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Minimum Critical Values Study

July 2005

Prepared by
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MINIMUM CRITICAL VALUES STUDY

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ABSTRACT

This report provides minimum critical values for various 30-cm water-reflected uranium and plutonium oxide and nitrate aqueous mixtures as calculated by the SCALE CSAS1X sequence using the 238-group ENDF/B-V neutron cross-section library. The minimum values were determined through parametric searches in one-dimensional geometry.

The calculations have been performed to obtain the minimum values: critical volume and mass for spheres, critical radius for cylinders, critical thickness for slabs, and minimum critical concentration (infinite geometry) for the following homogenous mixtures:

1. $\text{UO}_2\text{-H}_2\text{O}$ for 3, 4, 5, 20, and 100 wt % ^{235}U ;
2. UNH for 3, 4, 5, 20, and 100 wt % ^{235}U ;
3. $\text{PuO}_2\text{-H}_2\text{O}$ for 100/0/0, 95/5/0, 90/5/5, 80/10/10, and 71/17/11/1 wt % of $^{239}\text{Pu}/^{240}\text{Pu}/^{241}\text{Pu}(/^{242}\text{Pu})$; and
4. PuNH for 100/0/0, 95/5/0, 90/5/5, 80/10/10, and 71/17/11/1 wt % of $^{239}\text{Pu}/^{240}\text{Pu}/^{241}\text{Pu}(/^{242}\text{Pu})$.

All bounding surfaces were fully reflected by 30 cm of H_2O .

1. BACKGROUND AND PURPOSE

For several decades, and on a national and international basis, minimum critical values of fissionable systems characteristic of those found in fuel cycle operations have been used in consensus standards as technical guidance for establishing safe operational limits. In the mid-1990s, the international criticality safety community recognized that it would be highly desirable to develop an international consensus on minimum critical values for a variety of simple systems fueled with uranium and plutonium. An effort was initiated through the Organization for Economic Cooperation and Development (OECD) Nuclear Science Committee to address the problem of developing this consensus. Six countries have participated in this effort, and the results contained in this document detail the contribution from the United States.

The current status of the international effort was reported at the Seventh International Conference on Nuclear Criticality, ICNC2003 (ref. 1). The material described below was excerpted from this report.

An OECD/NEA expert group has compiled international data on existing minimum critical values for UO_2 , PuO_2 , UNH and PuNH systems to identify significant discrepancies in the data and to propose explanations. Minimum critical values given by criticality handbooks and other sources are known to show discrepancies that are sometimes considerable. Being unique like physical constants, minimum critical values should be the same, nevertheless. This is important with regard to licensing, though minimum critical values are used primarily in connection with safety factors. These discrepancies were brought to the attention of the OECD/NEA Working Party for Nuclear Criticality Safety (WPNCS) who appointed an expert group to investigate the issue.

The expert group confined the investigation to homogeneous systems. The work started with the compilation of a quite large number of chemical compositions of U-235, U-233, and plutonium, all with different reflectors. With over 900 data entries, the analytical effort was too high. The group thus focused on aqueous systems of UO_2 , PuO_2 , uranium-nitrate and plutonium-nitrate, with five enrichments of uranium and six Pu vectors, respectively.

With this scope of investigation, the objectives of the expert group became to

- collect data from different countries, including a short description of the methods used to derive the data;
- identify discrepancies and propose explanations;
- address effects of engineering data, density formulae, and reflector material;
- provide technical input to the international community; and
- supply a general reference for criticality safety analyses that use/include minimum critical values.

The minimum system parameters established for the critical systems include the spherical mass (kg of U or Pu), the spherical volume (liters), and the diameter (cm) and thickness (cm) of the axially infinite cylinder and the infinite slab, as well as the minimum concentrations (g U or Pu per liter) for fully infinite systems. Generally, the results submitted within the past 4 years agree to within 17% (spherical volume) for the nitrate-moderated systems. The PuO_2 systems generally agree to within 10% (spherical mass). The low-enriched UO_2 systems agree to within 19%, but the agreement is worse for the more highly enriched systems [20% enriched—33% (spherical mass), 100% enriched—36% (spherical volume)]. The complete summary of these results is given in Table 5 of this report.

The work reported in this document was performed as a subtask of the Applicable Ranges of Bounding Curves and Data (AROBCAD) work element of the U.S. Department of Energy (DOE)

Nuclear Criticality Safety Program. Again, it constitutes the contribution from the United States to this international comparison of minimum critical values.

2. INTRODUCTION

The OECD/Nuclear Energy Agency (OECD/NEA) Nuclear Science Committee Working Party on Nuclear Criticality Safety formed the Expert Group on Minimum Critical Values. The goal of the first task was to prepare a database of minimum critical values (mass, volume, radius, slab thickness, concentration, enrichment) that are often used as a basis for criticality safety considerations. Calculations reported in this document were performed with the CSAS1X sequence in SCALE² (Standardized Computer Analyses for Licensing Evaluation) using the 238-group ENDF/B-V cross-section library³ to obtain the minimum critical values for the following homogenous mixtures:

1. $\text{UO}_2\text{-H}_2\text{O}$ for 3, 4, 5, 20, and 100 wt % ^{235}U ;
2. UNH [i.e., $\text{UO}_2(\text{NO}_3)_2 + x\text{H}_2\text{O}$] for 3, 4, 5, 20, and 100 wt % ^{235}U ;
3. $\text{PuO}_2\text{-H}_2\text{O}$ for 100/0/0, 95/5/0, 90/5/5, 80/10/10, and 71/17/11/1 wt % of $^{239}\text{Pu}/^{240}\text{Pu}/^{241}\text{Pu}(/^{242}\text{Pu})$; and
4. PuNH [i.e., $\text{Pu}(\text{NO}_3)_4 + x\text{H}_2\text{O}$ for 100/0/0, 95/5/0, 90/5/5, 80/10/10, and 71/17/11/1 wt % of $^{239}\text{Pu}/^{240}\text{Pu}/^{241}\text{Pu}(/^{242}\text{Pu})$.

All bounding surfaces were fully reflected by 30 cm of H_2O .

The computational biases for the computed critical systems reported herein were not determined. However, the reader is directed to *Experience with the Scale Criticality Safety Cross-Section Libraries*.⁴ The latter report provides insights into computational biases that have been observed with the use of the computational methods utilized for the development of the information in the present report. Relative to the determination of the nearly unique minimum critical values when nearly optimally moderated with light water, computational biases are well characterized. For nearly optimally moderated systems, the computed values of k_{eff} for critical systems are approximately 1.00 ± 0.02 , 1.005 ± 0.015 , 1.015 ± 0.02 for high-enriched, low-enriched, and plutonium systems, respectively.

3. DESCRIPTION OF THE CODE PACKAGE

3.1. CSAS1X SEQUENCE

CSAS1X creates a microscopic cell-weighted library in the AMPX working library format. This control module sequentially activates the functional modules BONAMI and NITAWL-II to process the necessary cross sections and calculates the k-effective of the one-dimensional (1-D) problem using the discrete-ordinates code XSDRNPM. CSAS1X can perform a 1-D geometry search for these critical values: radius of sphere and cylinder, and slab thickness.

3.2. 238-GROUP NEUTRON CROSS-SECTION LIBRARY

The 238-group ENDF/B-V library is a general-purpose criticality analysis library and is the most complete library available in SCALE. This library is also known as the LAW (Library to Analyze Radioactive Waste) Library. It was initially released in version 4.3 of SCALE. The library contains data for all nuclides (more than 300) available in ENDF/B-V processed by the AMPX-77 system.⁵ It also contains data for ENDF/B-VI evaluations of ¹⁴N, ¹⁵N, ¹⁶O, ¹⁵⁴Eu, and ¹⁵⁵Eu. The library has 148 fast groups and 90 thermal groups (below 3 eV).

Most resonance nuclides in the 238-group ENDF/B-V library have resonance data (to be processed by NITAWL-II) in the resolved-resonance range and Bondarenko factors (to be processed by BONAMI) for the unresolved range. This library contains resolved-resonance data for *s*-wave, *p*-wave, and *d*-wave resonances ($l = 0$, $l = 1$, and $l = 2$, respectively). These data can have a significant effect on results for undermoderated, thermal, and intermediate-energy problems. Resonance structures in several light to intermediate-mass ENDF nuclides (i.e., ⁷Li, ¹⁹F, ²⁷Al, ²⁸Si) are accounted for using Bondarenko shielding factors. These structures can also be important in intermediate-energy problems. The ²³⁵U ENDF/B-V data result in slightly too much fission, while the ²³⁸U data result in slightly too much capture. Although better than the ENDF/B-IV data, the thermal-plutonium data still appear to have problems.

All nuclides in the 238-group LAW Library use the same weighting spectrum, consisting of

1. Maxwellian spectrum (peak at 300 K) from 10^{-5} to 0.125 eV,
2. a $1/E$ spectrum from 0.125 eV to 67.4 keV,
3. a fission spectrum (effective temperature at 1.273 MeV) from 67.4 keV to 10 MeV, and
4. a $1/E$ spectrum from 10 to 20 MeV.

The use of these spectra [as opposed to the $1/(E\sigma_i)$ spectrum used to generate the 218-group library] makes it difficult to collapse a general-purpose broad-group library that is valid over a wide range of problems.

All nuclides use a P_5 Legendre expansion to fit the elastic and discrete-level inelastic scattering processes in the fast range, thereby making the library suitable for both reactor and shielding applications. A P_3 fit was used for thermal scattering. Thermal scattering kernels are provided at temperatures (K) as presented in Table M4.2.7 of the SCALE manual. All other scattering processes use P_0 fits.

