Benchmark Critical Experiment of a Water Reflected Alpha-Phase Plutonium Sphere



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# BENCHMARK CRITICAL EXPERIMENT OF A WATER REFLECTED ALPHA-PHASE PLUTONIUM SPHERE

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### **IDENTIFICATION NUMBER:** PU-MET-FAST-011

**KEY WORDS:** acceptable, alpha-phase plutonium metal, critical experiment, fast, homogeneous, reflected plutonium metal sphere, unmoderated, water-reflected

### **1.0 DETAILED DESCRIPTION**

### 1.1 Overview of Experiment

In 1968, an experiment was performed at Los Alamos Scientific Laboratory using two subcritical spherical masses of alpha-phase plutonium reflected by water. From the two subcritical inverse multiplication measurements, an accurate prediction was made of the critical mass of an alpha-phase plutonium sphere reflected by water. The result of this experiment is considered to be acceptable as a benchmark critical experiment.

### 1.2 Description of Experimental Configuration

The experiment was performed using a plutonium sphere reflected by water. The sphere was painstakingly fabricated to obtain highly pure and highly dense plutonium. The plutonium was electrorefined to produce metal with total impurities of 230 ppm and cast into two split ingots. Two hemispheres were constructed from the split ingots, and the pieces were shrink-fitted<sup>a</sup> together and machined to exacting tolerances. Finally, the hemispheres were encased in 0.175-inch thick Lucite<sup>b</sup> that had been previously electroplated on the inside with 0.0005-in.-thick copper (Reference 1).

Figure 1 shows the experimental setup (Reference 1). The plutonium sphere was placed on a stand. The stand was constructed of Lucite. The seat of the stand was a 1.0-in.-thick circular disk with a central hole. It was supported on three legs about 10 in. long. The inside diameter of the seat was approximately 2.125 in., and the outside diameter was 10.0 in. (Reference 4). The stand was situated inside a 2-ft-diameter cylindrical aluminum-run tank. The run tank was attached to the fill tank by a flexible hose. The fill tank rode on a hydraulic lift. The operator raised the water level in the run tank by raising the fill tank with the hydraulic lift. The operator

<sup>&</sup>lt;sup>a</sup> Reference 1 describes shrink-fitted as a process whereby "the male piece was cooled with dry ice while the female piece self-heated."

<sup>&</sup>lt;sup>b</sup> There is some uncertainty associated with the thickness of the Lucite. Reference 1 gives the thickness as both 0.175 and 0.150 inches; Reference 2 gives the thickness as "about" 5/16 inch.

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lowered the run tank water level either by using the drain valve or by lowering the fill tank using the hydraulic lift

Two detectors were attached to the outside of the run tank, and two detectors were placed inside a cylindrical pipe 3 in. below the sphere. A  $^{252}$ Cf source inside an aluminum ball was used to calibrate detector response as a function of water height (References 1 and 2).

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Determination of the critical mass was accomplished by means of extrapolation to the critical radius using reciprocal multiplication measurements for two subcritical alpha-phase spheres (5546 and 5316 gram, respectively). Both measurements, for the purpose of extrapolation to critical, were taken with the run tank completely filled with water.

After the first subcritical measurement, the sphere was machined to reduce the mass, and a second subcritical experiment was performed. At the time of machining, it was observed that a 4 mm low density oxide film had formed on the equatorial parting plane which reduced the overall density of the second sphere to 19.68 g/cm<sup>3</sup>, as opposed to 19.74 g/cm<sup>3</sup> for the first sphere (Reference 1).

Originally, the plan had been to refabricate a larger sphere whose mass was halfway between the first sphere and the new empirical estimate of critical mass, but plans were revised to machine down the first sphere. This was done for two reasons:

- 1. Final count rate of the first sphere was so high that doubling the count rate would have saturated the counting system.
- 2. If the metal were recast, the density might have been significantly different.

The corrected inverse multiplications were given in Reference 1 and reproduced in Table 1. The corrections are discussed in more detail in Section 2.0 and Appendix B. The corrected inverse multiplications were used to extrapolate to critical as shown in Figure 2.

