Analytical Review of Minimum Critical Mass Values for Selected Uranium and Plutonium Materials+

by

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I. INTRODUCTION

Current subcritical limits for a number of uranium and plutonium materials (metals and compounds) as given in the ANSI/ANS standards for criticality safety are based on evaluations[1,2] performed in the late 1970s and early 1980s. This paper presents the results of an analytical study of the minimum critical mass values for a set of materials using current codes and standard cross section sets. This work is meant to produce a consistent set of minimum critical mass values that can form the basis for adding new materials to the single-parameter tables in ANSI/ANS-8.1[3].

Minimum critical mass results are presented for bare and water reflected full-density spheres and for full density moist (1.5 wt-% water) as calculated with KENO-Va, MCNP4A and ONEDANT.

Calculations were also performed for both dry and moist materials at one-half density. Some KENO calculations were repeated using several cross section sets to examine potential bias differences. The results of the calculations were compared to the currently accepted subcritical limits. However, discussion of the entire set of results is beyond the scope of this summary.

II. CALCULATIONAL APPROACH

Full density bare (unreflected) models consisted of a single spherical region with vacuum boundary conditions, and reflected models included a 30-cm thick region of water (density 0.9982 g/cm^3) around the fissionable sphere. The theoretical densities were taken from standard chemistry references. Moist material densities were calculated according to:

$$\rho_{moist} = \left[\frac{0.015}{\rho_{water}} + \frac{0.985}{\rho_{material}}\right]^{-1}$$

The specific implementation of each of the three codes for the current study is described below. The general approach used was to perform a series of k-effective calculations with slightly different fissionable mass values whose results were approximately centered around a value of $k_{eff} = 1$. A linear fit to the k_{eff} values as a function of mass was then interpolated to a value of $k = 1.0$, and that value is taken to be the minimum critical mass. Uncertainty values were assigned by adding and subtracting the one-sigma uncertainties from the calculated k_{eff} values, then performing linear fits on the resulting data sets.

The ONEDANT calculations were made with the 1-D option of the TWODANT code package, version 02-05-90. The cross section set was the standard 27-group ENDF/B-IV set from the SCALE

4.2 code package, which was processed using modules BONAMI, NITAWL AND ICE. The code was executed on several HP work stations. Number densities were explicitly input to the code.

The KENO-Va code was executed as part of the SCALE 4.3 code package as distributed by RSIC. The executable modules were updated to the July 1996 release, and the revised 44-group ENDF/B-V cross sections were installed. The code was run on a personal computer platform.

KENO-Va was run under CSAS25 which incorporates the BONAMI and NITAWL modules for cross section preparation. For all calculations the standard 238-group ENDF/B-V cross section set was used. Number densities were calculated by the code based on input density values.

MCNP4A, version 10/01/93 was executed on on several HP work stations using continuous-energy ENDF/B-V cross sections. Number densities were explicitly input to the code.

III. RESULTS

Table 1 shows the calculated minimum critical mass values for full-density materials with and without a water reflector. Table 2 shows the results for full density moist materials. The calculated minimum critical mass values are reasonably consistent for the three codes, and differences most likely reflect differences in the cross section sets. The results are also consistent with values given in ANSI/ANS-8.1, which are single parameter subcritical mass limits. More detailed comparisons and discrepancies between cross section sets will be studied as a continuation of this work.