3.3. CALCULATION METHODS

The 238-group cross-section library and the CSAS1X sequences in SCALE were used to determine critical data for spheres, cylinders, and slabs. The discrete-ordinates code XSDRNPM was used to

generate the calculated values. The XSDRNPM overall point convergence is 1.0E–06, while the meshing is generated by SCALE. Cross sections were calculated with a temperature of 293 K.

Default values in SCALE have been used whenever considered appropriate or when no other information is known. If SCALE generates a warning or error message suggesting changes to default parameters, the default values may be changed.

The compositions of the homogeneous uranium oxide–water mixture and the plutonium oxide–water mixture were calculated using available options in CSAS1X. The default density of 10.96 g/cm³ was used for UO₂, and 11.86 g/cm³ was used for PuO₂. The density of H₂O was 0.9982 g/cm³ for both mixtures.

The homogeneous uranium nitrate–water mixture and plutonium nitrate–water mixture were calculated using the built-in SCALE atomic number density equations for uranyl nitrate [UO₂(NO₃)₂] solution and for plutonium nitrate [(Pu(NO₃)₄)] solution per ARH-600 (ref. 6).

The equations for UO₂(NO₃)₂ solution are as follows:

$$\rho_{UO_2(NO_3)_2} = \rho_U (A_U + 8A_O + 2A_N) / A_U ,$$

$$\rho_{HNO_3} = M_{HNO_3} (A_H + A_N + 3A_O) / 1000 ,$$

and

$$\rho_{soln} = \left[\frac{1.0012 + 317.7 * \rho_U / A_{NU} + 0.03096 * M_{HNO_3}}{(1 - 5 * 10^{-4} \Delta T) + 1.45 * 10^{-4} \Delta T} \right]$$

The equations for (Pu(NO₃)₄) solution are as follows:

$$\begin{aligned} \rho_{soln} = & 0.9982 * \rho_{P_U} / A_{N_P} (1 - 0.3619 * \rho_{P_U} - 0.0246 * M_{HNO_3}) \\ & + \rho_{P_U(NO_3)_4} + \rho_{HNO_3} , \end{aligned}$$

$$\rho_{P_U(NO_3)_4} = \rho_{P_U} (A_{P_U} + 12A_O + 4A_N) / A_{P_U} ,$$

and

$$\rho_{HNO_3} = M_{HNO_3} (A_H + A_N + 3A_O) / 1000 .$$

In both sets of equations, the parameter definitions are as follows:

ρ_i = density of nuclide (g/cm³),

M_{HNO_3} = molarity (mol/L),

A_i = isotopic mass (g/mol),

ΔT = (T_{°C} – 22.5).

4. CALCULATIONS

4.1. URANIUM OXIDE–WATER MIXTURES

The uranium isotopic compositions involve only ^{235}U and ^{238}U . The following uranium oxide enrichments have been considered in this study: U(3), U(4), U(5), U(20), and U(100) wt % ^{235}U .

In all calculations the composition of the homogeneous uranium oxide–water mixture was calculated using available options in CSAS1X. The default density value of 10.96 g/cm^3 was used for UO_2 , and 0.9982 g/cm^3 was used for the density of H_2O . Cross sections were calculated with a temperature of 293 K.

A series of CSAS1X calculations of the critical parameters (dimensions, masses, and concentrations) with varying volume fractions of UO_2 and H_2O were performed for spheres, cylinders, slabs, and infinite media. Plots of these calculations are shown in Figs. A.1–A.20 (Appendix A). For each sphere radius, mass was calculated as density times volume, where density was calculated as $N_{235} * 235.0441 / 0.60221368$ (ref. 7) and the volume is from the output of the calculation. Total grams of uranium as shown in Table 1 are grams of ^{235}U divided by weight fraction of ^{235}U .

Concentration was calculated as $10.96 \text{ g UO}_2/\text{cm}^3 * \text{VF} * (\text{U}/\text{UO}_2) * 1000$, where VF is the volume fraction of the CSAS1X case where k-infinity equals 1. The value of U/UO_2 was calculated as $(^{235}\text{U} \text{ wt \% * } 235.0441 + ^{238}\text{U} \text{ wt \% * } 238.0510) / (^{235}\text{U} \text{ wt \% * } 235.0441 + ^{238}\text{U} \text{ wt \% * } 238.0510 + 2 * 15.9906)$. Atomic weights are from the SCALE Standard Composition Library (Sect. M8) based on ENDF/B-V data.

4.2. URANIUM NITRATE–WATER SOLUTIONS

The uranium isotopic compositions involve only ^{235}U and ^{238}U . The following uranium nitrate enrichments have been considered in this study: U(3), U(4), U(5), U(20), and U(100) wt % U.

The composition of the homogeneous uranium nitrate–water material was calculated using the built-in SCALE atomic number density equations for uranyl nitrate ($\text{UO}_2(\text{NO}_3)_2$) solution per ARH-600 (ref. 6). The uranium density of a hydrated uranyl nitrate crystal is about 1296 g/L , which is about the density of the 3% minimum volume. Cross sections were calculated with a temperature of 293 K.

A series of CSAS1X calculations of the critical parameters (dimensions, masses, and concentrations) with varying uranium density were performed for spheres, cylinders, slabs, and infinite media. Plots of these calculations are shown in Figs. B.1–B.20 (Appendix B). SCALE warns the user when the uranium density is higher than what a real solution would support. If the input uranium density is higher than the theoretical density for a hydrated uranyl crystal, an error is given by SCALE and the calculations are stopped. The maximum uranium density is about 1296 g/L for 3% enriched uranium. The uranium density is dependent on the enrichment. Values calculated and reported above 1296 g/L are higher than those for a pure solution. Values above 1296 g/L were calculated using volume fractions in the CSAS1X calculations. The volume fraction for uranyl nitrate ($\text{UO}_2(\text{NO}_3)_2$) was determined as the selected $\text{UO}_2(\text{NO}_3)_2$ density divided by the theoretical density of $\text{UO}_2(\text{NO}_3)_2$. The volume fraction for the water was determined as 1.5644 minus the volume fraction of $\text{UO}_2(\text{NO}_3)_2$, where 1.5644 is the sum of the density multipliers at the point where the solution reaches the fully hydrated salt [i.e., $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$]. It was assumed that this sum remained constant for some range of uranium density above this point; therefore, adding uranium reduced the amount of hydration.

The minimum critical mass was calculated as volume times density (in grams per liter). Infinite geometry CSAS1X cases with varying uranium density for the uranyl nitrate [$\text{UO}_2(\text{NO}_3)_2$] solution were performed to obtain k-infinity. Concentration (in grams per liter) is the density resulting in k-infinity equal to 1. Minimum values for uranium nitrate–water systems are shown in Table 2.

Table 1. UO₂–H₂O, water reflected (30 cm)

Enrichment 3 wt % ²³⁵ U								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g U/L)	(H/U)
Radius (cm)	26.85	23.43	Diameter (cm)	32.7	Thickness (cm)	16.7	521	47.9
Density (g U/L)	1256	2319	Density (g U/L)	2415	Density (g U/L)	2512		
Volume (L)	81.06	53.9	Linear density ^a (kg U/m)	202.816	Areal density ^b (kg U/m ²)	419.504		
Mass (kg U)	101.8	125.0						
Enrichment 4 wt % ²³⁵ U								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g U/L)	(H/U)
Radius (cm)	22.43	20.47	Diameter (cm)	28.2	Thickness (cm)	13.8	369.4	68.7
Density (g U/L)	1159	2029	Density (g U/L)	2125	Density (g U/L)	2319		
Volume (L)	47.3	35.9	Linear density ^a (kg U/m)	132.723	Areal density ^b (kg U/m ²)	320.022		
Mass (kg U)	54.8	72.8						
Enrichment 5 wt % ²³⁵ U								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g U/L)	(H/U)
Radius (cm)	20.86	18.81	Diameter (cm)	25.7	Thickness (cm)	12.2	286.6	89.5
Density (g U/L)	966	1932	Density (g U/L)	1932	Density (g U/L)	2125		
Volume (L)	38.02	27.9	Linear density ^a (kg U/m)	100.222	Areal density ^b (kg U/m ²)	259.250		
Mass (kg U)	36.7	53.9						

Table 1 (continued)

Enrichment 20 wt % ^{235}U								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g U/L)	(H/U)
Radius (cm)	16.24	13.68	Diameter (cm)	17.9	Thickness (cm)	7.2	64.7	411.5
Density (g U/L)	290	1062	Density (g U/L)	1159	Density (g U/L)	1449		
Volume (L)	17.9	10.7	Linear density ^a (kg U/m)	29.166	Areal density ^b (kg U/m ²)	104.328		
Mass (kg U)	5.2	11.4						
Enrichment 100 wt % ^{235}U								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g U/L)	(H/U)
Radius (cm)	14.82	10.08	Diameter (cm)	12.4	Thickness (cm)	3.4	12.17	2141.2
Density (g U/L)	58	9647	Density (g U/L)	10574	Density (g U/L)	9647		
Volume (L)	13.6	4.3	Linear density ^a (kg U/m)	127.694	Areal density ^b (kg U/m ²)	327.998		
Mass (kg U)	0.79	41.5						

^aLinear density (cylinders): density (g U/L) * $(\pi/4)(d^2) * 10^{-4}$ = kg U/m, d = diameter.

^bAreal density (slab): density (g U/L) * t * 10^{-2} = kg U/m², t = thickness.