	Mass	1/M
Second Sphere	5343	-
Density Adjusted to 19.74 g/cm <sup>3</sup>	5316	0.03604
First Sphere	5546	0.01844
Critical Mass at 19.74 g/cm <sup>3</sup>	5790	0

Table 1.	Extrapolation Data.
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Figure 2. Plutonium Sphere, 10 inch Water Reflector.

A plot of inverse multiplication versus the height of the water in the tank for the first sphere was given in Reference 1 and is reproduced here as Figure 3. Small amounts of water were added to the tank, and the count rate was observed and recorded at each increment. After ~2 inches of water covered the sphere (water height  $\approx$  16 in.), the count rate increase, and, therefore, the increase in multiplication due to a reduction in leakage, was small (see Figure 3).



Figure 3. 5546 gram Plutonium Ball with 5/16 inch Thick Lucite Shell on Stool.

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# 1.3 Description of Material Data

The composition of the alpha-phase plutonium core is given in Table 2 (Reference 1). The impurities in the core are shown in Table 3 (Reference 3). The impurities are average values based upon before and after samples of the core.<sup>a</sup>

Isotope	At.%
<sup>239</sup> Pu	94.50%
<sup>240</sup> Pu	5.18%
<sup>241</sup> Pu	0.30%
<sup>242</sup> Pu	0.02%

Table 2. Isotopic Abundances of the Core.

<sup>&</sup>lt;sup>a</sup> Personal communication, D. R. Smith, December 1994.

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Element	Concentration (ppm)
Americium	90
Tungsten	60
Carbon	25
Oxygen	20
Silicon	7
Iron	5
Nickel	4
Nitrogen	4
Copper	3
Thorium	3
Magnesium	1
Gallium	0.5
Aluminum	0.5
Manganese	0.2

# 1.4 <u>Supplemental Experimental Measurements</u>

No additional experimental data were found.

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## 2.0 EVALUATION OF EXPERIMENTAL DATA

No significant deficiencies exist in the published data. The Lucite stand upon which the plutonium sphere sat was not physically described in the references. It was determined, however, that the stand was similar to that used for the water reflected oralloy sphere experiment.<sup>a</sup> The stand was made of Lucite. The seat of the stand was approximately a 1.0-in.-thick circular disk with a central hole. It was supported on three legs about 10 in. long. The inside diameter of the seat was approximately 2.125 in., and the outside diameter was 10.0 in. (Reference 4 and HEU-MET-THERM-001). The worth of the stand was determined by placing a second stand on top of the plutonium sphere.

No mention was made as to the proximity of the room walls, but this fact is moot because the water reflector was essentially infinitely (10 inches) thick (see Appendix B, Section B.2). No correction was made for the fact that the run tank was cylindrical rather than spherical. The thick water reflection in all directions (radially and axially) negated any geometrical effects of the run tank.

Corrections were made by the experimenters to account for the effects of the Lucite stand  $(+0.0024 \ \Delta k)$  and the Lucite shell  $(+0.0020 \ \Delta k)$ . These corrections were measured by situating a second Lucite stand and then a second Lucite shell in the assembly.<sup>b</sup> A third correction, density, was derived from the approximation that critical mass is inversely proportional to density raised to the 1.6 power (see Appendix C). Finally, the critical mass of a simple Pu sphere with thick water reflection was derived from a straight line extrapolation, by radius, using the three previous corrections and the two experimental configurations (the two different masses of plutonium spheres). This idealized one dimensional, water-reflected 5790 ± 25 gram Pu sphere is what is accepted as the calculational benchmark model. The sensitivity of the calculational model to various parameters is assessed in Appendix B, Section B.1.

<sup>&</sup>lt;sup>a</sup> Personal communication, R. E. Anderson, November 1993.

<sup>&</sup>lt;sup>b</sup> Personal communication, D. R. Smith, December 1993.

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## **3.0 BENCHMARK SPECIFICATIONS**

## 3.1 Description of Model

The idealized experimental benchmark model is a simple alpha-phase plutonium sphere with a density of  $19.74 \text{ g/cm}^3$  and a mass of 5790 grams reflected by a 10-inch-thick spherical shell of water (References 1 and 2). The model is an idealized configuration derived by the experimenters.