Table 1. Calculated Minimum Critical Mass, Full Density Dry Materials						
	Mass [a] (kg)					
	Bare Spheres			Water-Reflected Spheres		
Material	ONEDANT	KENO-Va	MCNP-4A	ONEDANT	KENO-Va	MCNP-4A
^{235}U	44.29	46.6 ± 0.3	46.5 ± 0.2	20.21	21.7 ± 0.2	21.3 ± 0.1
$^{235}UC_2$	87.04	92.6 ± 0.5	92.4 ± 0.3	38.03	40.8 ± 0.3	40.8 ± 0.2
$^{235}UO_2$	100.1	107.8 ± 0.6	105.4 ± 0.4	43.06	47.1 ± 0.3	46.5 ± 0.3
$^{235}UO_3$	209.2	227 ± 1	223.5 ± 0.7	79.48	88.3 ± 0.7	87.2 ± 0.5
$^{235}U_3O_8$	165.1	179.0 ± 0.9	175.5 ± 0.6	65.32	72.5 ± 0.5	71.0 ± 0.4
$^{235}UF_4$	202.5	219 ± 1	223.9 ± 0.8	79.55	87.4 ± 0.7	88.5 ± 0.5
$^{235}UF_6$	380.7	417 ± 2	429 ± 1	138.9	156 ± 1	157.3 ± 0.9
^{233}U	16.97	15.5 ± 0.1	15.53 ± 0.07	7.191	7.19 ± 0.05	7.02 ± 0.05
$^{239}Pu(\alpha)$	10.44	10.12 ± 0.06	9.75 ± 0.05	5.395	5.49 ± 0.05	5.20 ± 0.03
$^{239}PuO_2$	26.72	27.7 ± 0.2	26.43 ± 0.09	12.25	13.13 ± 0.09	12.57 ± 0.06
$^{239}Pu_2O_3$	27.10	27.9 ± 0.2	26.61 ± 0.08	12.06	13.0 ± 0.1	12.28 ± 0.06
$^{239}PuF_3$	39.51	40.4 ± 0.2	39.7 ± 0.1	17.05	18.1 ± 0.1	17.54 ± 0.08
$^{239}PuF_4$	70.12	72.2 ± 0.4	81.2 ± 0.2	27.17	29.0 ± 0.2	28.2 ± 0.1
$^{239}PuCl_3$	233.1	235 ± 1	229.2 ± 0.6	74.06	77.1 ± 0.6	74.2 ± 0.4

[a] Mass values are total compound or material mass. Theoretical material densities (g/cm^3):

^{235}U	19.05	UF_4	6.70	Pu_2O_3	11.47
UC_2	11.28	UF_6	4.68	PuF_3	9.32
UO_2	10.96	^{233}U	19.05	PuF_4	7.00
UO_3	7.29	^{239}Pu	19.84	$PuCl_3$	5.70
U_3O_8	8.30	PuO_2	11.46		

Table 2. Calculated Minimum Critical Mass, Full Density Moist Materials

	Mass [a] (kg)					
	Bare Spheres			Water-Reflected Spheres		
Material	ONEDANT	KENO-Va	MCNP-4A	ONEDANT	KENO-Va	MCNP-4A
$^{235}UC_2$	93.21	87.6 ± 0.5	88.9 ± 0.4	35.90	38.5 ± 0.3	38.9 ± 0.2
$^{235}UO_2$	94.76	101.2 ± 0.5	100.9 ± 0.4	40.29	43.4 ± 0.4	43.6 ± 0.3
$^{235}UO_3$	179.4	194 ± 1	193.4 ± 0.7	68.64	74.8 ± 0.7	75.2 ± 0.4
$^{235}U_3O_8$	145.6	156.0 ± 0.9	157.2 ± 0.6	57.66	62.4 ± 0.6	63.1 ± 0.4
$^{235}UF_4$	171.3	181 ± 1	186.2 ± 0.7	67.90	73.7 ± 0.6	74.9 ± 0.4
$^{235}UF_6$	300.9	322 ± 2	331 ± 1	111.7	120 ± 1	124.1 ± 0.7
$^{239}PuO_2$	31.55	32.8 ± 0.2	31.8 ± 0.1	13.66	14.6 ± 0.1	14.16 ± 0.07
$^{239}Pu_2O_3$	32.03	33.0 ± 0.2	32.04 ± 0.09	13.46	14.3 ± 0.1	13.90 ± 0.07
$^{239}PuF_3$	43.31	44.3 ± 0.2	44.0 ± 0.1	18.05	18.9 ± 0.1	18.72 ± 0.09
$^{239}PuF_4$	71.46	73.2 ± 0.4	73.4 ± 0.3	27.24	28.7 ± 0.2	28.6 ± 0.1
$^{239}PuCl_3$	220.9	227 ± 1	224.9 ± 0.7	71.50	73.7 ± 0.6	72.5 ± 0.4

[a] Mass value is total moist compound mass.

IV. REFERENCES

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3. "Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors," American National Standard ANSI/ANS-8.1-1983-R88.