Table 2. UNH–H₂O, water reflected (30 cm)

Enrichment 3 wt % ²³⁵ U								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g U/L)	(H/U)
Radius (cm)	46.86	45.9	Diameter (cm)	66.96	Thickness (cm)	38.72	624.6	34.0
Density (g U/L)	1140	1295	Density (g U/L)	1297	Density (g U/L)	1300		
Volume (L)	431.1	405	Linear density ^a (kg U/m)	456.731	Areal density ^b (kg U/m ²)	503.360		
Mass (kg U)	491	524						
Enrichment 4 wt % ²³⁵ U								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g U/L)	(H/U)
Radius (cm)	33.95	32.69	Diameter (cm)	46.89	Thickness (cm)	25.77	415.5	55.3
Density (g U/L)	910	1145	Density (g U/L)	1155	Density (g U/L)	1180		
Volume (L)	163.95	146.3	Linear density ^a (kg U/m)	199.449	Areal density ^b (kg U/m ²)	304.086		
Mass (kg U)	149.1	167.5						
Enrichment 5 wt % ²³⁵ U								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g U/L)	(H/U)
Radius (cm)	28.84	27.39	Diameter (cm)	38.79	Thickness (cm)	20.57	311.5	76.6
Density (g U/L)	765	1050	Density (g U/L)	1055	Density (g U/L)	1090		
Volume (L)	100.433	86.1	Linear density ^a (kg U/m)	124.676	Areal density ^b (kg U/m ²)	224.213		
Mass (kg U)	76.8	90.4						

Table 2 (continued)**Enrichment 20 wt % ^{235}U**

Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g U/L)	(H/U)
Radius (cm)	18.57	15.72	Diameter (cm)	21.04	Thickness (cm)	9.24	65.0	398.2
Density (g U/L)	225	600	Density (g U/L)	620	Density (g U/L)	710		
Volume (L)	26.816	16.3	Linear density ^a (kg U/m)	21.556	Areal density ^b (kg U/m ²)	65.604		
Mass (kg U)	6.0	9.8						

Enrichment 100 wt % ^{235}U								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g U/L)	(H/U)
Radius (cm)	15.20	11.74	Diameter (cm)	15.04	Thickness (cm)	5.44	12.25	2,127.2
Density (g U/L)	55	350	Density (g U/L)	375	Density (g U/L)	500		
Volume (L)	14.698	6.8	Linear density ^a (kg U/m)	6.662	Areal density ^b (kg U/m ²)	27.200		
Mass (kg U)	0.808	2.4						

^aLinear density (cylinders): density (g U/L) * $(\pi/4)(d^2) * 10^{-4}$ = kg U/m, d = diameter.

^bAreal density (slab): density (g U/L) * t * 10^{-2} = kg U/m², t = thickness.

4.3. PLUTONIUM OXIDE-WATER MIXTURES

The plutonium isotopic compositions include ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu . The following plutonium isotopic weight percent compositions have been considered in this study: Pu(100/0/0/0), Pu(95/5/0/0), Pu(90/5/5/0), and Pu(80/10/10/0), plus the more realistic vector Pu(71/17/11/1).

In all calculations the composition of the homogeneous plutonium oxide–water mixture was calculated using available options in CSAS1X. The default density value of 11.86 g/cm^3 was used for PuO_2 , and 0.9982 g/cm^3 was used for the density of H_2O . Cross sections were calculated with a temperature of 293 K.

A series of CSAS1X calculations of the critical parameters (dimensions, masses, and concentrations) with varying volume fractions of PuO_2 and H_2O were performed for spheres, cylinders, slabs, and infinite media. Plots of these calculations are shown in Figs. C.1–C.20 (Appendix C), where “enrichment” refers to the ^{239}Pu weight percent of the plutonium. The minimum critical mass is calculated as density times volume, where density is calculated as $[(N_{239} * 239.0526) + (N_{240} * 240.0542) + (N_{241} * 241.0487) + (N_{242} * 242.0584)] / 0.60221368$. Atomic weights are from the SCALE Standard Composition Library based on ENDF/B-V data.

Infinite geometry CSAS1X cases with varying volume fractions for the PuO_2 and H_2O were run to obtain k-infinity. Concentrations can be calculated from this data. The minimum critical concentration was calculated as $\text{Pu}/\text{PuO}_2 * \text{VF}$, where VF is the PuO_2 volume fraction of the CSAS1X case where k-infinity equals 1. The value of Pu/PuO_2 is calculated as $[(N_{239} * 239.0526) + (N_{240} * 240.0542) + (N_{241} * 241.0487) + (N_{242} * 242.0584)] / [(N_{239} * 239.0526) + (N_{240} * 240.0542) + (N_{241} * 241.0487) + (N_{242} * 242.0584) + 31.9812]$. Minimum values for plutonium oxide–water systems are shown in Table 3.

Table 3. PuO₂–H₂O, water reflected (30 cm)

Pu(100,0,0,0)O ₂								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g Pu/L)	(H/Pu)
Radius (cm)	15.76	6.49	Diameter (cm)	7.667	Thickness (cm)	1.723	7.091	3691
Density (g Pu/L)	30.32	10107	Density (g Pu/L)	10107	Density (g Pu/L)	10107		
Volume (L)	16.389	1.144	Linear density ^a (kg Pu/m)	46.661	Areal density ^b (kg Pu/m ²)	174.143		
Mass (kg Pu)	0.497	11562						
Pu(95,5,5,0)O ₂								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g Pu/L)	(H/Pu)
Radius (cm)	16.80	6.64	Diameter (cm)	7.926	Thickness (cm)	1.927	7.640	3426
Density (g Pu/L)	30.32	10108	Density (g Pu/L)	10108	Density (g Pu/L)	10108		
Volume (L)	19.854	1.227	Linear density ^a (kg Pu/m)	49.872	Areal density ^b (kg Pu/m ²)	194.781		
Mass (kg Pu)	0.602	12404						
Pu(90,5,5,0)O ₂								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g Pu/L)	(H/Pu)
Radius (cm)	16.56	6.62	Diameter (cm)	7.871	Thickness (cm)	1.845	7.470	3505
Density (g Pu/L)	30.31	10108	Density (g Pu/L)	10108	Density (g Pu/L)	10108		
Volume (L)	19.039	1.217	Linear density ^a (kg Pu/m)	49.183	Areal density ^b (kg Pu/m ²)	186.492		
Mass (kg Pu)	0.577	12301						

Table 3 (continued)

Pu(80,10,10,0)O₂								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g Pu/L)	(H/Pu)
Radius (cm)	17.37	6.73	Diameter (cm)	8.022	Thickness (cm)	1.908	7.889	3320
Density (g Pu/L)	24.24	10109	Density (g Pu/L)	10109	Density (g Pu/L)	10109		
Volume (L)	21.945	1.279	Linear density ^a (kg Pu/m)	51.093	Areal density ^b (kg Pu/m ²)	129.879		
Mass (kg Pu)	0.665	12929						
Pu(71,17,11,1)O₂								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g Pu/L)	(H/Pu)
Radius (cm)	18.08	6.94	Diameter (cm)	8.335	Thickness (cm)	2.096	8.968	2922
Density (g Pu/L)	35.40	10109	Density (g Pu/L)	10109	Density (g Pu/L)	10109		
Volume (L)	24.743	1.400	Linear density ^a (kg Pu/m)	55.158	Areal density ^b (kg Pu/m ²)	211.884		
Mass (kg Pu)	0.876	14152						

^aLinear density (cylinders): density(g Pu/L) * $(\pi/4)(d^2) * 10^{-4}$ = kg Pu/m, d = diameter.

^bAreal density (slab): density (g Pu/L)* t * 10^{-2} = kg Pu/m², t = thickness.

4.4. PLUTONIUM NITRATE-WATER SOLUTIONS

The plutonium isotopic compositions involve $^{239}\text{Pu}/^{240}\text{Pu}/^{241}\text{Pu}/^{242}\text{Pu}$. The following plutonium isotopic weight percents have been considered in this study: Pu(100/0/0/0), Pu(95/5/0/0), Pu(90/5/5/0), and Pu(80/10/10/0), plus the more realistic vector Pu(71/17/11/1).

The composition of the homogeneous plutonium nitrate–water material was calculated using the built-in SCALE atomic number density equations for plutonium nitrate [$(\text{Pu}(\text{NO}_3)_4)$] solution per ARH-600 (ref. 6). Cross sections were calculated with a temperature of 293 K.

A series of CSAS1X calculations of the critical parameters (dimensions, masses, and concentrations) with varying solution fuel density were performed for spheres, cylinders, slabs, and infinite media. Plots of these calculations are shown in Figs. D.1–D.20 (Appendix D), where “enrichment” refers to the ^{239}Pu weight percent of the plutonium. The minimum critical mass values for spheres were calculated as volume times density. Infinite CSAS1X cases with varying plutonium density for the plutonium nitrate [$(\text{Pu}(\text{NO}_3)_4)$] solution were performed to obtain k-infinity. Concentration (in grams per liter) is the density resulting in k-infinity equal to 1. Minimum values for plutonium nitrate–water systems are shown in Table 4.