### 3.2 Dimensions

The radius of the 5790 gram alpha-phase plutonium sphere with a density of 19.74 g/cm<sup>3</sup> was 4.1217 cm. The sphere was reflected by 10 inches of water.

### 3.3 Material Data

The calculated atom densities, of the alpha-phase plutonium sphere for the isotopic composition given previously in Tables 2 and 3, are shown in Table 4. The small amount of <sup>242</sup>Pu is replaced with <sup>240</sup>Pu. As shown by the sensitivity studies in Appendix B, Section B.1, the effect of impurities in the plutonium was negligibly small, so the impurities are omitted for simplicity.

Isotope/Element	Atom Density (atoms/barn-cm)	
α-phase Pu		
<sup>239</sup> Pu	4.6982×10 <sup>-2</sup>	
<sup>240</sup> Pu	2.5852×10 <sup>-3</sup>	
<sup>241</sup> Pu	1.4915×10 <sup>-4</sup>	
<sup>242</sup> Pu	9.9432×10 <sup>-6</sup>	
Water		
Hydrogen	6.6766×10 <sup>-2</sup>	
Oxygen	3.3383×10 <sup>-2</sup>	

Table 4. Atom Densities

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### 3.4 <u>Temperature Data</u>

No mention is made in the references in regard to experimental temperature. The experimental temperature is assumed to be at room temperature (293 K).

## 3.5 Experimental and Benchmark-Model keff

The experimental  $k_{eff}$  for the first plutonium sphere with corrections was about 0.98, which, assuming  $k_{eff}$  is "close to one,"<sup>a</sup> corresponds to a normalized inverse multiplication of 0.01844. The idealized benchmark-model  $k_{eff}$  is 1.000 ± 0.001. The uncertainty in  $k_{eff}$  is due to the 25 gram uncertainty in the critical mass reported by the experimenters.

<sup>&</sup>lt;sup>a</sup> Inverse multiplication is related to  $k_{eff}$  by:  $1/M = 1 - k_{eff}$ . This relationship is valid if  $k_{eff}$  is "close to one." The statement, "close to one," varies depending on the system.

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# 4.0 RESULTS OF SAMPLE CALCULATIONS

The idealized experimental configuration is a simple one dimensional sphere, and, therefore, it can easily be calculated with any computational code. The results of these calculations are shown in Table 5. Input listings are given in Appendix A.

KENO (Hansen-Roach)	KENO (27-Group ENDF/B-IV)	MCNP (Continuous Energy ENDF/B-V)	ONEDANT (27-Group ENDF/B- IV)
$1.0009 \pm 0.0016$	$1.0016 \pm 0.0013$	$0.9993 \pm 0.0011$	1.0034

 Table 5. Sample Calculation Results (United States).

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## **5.0 REFERENCES**

- 1. D. R. Smith and W. U. Geer, "Critical Mass of a Water-Reflected Sphere," Nuc. Applic. Technol., 7, pp. 405-408, November 1969.
- 2. D. R. Smith and W. U. Geer, "Measurement of the Critical Mass of a Water Reflected Plutonium Sphere," Transactions of the American Nuclear Society, **11**, 378, June 1968.
- 3. G. E. Hansen and H. C. Paxton, "Reevaluated Critical Specifications of Some Los Alamos Fast - Neutron Systems," LA-4208, September 1969.
- 4. C. C. Byers, J. J. Koelling, G. E. Hansen, D. R. Smith, and H. R. Dyer, "Critical Measurements of a Water-Reflected Oralloy Sphere," Transactions of the American Nuclear Society, 27, 412, November 1977.
- 5. K. D. Lathrop, "DTF-IV, A FORTRAN-IV Program for Solving the Multi-group Equation with Anistropic Scattering," LA-3373, 1965.
- 6. H. C. Paxton, "Criticality Control in Operations with Fissile Material," LA-3366, p. 25, November 1972.

