**	42.529	**21.9**	**34.1**	**28.4**	**21.7**	
^{247}**Cm**	7.206	**3.6**	**5.6**	**4.7**	**3.7**	2.104

In blue, rounded values calculated by fitted relationships – see Fig. 1- for comparison. * note (see text) that these values are noticeable conservatives against available experimental validation [11)]

Table 2 Metallic Critical Masses Comparison (kg)

	Reflector														
	None				Stainless Steel 30 cm		Water 20 cm			Concrete 60 cm		Lead+Water 25cm +20cm			
	A2 * Sn Normes	A2 MORET4	A2 Sn Keff	TRIPOLI4.1	A2 Sn Normes	A2 Sn Keff	A2 Sn Normes	A2 Sn Keff	TRIPOLI4.1	A2 Sn Normes	A2 Sn Keff	A2 Sn Normes	A2 Sn Keff		
Groups	20	172	172	p***	20	172	20	172	p***	20	172	20	172		
^{232}U	**3.48**	**3.52**	**3.70**	**3.65**	**1.84**	**1.97**	**2.05**	**2.18**	**2.16**	**2.06**	**2.14**	**2.07**	**2.21**		
^{233}U	**15.51**	**15.37**	**16.34**	**17.70**	**6.03**	**6.40**	**7.10**	**7.46**	**7.60**	**6.76**	**7.16**	**7.03**	**7.82**		
^{234}U	**153.78**	**142.91**	**148.53**	**145.99**	**85.18**	**85.33**	**132.09**	**137.35**	**135.54**	**110.37**	**115.60**	**78.45**	**82.62**		
^{235}U	**46.56**	**44.32**	**48.24**	**47.31**	**17.08**	**17.16**	**21.35**	**22.09**	**21.77**	**18.73**	**19.45**	**20.11**	**21.31**		
^{237}Np	**81.66**	**81.17**	**81.94**	**80.62**	**48.07**	**49.96**	**74.70**	**75.44**	**74.03**	**62.53**	**63.60**	**45.25**	**46.85**		
^{236}Pu	**8.16**	**8.15**	**8.42**	**8.22**	**3.75**	**4.01**	**4.84**	**5.04**	**5.02**	**4.45**	**4.63**	**4.09**	**4.48**		
^{238}Pu	**9.12**	**8.95**	**9.16**	**8.95**	**4.42**	**4.78**	**7.28**	**7.38**	**7.29**	**5.96**	**6.08**	**5.06**	**5.20**		
^{239}Pu	**10.23**	**10.15**	**10.33**	**10.09**	**4.66**	**4.79**	**5.99**	**6.00**	**5.50**	**5.53**	**5.54**	**5.36**	**5.52**		
^{240}Pu	**39.31**	**40.13**	**39.03**	**37.55**	**21.69**	**22.58**	**34.68**	**34.95**	**33.61**	**28.65**	**29.05**	**21.39**	**22.04**		
^{241}Pu	**13.14**	**13.33**	**13.04**	**12.77**	**5.31**	**5.49**	**6.53**	**6.68**	**6.01**	**6.07**	**6.18**	**6.48**	**6.69**		
^{242}Pu	**77.66**	**78.05**	**75.83**	**74.95**	**42.16**	**44.24**	**68.85**	**69.35**	**68.41**	**56.62**	**57.84**	**40.44**	**41.89**		
^{241}Am	**75.50**	**73.65**	**75.61**	**72.70**	**40.07**	**44.00**	**66.81**	**67.77**	**65.78**	**53.20**	**55.12**	**35.26**	**37.55**		
^{242m}Am	**14.38**	**14.38**	**14.50**	**14.58**	**4.51**	**4.62**	**6.37**	**6.44**	**6.85**	**5.41**	**5.49**	**5.83**	**6.01**		
^{243}Am	**214.30**	**214.30**	**209.64**	**203.92**	**122.54**	**132.35**	**195.44**	**192.84**	**189.35**	**159.04**	**160.82**	**104.41**	**109.76**		
^{242}Cm	**25.15**	**	**25.77**	**24.82**	**12.79**	**12.23**	**19.90**	**17.60**	**16.99**	**16.60**	**15.10**	**12.90**	**12.25**		
^{243}Cm	**7.34**	**7.34**	**7.52**	**7.42**	**2.76**	**2.87**	**2.83**	**2.90**	**2.86**	**2.94**	**3.00**	**3.39**	**3.54**		
^{244}Cm	**32.97**	**32.18**	**33.05**	**32.31**	**16.01**	**16.81**	**26.87**	**27.07**	**26.52**	**21.61**	**21.94**	**15.48**	**16.10**		
^{245}Cm	**6.81**	**6.81**	**6.85**	**6.74**	**2.66**	**2.75**	**2.61**	**2.64**	**2.35**	**2.62**	**2.81**	**3.21**	**3.33**		
^{247}Cm	**7.21**	**	**7.12**	**6.98**	**3.60**	**2.99**	**5.60**	**3.46**	**3.49**	**4.70**	**3.37**	**3.71**	**3.48**		

*A2 = APOLLO2, Nnormes = standard route for MCV, ** not calculated ,* ** p = point wise cross-section

Table 3 Water Moderated and Reflected Critical Masses Comparison

	APOLLO2 Sn Normes		APOLLO2 Sn Keff		TRIPOLI 4.1
groups	20		172		p
	C opt. (g/l)	Mass (kg)	C opt (g/l)	Mass (kg)	
^{233}U	60	0,5534	59	0,5594	0,5415
^{235}U	52	0,779	57,5	0,7846	0,7930
^{239}Pu	31	0,498	32	0,5030	0,5066
^{241}Pu	26,7	0,267	26,4	0,2690	0,2730
^{242m}Am	3	0,023	3,5	0,023	
^{243}Cm	60	0,264	58,2	0,2689	0,2687
^{245}Cm	12	0,047	11,5	0,0473	0,0473
^{247}Cm	250	2,104	244,2	2.195	2.2101

In these calculations, critical masses of water moderated ^{232}U and ^{236}Pu are larger than metallic ones, thus they are not mentioned.

Table 4 PWTER Calculations for Actinides Fissile in Solution

	Mc	H/X opt. for Critical Mass	C (X)	0,7.Mc	M(k_{eff} =0,95)	Mlimit	Radius corresponding to Mlimit	Radius for 15 g
X	(kg)		(g/l)	(kg)	(kg)	(g)	(cm)	(cm)
^{233}U	0.5594	435.84	59	0.3916	0.4594	13.818	3.8239	3.930
^{235}U	0.7846	451.10	57.5	0.5492	0.6287	15.579	4.0141	3.964
^{239}Pu	0.5030	825.58	32	0.3521	0.3994	9.174	4.0905	4.819
^{241}Pu	0.2690	1010.00	26.4	0.1883	0.2193	6.361	3.8611	5.139
^{242m}Am	0.0230	7653.50	3.5	0.0161	0.0193	0.708	3.6417	10.076
^{243}Cm	0.2689	460.28	58.2	0.1882	0.2291	10.224	3.4744	3.948
^{245}Cm	0.0473	2356.85	11.5	0.0331	0.0408	1.929	3.4211	6.778
^{247}Cm	2.1955	110.00	244.2	1.5369	1.7977	55.990	3.7969	2.448