Table 4. PuNH–H₂O, water reflected (30 cm)

Pu(100,0,0,0)(NO₃)₄								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g Pu/L)	(H/Pu)
Radius (cm)	15.95	12.18	Diameter (cm)	15.63	Thickness (cm)	5.68	7.21	3666
Density (g Pu/L)	30	290	Density (g Pu/L)	340	Density (g Pu/L)	525		
Volume (L)	16.992	7.564	Linear density ^a (kg Pu/m)	6.524	Areal density ^b (kg Pu/m ²)	29.820		
Mass (kg Pu)	0.509	2.193						
Pu(95,5,0,0)(NO₃)₄								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g Pu/L)	(H/Pu)
Radius (cm)	17.01	13.66	Diameter (cm)	17.88	Thickness (cm)	7.15	7.77	3403
Density (g Pu/L)	30	140	Density (g Pu/L)	150	Density (g Pu/L)	200		
Volume (L)	20.625	10.678	Linear density ^a (kg Pu/m)	3.766	Areal density ^b (kg Pu/m ²)	14.300		
Mass (kg Pu)	0.618	1.495						
Pu(90,5,5,0)(NO₃)₄								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g Pu/L)	(H/Pu)
Radius (cm)	16.41	13.47	Diameter (cm)	17.60	Thickness (cm)	6.96	7.6	3,505
Density (g Pu/L)	32	150	Density (g Pu/L)	150	Density (g Pu/L)	225		
Volume (L)	18.511	10.245	Linear density ^a (kg Pu/m)	3.649	Areal density ^b (kg Pu/m ²)	15.660		
Mass (kg Pu)	0.572	1.537						

Table 4 (continued)

Pu(80,10,10,0)(NO₃)₄								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g Pu/L)	(H/Pu)
Radius (cm)	17.59	14.22	Diameter (cm)	18.71	Thickness (cm)	7.65	8.05	3291
Density (g Pu/L)	30	140	Density (g Pu/L)	150	Density (g Pu/L)	200		
Volume (L)	22.790	12.057	Linear density ^a (kg Pu/m)	4.124	Areal density ^b (kg Pu/m ²)	15.300		
Mass (kg Pu)	0.683	1.688						
Pu(71,17,11,1)(NO₃)₄								
Sphere			Cylinder		Slab		Concentration	
	Min. mass	Min. vol.		Min. values		Min. values	(g Pu/L)	(H/Pu)
Radius (cm)	18.16	15.52	Diameter (cm)	20.65	Thickness (cm)	8.88	9.15	2897
Density (g Pu/L)	36	125	Density (g Pu/L)	135	Density (g Pu/L)	175		
Volume (L)	25.098	15.645	Linear density ^a (kg Pu/m)	4.521	Areal density ^b (kg Pu/m ²)	15.540		
Mass (kg Pu)	0.902	1.956						

^aLinear density (cylinders): density (g Pu/L) * $(\pi/4)(d^2) * 10^{-4}$ = kg Pu/m, d = diameter.

^bAreal density (slab): density (g Pu/L) * t * 10^{-2} = kg Pu/m², t = thickness.

4.5. RANGE OF DATA AND DISCREPANCIES

Tables 5 and 6 give an overview on the largest and smallest values found for every minimum critical parameter of the examined UO₂, UHN, PuO₂, and PuNH systems as reported by W. Weber and D. Mennerdahl in “Results of an OECD/NEA Comparison of Minimum Critical Values,” which was presented at The Seventh International Conference on Nuclear Criticality Safety (ICNC2003), Tokaimura, Japan, October 20–24, 2003 (ref. 1). ORNL results are shown in comparison to the Weber report. To identify discrepancies of major size, the percentage differences (Δ_{all}) between the largest and the smallest value for every critical parameter for all systems of the compilation are given.

Table 5. Spheres: largest and smallest minimum critical values and differences (Δ_{all}) for all entries provided

UO₂ systems								
Sphere mass (kg U)					Sphere volume (L)			
²³⁵ U (wt %)	Smallest, all	ORNL	Largest, all	$\Delta_{\text{all}} (\%)$	Smallest, all	ORNL	Largest, all	$\Delta_{\text{all}} (\%)$
3	88	101.8	102.8	14	45	53.9	55.8	19
4	53.9	54.8	56.9	5	32.9	35.9	37	11
5	35	38.2	38.2	8	27.4	27.9	29.1	6
20	4.95	5.2	7.43	33	9.9	10.7	11.5	14
100	0.77	0.79	0.84	8	3.9	4.3	6.1	36
UHN systems								
Sphere mass (kg U)					Sphere volume (L)			
²³⁵ U (wt %)	Smallest, all	ORNL	Largest, all	$\Delta_{\text{all}} (\%)$	Smallest, all	ORNL	Largest, all	$\Delta_{\text{all}} (\%)$
3	429	491	529	19	361	405	434	17
4	144.2	149.1	164	12	134.9	146.3	160	16
5	74	76.8	82.2	10	80	86.1	89.4	11
20	4	6.0	6.13	35	15	16.3	16.93	11
100	0.79	0.808	0.86	8	6.38	6.8	7	9
PuO₂ systems								
Sphere mass (kg Pu)					Sphere volume (L)			
Pu vector: ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu	Smallest, all	ORNL	Largest, all	$\Delta_{\text{all}} (\%)$	Smallest, all	ORNL	Largest, all	$\Delta_{\text{all}} (\%)$
100/0/0/0	0.48	0.497	0.531	10	1.00	1.144	1.22	18
95/5/0/0	0.58	0.602	0.65	10	1.23	1.227	1.31	6
90/5/5/0	0.585	0.577	—	—	1.24	1.217	—	—
80/10/10/0	0.665	0.665	0.715	7	1.28	1.279	1.36	6
71/17/11/1	0.88	0.88	0.95	8	1.40	1.4	1.5	6
PuHN systems								
Sphere mass (kg Pu)					Sphere volume (L)			
Pu vector: ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu	Smallest, all	ORNL	Largest, all	$\Delta_{\text{all}} (\%)$	Smallest, all	ORNL	Largest, all	$\Delta_{\text{all}} (\%)$
100/0/0/0	0.509	0.509	0.544	6	7.256	7.564	8.3	13
95/5/0/0	0.61	0.618	0.663	8	10.67	10.678	11.45	7
90/5/5/0	0.598	0.572	0.60	0.3	10.175	10.245	10.27	1
80/10/10/0	0.66	0.683	0.735	10	11.8	12.057	12.96	9
71/17/11/1	0.90	0.902	0.978	8	15.6	15.645	16.92	8

5. CONCLUSIONS

Basic minimum critical values are important physical constants needed for assessing safety margins in criticality and are used for licensing. The scope of the Expert Group is to compile minimum critical values from different countries. Some of the members use identical or almost identical methods and data.

The built-in SCALE atomic number density equations for uranyl nitrate (UNH) and plutonium nitrate (PuNH) were used. The maximum uranium density is about 1296 g/L. In the calculations for 3 wt % enriched uranium nitrate, the uranium density for minimum values was higher than what a real solution would support. Therefore, a great deal of confidence should not be placed in the results above the uranium density of 1296 g/L. SCALE warns the user when the uranium density is higher than what a real solution would support.

6. REFERENCES

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APPENDIX A

**GRAPHS OF UO₂–H₂O, WATER REFLECTED (30 cm):
CRITICAL PARAMETERS vs FUEL VOLUME FRACTIONS**

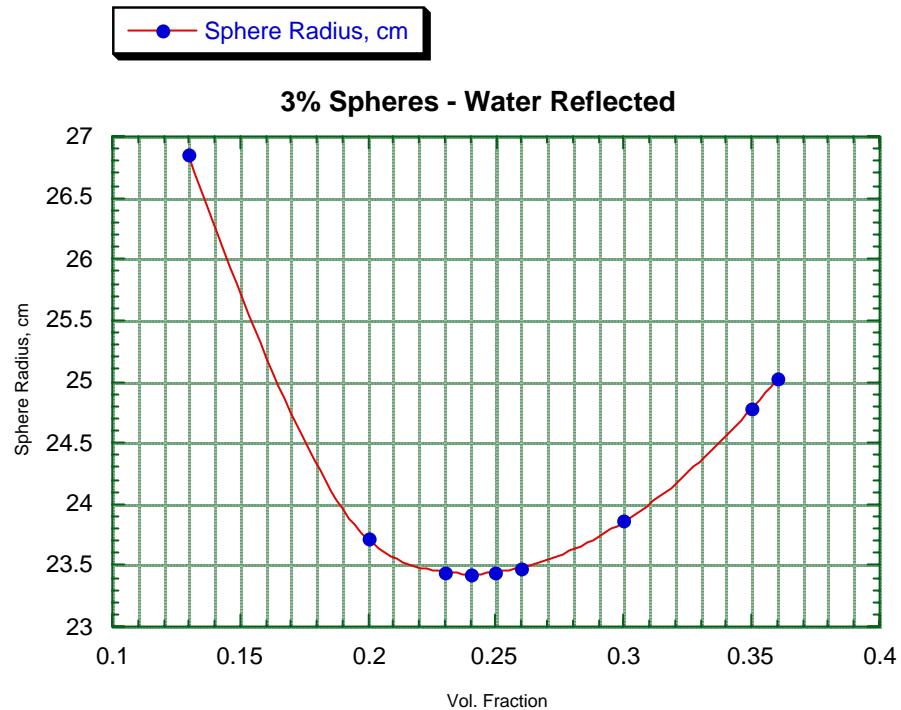


Fig. A.1. Radius vs volume fraction for 3% enriched $\text{UO}_2\text{-H}_2\text{O}$ spheres.