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# **APPENDIX A: TYPICAL INPUT LISTINGS**

## A.1 KENO Input Listings

### Hansen-Roach 16-Group Cross Sections and 27-Group ENDF/B-IV Cross Sections

Listed below are the input files for KENO V.a with 16-group Hansen-Roach and 27-group SCALE4 cross sections for the sample calculations. Both files use 300 active generations with 1500 histories per generation after skipping the first five generations.

KENO-V.a Input Listing for Table 5 (16-Energy-Group Hansen-Roach Cross Sections).

KENO V.a with Hansen-Roach cross sections input file WATER REFLECTED PU(5.2) SPHERE READ PARAM RUN=yes FAR=YES LIB=41 GEN=305 NPG=1500 NSK=5 END PARAM READ MIXT SCT=1 MIX=1 94901 0.046982 94001 0.0025852 94100 0.00014915 MIX=2 1101 0.066766 8100 0.033383 END MIXT READ GEOM UNIT 1 SPHERE 1 1 4.1217 SPHERE 2 1 29.5217 END GEOM END DATA END

KENO-V.a Input Listing for Table 5 (27-Energy-Group SCALE4 Cross Sections).

SCALE with 27 group ENDF/B-IV cross sections input file =CSAS25 PU(5.2) WATER REFLECTED SPHERE 27GROUPNDF4 INFHOMMEDIUM PU-239 1 0 0.046982 END PU-240 1 0 0.0025852 END PU-241 1 0 0.00014915 END H 200.066766 END 0 200.033383 END END COMP READ PARAMETERS TME=1000 TBA=10 GEN=305 NPG=1500 NSK=5 END PARAMETERS READ GEOM UNIT 1 SPHERE 1 1 4.1217 SPHERE 2 1 29.5217 END GEOM END DATA END

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# A.2 MCNP Input Listings

Listed below is the input file for MCNP 4.2 with continuous-energy ENDF/B-V cross sections. The input file uses 300 active generations with 1500 histories per generation after skipping the first 10 generations.

MCNP Input Listing for Table 5.

WATER REFLECTED PU(5.2) SPHERE 1 1 0.04971635 -1 imp:n=1 2 2 0.100149 1 -2 imp:n=1 2 imp:n=0 3 0 1 so 4.1217 2 so 29.5217 m1 94239.55c 0.046982 94240.50c 0.0025852 94241.50c 0.00014915 m2 1001.50c 0.066766 8016.50c 0.033383 mt2 lwtr.01t kcode 1500 1.0 10 310 ksrc 000 print

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## A.3 **ONEDANT Input Listings**

Listed below are the input files for ONEDANT version 2.3h.2 with 27-group SCALE4 cross sections which have  $P_3$  scatter data. The quadrature set is  $S_{48}$ . The convergence criteria is  $10^{-4}$  for eigenvalue and flux by default. The mesh size is approximately 19 mesh/cm in the core and 2 mesh/cm in the reflector. The core was heavily meshed to ensure proper convergence of the eigenvalue and the flux. The first input file listed is used to generate the 27-group SCALE cross sections for ONEDANT.

ONEDANT Input Listing for Table 5.

=CSASI RUN TO GET XSCTS FOR WATER REFLECTED PU(5.2) SPHERE 27GROUPNDF4 INFHOMMEDIUM 
 PU-239
 1 0 0.046982 END

 PU-240
 1 0 0.0025852 END

 PU-241
 1 0 0.00014915 END
 200.066766 END Η 200.033383 END 0 END COMP END 1 Water reflected Pu(5.2) sphere /BLOCK 1 igeom=sph ngroup=27 niso=2 isn=48 mt=2 nzone=2 im=2 it=130 t /BLOCK 2 xmesh=0.0,4.1217,29.5217 xints=80,50 zones=1,2 t /BLOCK 3 lib=xs27.w12 maxord=3 ihm=42 iht=3 ihs=16 ititl=1 ifido=2 i2lp1=1 t /BLOCK 4 matls=isos assign=matls t /BLOCK 5 chi=.026 .203 .217 .123 .161 .172 .084 .013 .001 18z ievt=1 isct=3 t