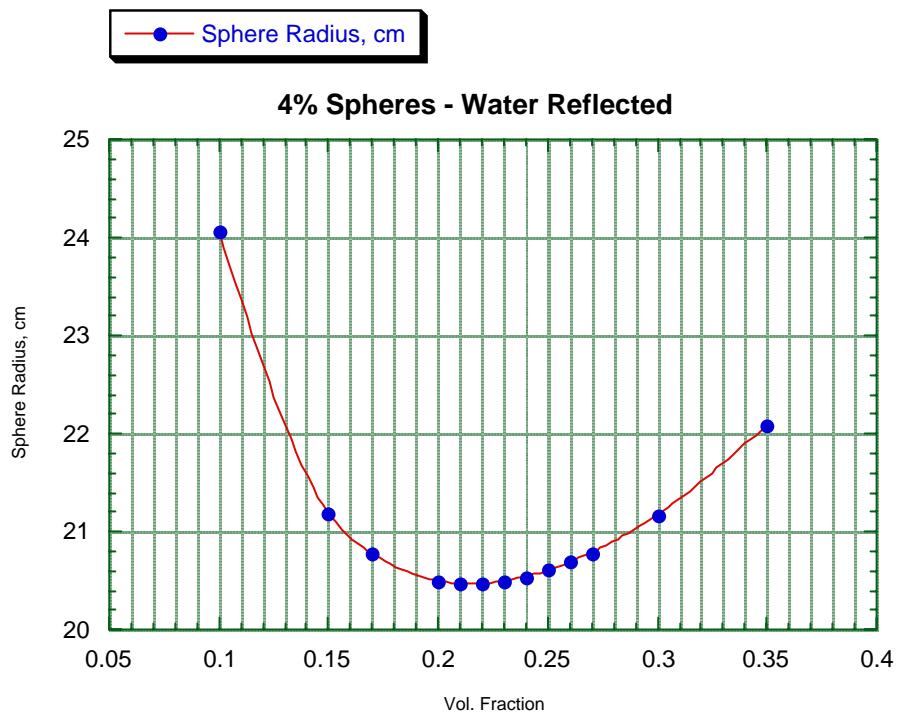


Fig. A.2. Radius vs volume fraction for 4% enriched $\text{UO}_2\text{-H}_2\text{O}$ spheres.

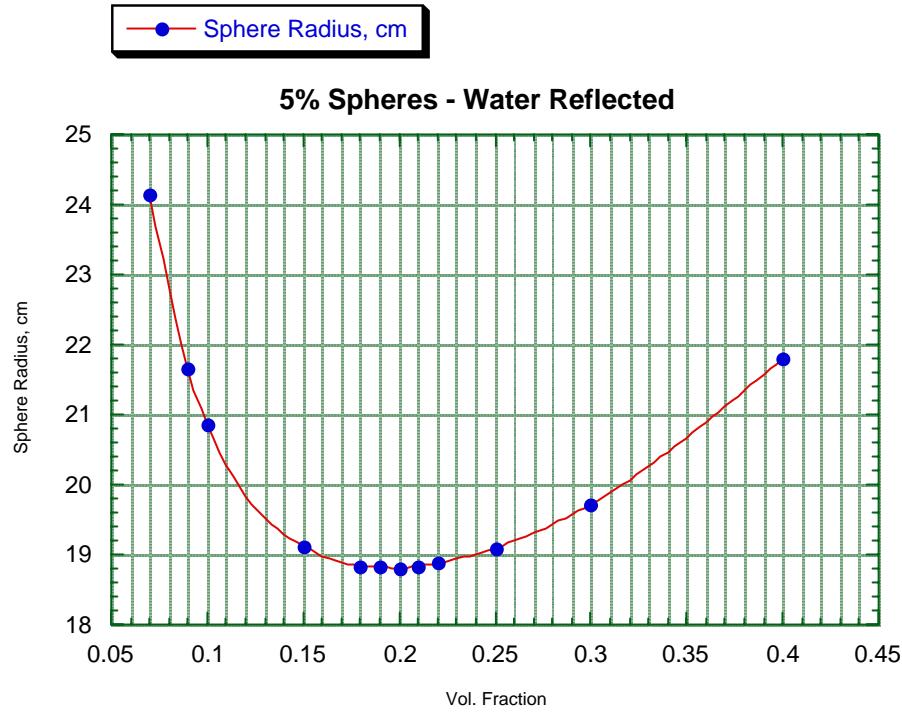


Fig. A.3. Radius vs volume fraction for 5% enriched $\text{UO}_2\text{-H}_2\text{O}$ spheres.

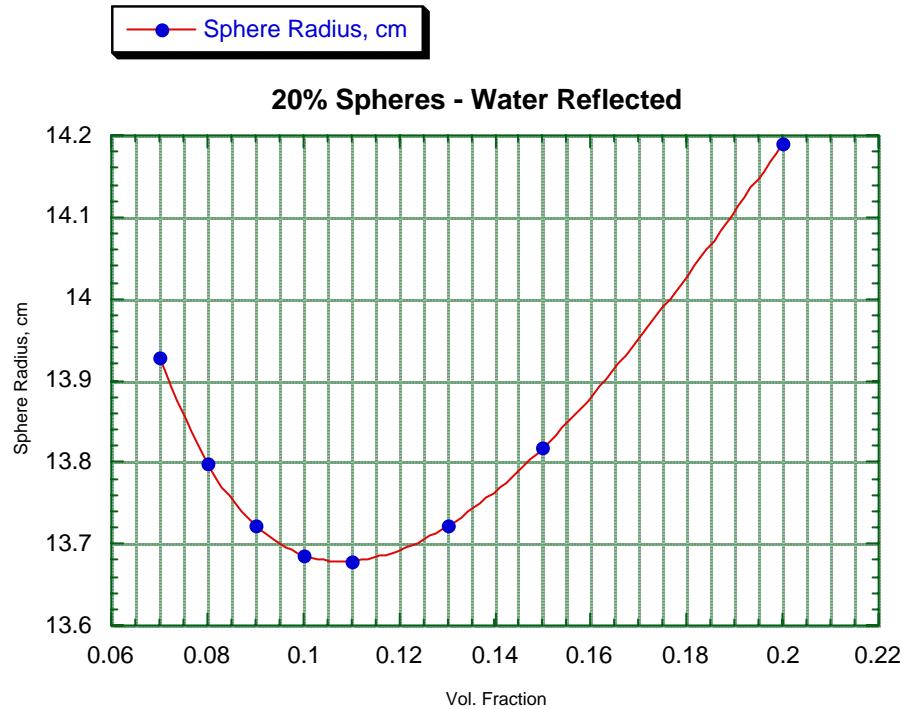


Fig. A.4. Radius vs volume fraction for 20% enriched $\text{UO}_2\text{-H}_2\text{O}$ spheres.

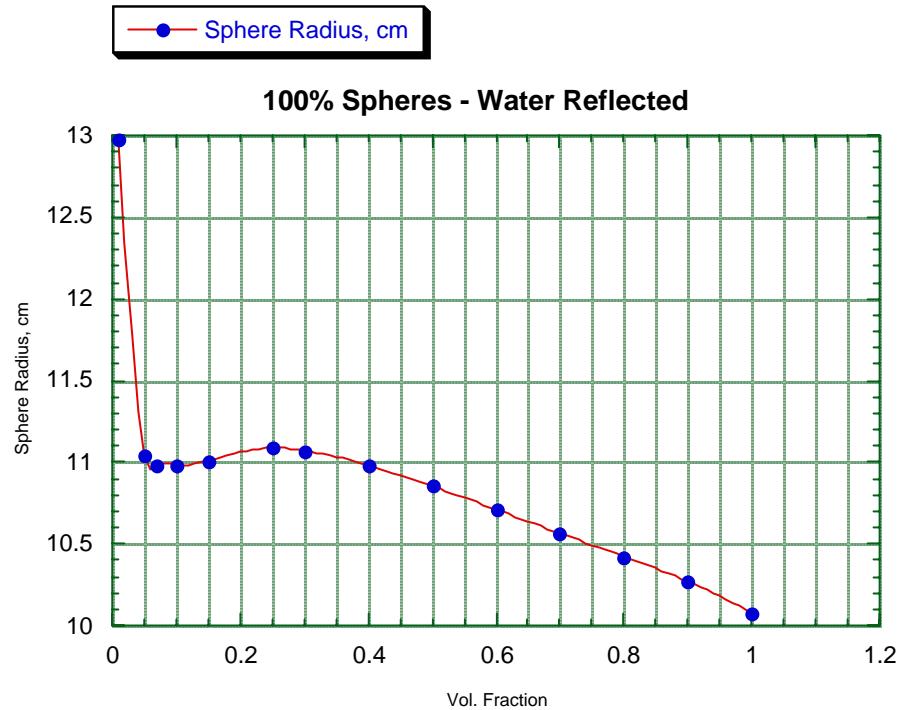


Fig. A.5. Radius vs volume fraction for 100% enriched $\text{UO}_2\text{-H}_2\text{O}$ spheres.

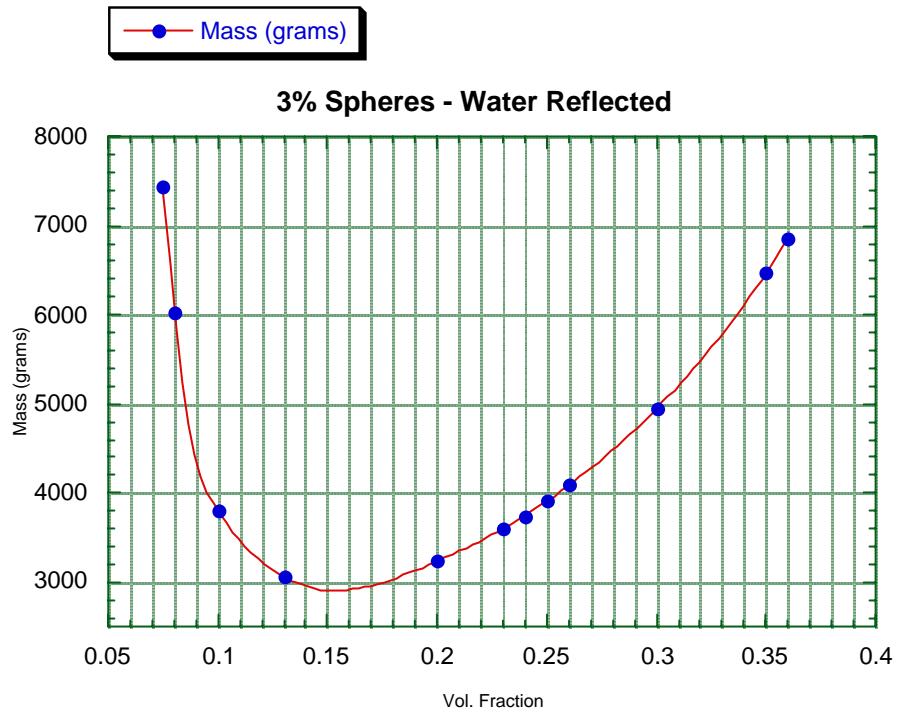


Fig. A.6. Mass vs volume fraction for 3% enriched $\text{UO}_2\text{-H}_2\text{O}$ spheres.

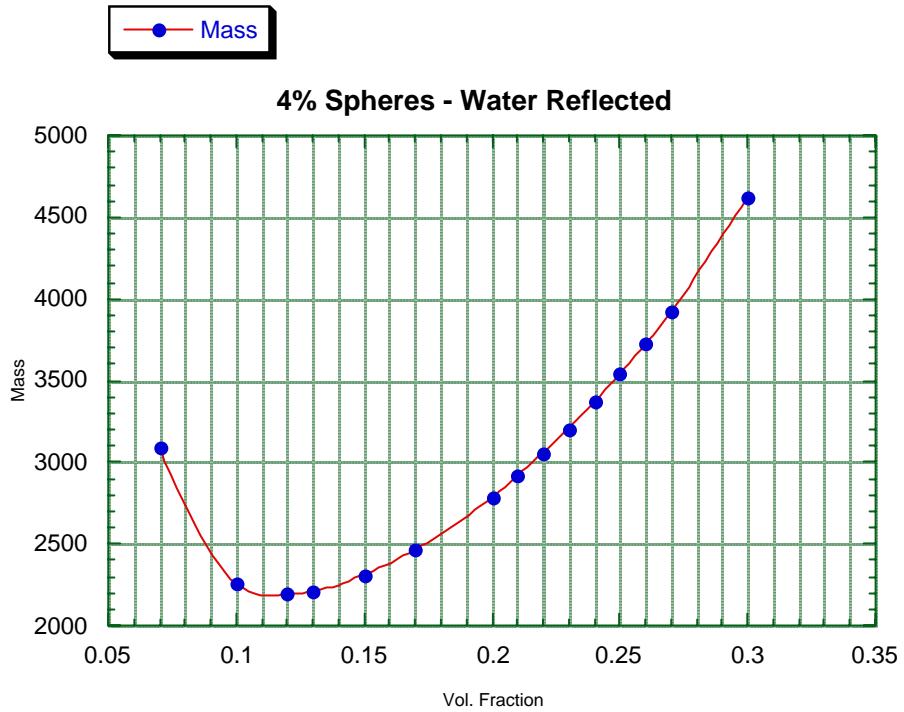


Fig. A.7. Mass vs volume fraction for 4% enriched $\text{UO}_2\text{-H}_2\text{O}$ spheres.

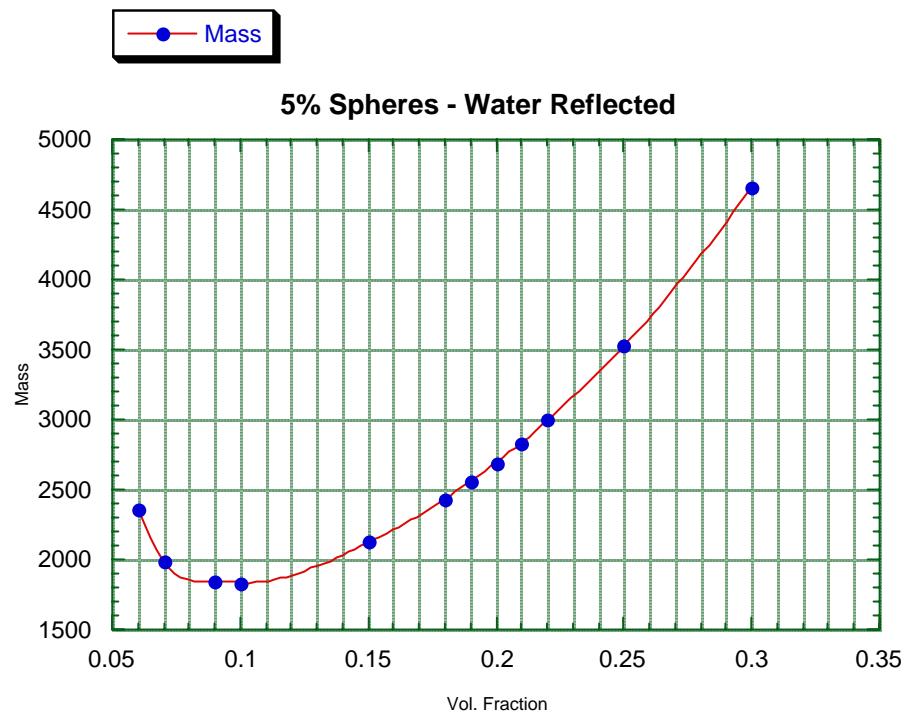


Fig. A.8. Mass vs volume fraction for 5% enriched $\text{UO}_2\text{-H}_2\text{O}$ spheres.

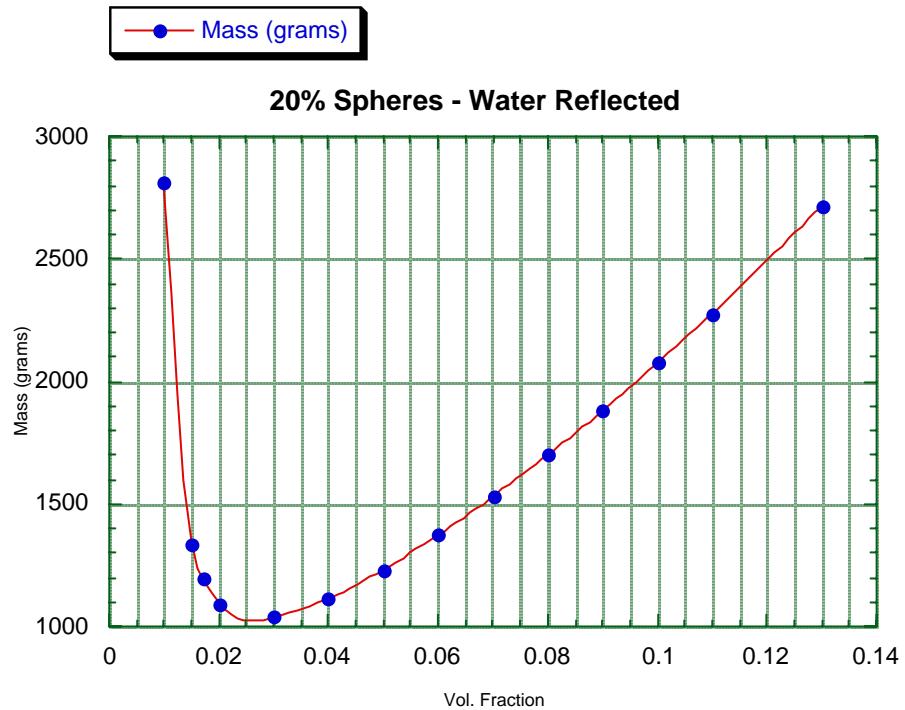


Fig. A.9. Mass vs volume fraction for 20% enriched $\text{UO}_2\text{-H}_2\text{O}$ spheres.

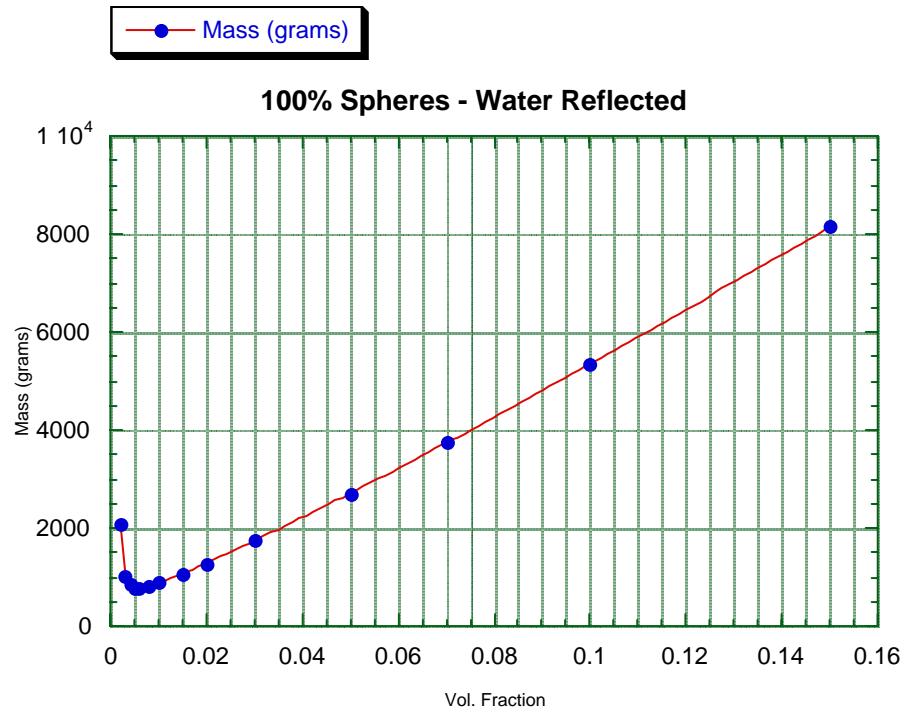


Fig. A.10. Mass vs volume fraction for 100% enriched $\text{UO}_2\text{-H}_2\text{O}$ spheres.