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# **APPENDIX B: SENSITIVITY STUDIES**

### **B.1** Worth of Corrections and Other Parameters

The results of calculations to determine the sensitivity of the model to numerous parameters are reported in this Appendix. ONEDANT/TWODANT with Hansen-Roach cross sections was used for the sensitivity studies. The results are shown in Table B.1. Based on these calculations and the opinions of the experimenters, the effects of the copper electroplate, impurities, and the aluminum run tank, as well as anything outside of the run tank, are negligible. The Lucite shell accounts for a  $\Delta k$  of 0.0022 (0.0020  $\Delta k$  experimentally), and the Lucite stand accounts for a  $\Delta k$  of 0.0010 (0.0024  $\Delta k$  experimentally).

Effect	$\Delta k_{ m eff}^{a}$
0.0005 inch Copper Plate	+0.0000
Impurities (concentration ≥ 10 ppm)	+0.0007
±25 grams	±0.0012
5/16 inch Lucite Shell	+0.0022
Aluminum Run Tank	+0.0000
Room Return	+0.0000
1.0 inch Lucite Collar (approximation for the Lucite Stand)	+0.0010

Table B.1.	Sensitivity	Studies.
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In the previous table, the difference between the experimental  $\Delta k$  and the calculated  $\Delta k$  of the Lucite shell and stand appears to be larger than the  $\Delta k$  associated with the 25 gram uncertainty. However, the corrections are applied, and the resultant inverse multiplications are extrapolated to zero. Therefore, a direct correlation between the  $\Delta k$  associated with the uncertainty and the  $\Delta k$  associated with the corrections is not valid, see Section B.3 for further explanation.

 $<sup>^{\</sup>rm a}~$  The  $\Delta k_{\rm eff}$  is relative to a base case of 1.0023.

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## **B.2** Justification of Infinite Water Reflection

A sensitivity study was performed using ONEDANT with Hansen-Roach cross sections to show the effect of changing the thickness of the water. The results are shown in Figure B.1. The reflector is infinitely thick at 10.0 in.



Figure B.1. Plot of k<sub>eff</sub> versus Reflector Thickness.

# **B.3** Calculation of the Worth of the Lucite Structures

Figure B.2 shows the extrapolation by radius of the corrected inverse multiplication. Corrections were made for the Lucite stand and shell by subtracting the increase in multiplication measured for the addition of a second stand and shell from the measured value for a single stand and shell to obtain the predicted multiplication for no stand or shell. The extrapolation to a 1/M of 0.0 was accomplished by straight line fit to the data for the two alphaphase plutonium spheres. The predicted critical radius was found to be 4.1217 cm.

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Figure B.2. Extrapolation, by Radius, to 1/M of 0.0 (corrected 1/M).

Figure B.3 shows the extrapolation by mass of the corrected inverse multiplication (corrections made for the Lucite stand and the Lucite shell). The extrapolation to a 1/M of 0.0 was accomplished by straight line fit to the data for the two alpha-phase plutonium spheres. The predicted critical mass was found to be 5787 grams, which is in excellent agreement with the 5790 grams, which can be derived from the predicted critical radius of Figure B.2.



Figure B.3. Extrapolation, by Mass, to a 1/M of 0.0 (corrected 1/M).

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Figure B.4 shows the extrapolation by mass of the uncorrected inverse multiplication. The extrapolation to a 1/M of 0.0 was accomplished by a linear fit to the data for the two alpha-phase plutonium spheres. The predicted critical mass for the uncorrected multiplications was found to be 5730 grams.



Figure B.4. Extrapolation, by Mass, to Critical (uncorrected 1/M).

Using the predicted critical masses shown by the previous two figures, the  $\Delta k$  associated with the Lucite stand and shell was calculated. The results are shown in Table B.2. ONEDANT with 16-group Hansen-Roach, 27-group ENDF/B-IV, and 30-group ENDF/B-V cross sections was used for these calculations. The calculated  $\Delta k$  is approximately 0.0030 for all cross section sets described above. The reported  $\Delta(1/M)$  of the stand and shell was 0.0044.  $\Delta k$  is equal to  $\Delta(1/M)$  if  $k_{eff}$  is "close to one."