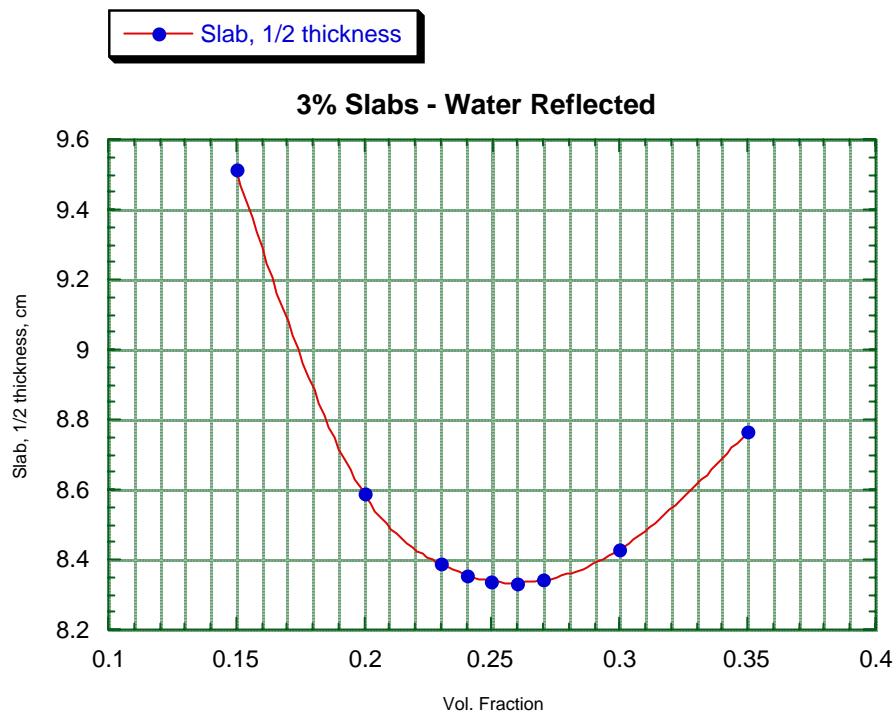


Fig. A.11. One-half thickness vs volume fraction for 3% enriched $\text{UO}_2\text{-H}_2\text{O}$ slabs.

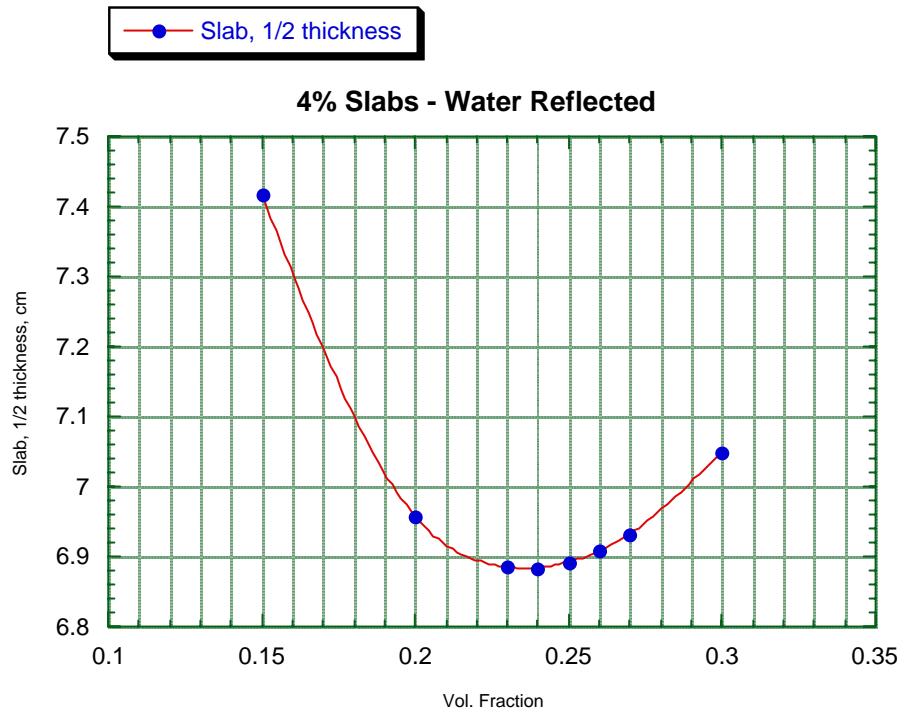


Fig. A.12. One-half thickness vs volume fraction for 4% enriched $\text{UO}_2\text{-H}_2\text{O}$ slabs.

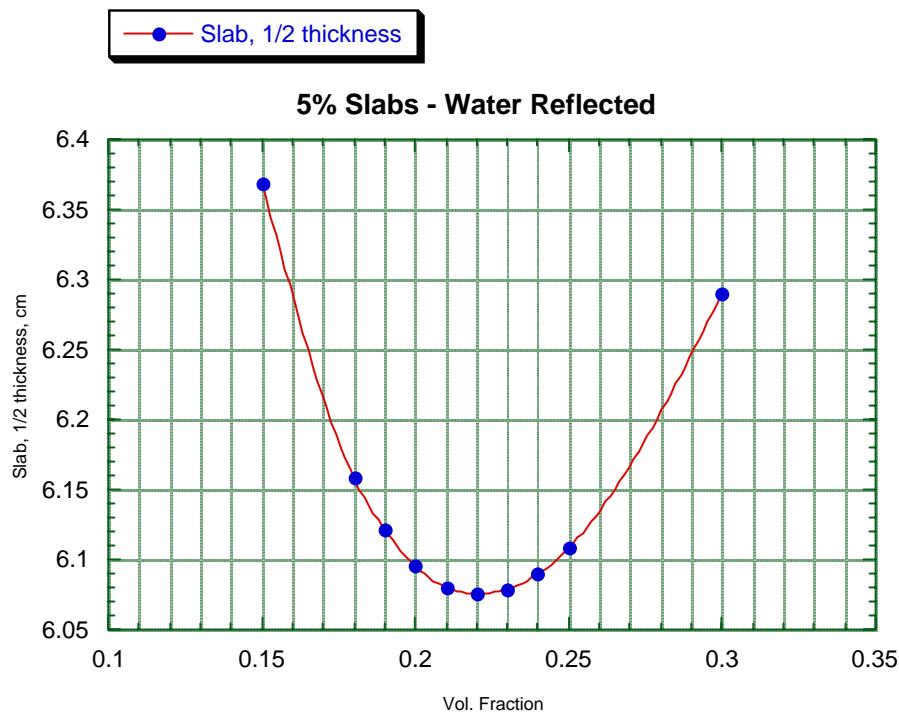


Fig. A.13. One-half thickness vs volume fraction for 5% enriched $\text{UO}_2\text{-H}_2\text{O}$ slabs.

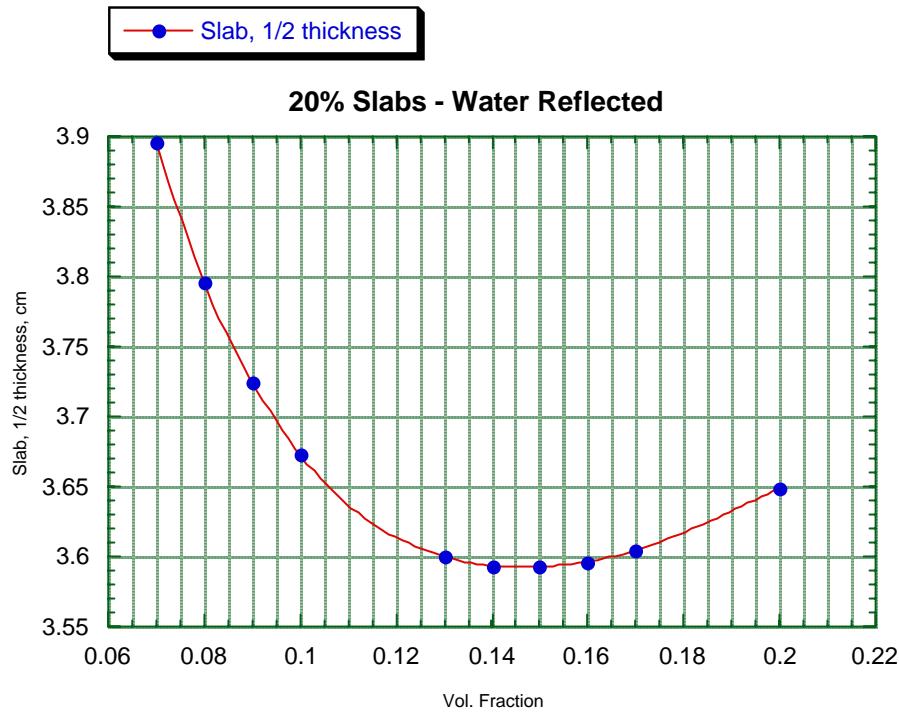


Fig. A.14. One-half thickness vs volume fraction for 20% enriched $\text{UO}_2\text{-H}_2\text{O}$ slabs.