Table B.2. Results of Calculations to Assess the  $\Delta k$  Associated with the Lucite Stand and Shell

Cross Sections	Mass=5730 grams	Mass=5790 grams	$\Delta \mathbf{k}$
ENDF/B-IV, 27 Group	1.0020	1.0050	0.0030
Hansen-Roach	0.9985	1.0013	0.0028
ENDF/B-V, 30 Group	1.1072	1.1101	0.0029

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Figure B.5 shows the results of extrapolation to critical based upon a  $\Delta 1/M$ , [= $\Delta k$ ], of 0.0030 calculated correction for the Lucite stand and shell. The extrapolated critical mass for this  $\Delta k$  is 5770 grams. The change in critical mass using the calculated worth of the Lucite is 20 grams, which is within the given experimental uncertainty of ±25 grams.



Figure B.5. Extrapolation to Critical Based on the Calculated  $\Delta k$  Associated with the Lucite Stand and Shell.

# **B.4** Verification of Linear Extrapolation

Calculations were conducted to ensure that a linear extrapolation through the two experimentally determined points is valid. Multiplication calculations were performed using ONEDANT for various plutonium masses to show that the linearity assumption is valid. The results of these calculations were used to extrapolate to the critical mass. The results are shown in Figure B.6 and Table B.3. The predicted critical mass was found to be 5795 grams, calculationally. Results of this study are within the 25 gram uncertainty of the experimentally predicted critical mass of 5790 grams.

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Mass (grams)	Corrected 1/M
5316	0.03555
5400	0.02915
5450	0.02545
5546	0.01837
5600	0.01450
5700	0.00710

# Table B.3. Multiplication Calculation Results.



Figure B.6. Extrapolation, by Mass, to Critical (corrected 1/M).

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# **APPENDIX C: EXPLANATION OF THE** $m_{critical} \propto \rho^{-1.6}$ **RELATIONSHIP**

The relationship,  $m_{critical} \propto \rho^{-2}$  for a bare system, is easily derived from the following:

### $\rho_1 r_1 = \rho_2 r_2$ then $k_1 = k_2$ ,

where  $\rho$  is the density, and *r* is the characteristic dimension of the system, and *k* is the effective multiplication factor of the system (Reference 6).

$$\frac{\rho_1}{\rho_2} = \frac{r_2}{r_1}$$

$$(\frac{\rho_1}{\rho_2})^3 = (\frac{r_2}{r_1})^3$$

$$\frac{V_2}{V_1} = \frac{\frac{4}{3}\pi r_2^3}{\frac{4}{3}\pi r_1^3} = (\frac{r_2}{r_1})^3$$

$$(\frac{\rho_1}{\rho_2})^3 = \frac{V_2}{V_1}$$

$$\frac{\rho_1 V_1 = m_1}{\rho_2 V_2 = m_2}$$

$$\rho_1^3 V_1 = \rho_2^3 V_2$$

$$(\rho_1 V_1) = \rho_2^2 (\rho_2 V_2)$$

 $\rho_1^2 m_1 = \rho_2^2 m_2$ 

 $\rho_1^2 \\$ 

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$$m_2 = m_1 (\frac{\rho_1}{\rho_2})^2$$

If the system is not bare, then:

$$m_2 = m_1 \left(\frac{\rho_1}{\rho_2}\right)^n$$

where *n* is an arbitrary constant dependant on the reflector. Normally n = 1.6 for a close fitting reflector, but *n* always lies between 1 and 2. The value of *n* was derived from many discrete ordinates calculations using the DTF code (Reference 5) performed at the time of the experiment.

$$m_{critical} = 5343 \left(\frac{19.68}{19.74}\right)^{1.6} = 5317 \ grams$$

The calculational results in Reference 1 show a mass of 5316 grams for the second sphere using the previously described density correction. These calculations were reproduced as shown above with a calculated mass of 5317 grams. This is believed to be due to round-off error. This difference is well within the reported uncertainty of  $\pm 25$  grams in the final critical mass.



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