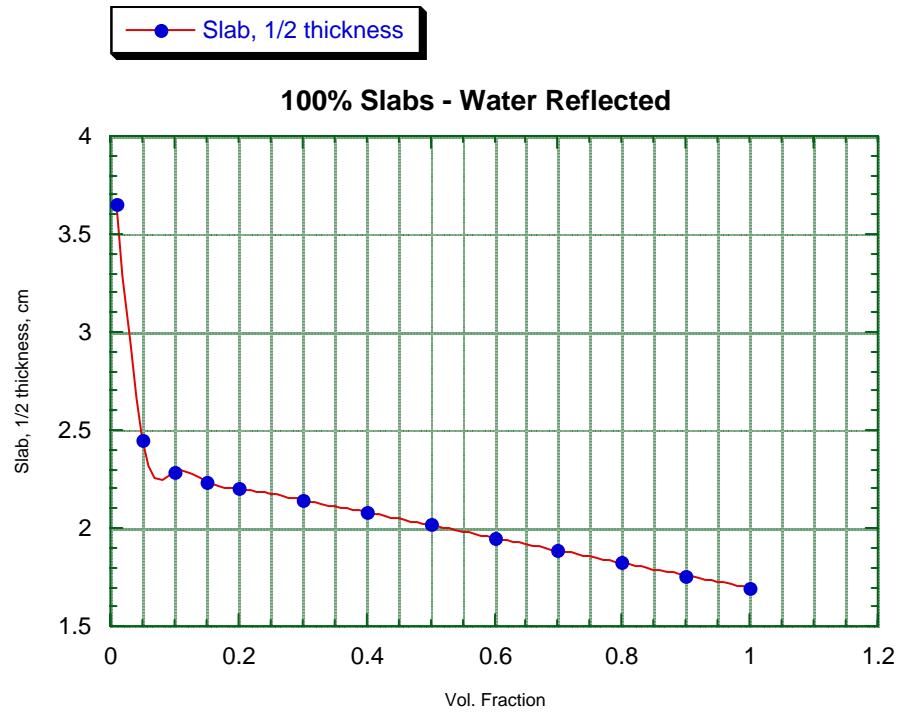


Fig. A.15. One-half thickness vs volume fraction for 100% enriched $\text{UO}_2\text{-H}_2\text{O}$ slabs.

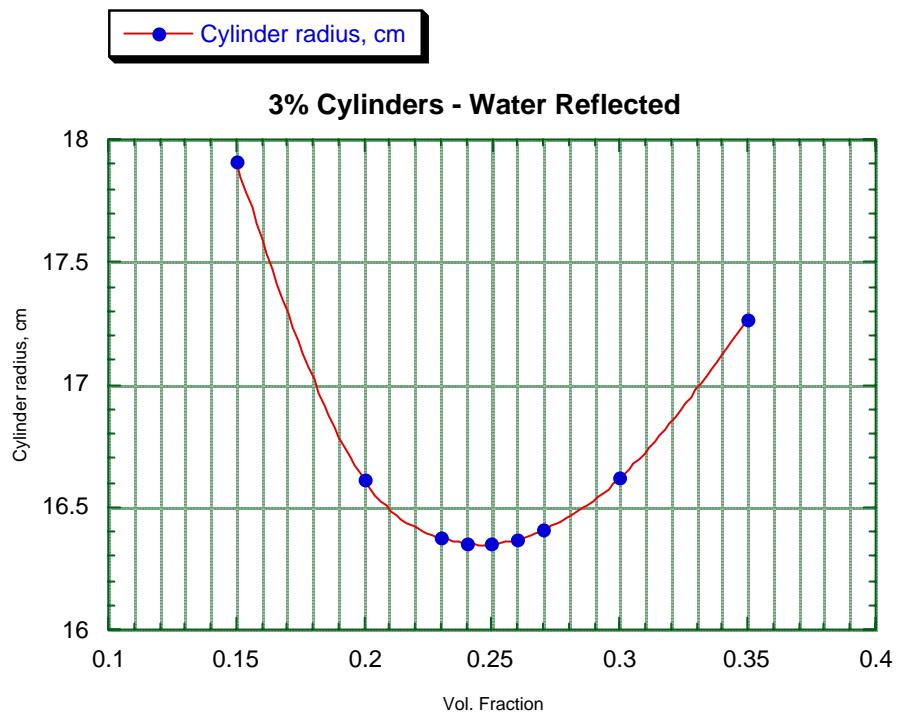


Fig. A.16. Radius vs volume fraction for 3% enriched $\text{UO}_2\text{-H}_2\text{O}$ cylinders.

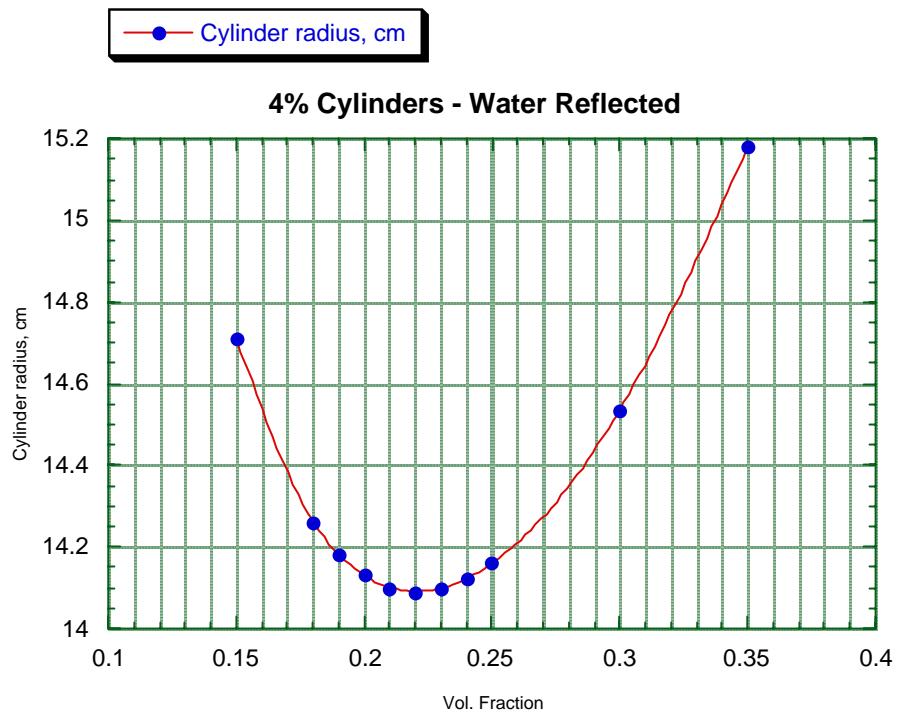


Fig. A.17. Radius vs volume fraction for 4% enriched $\text{UO}_2\text{-H}_2\text{O}$ cylinders.

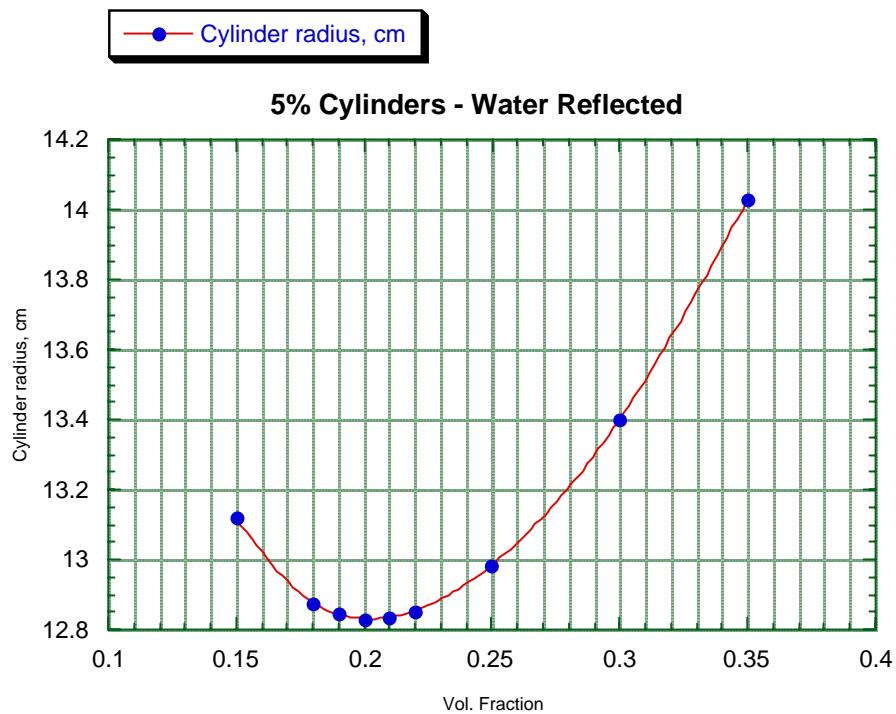


Fig. A.18. Radius vs volume fraction for 5% enriched $\text{UO}_2\text{-H}_2\text{O}$ cylinders.

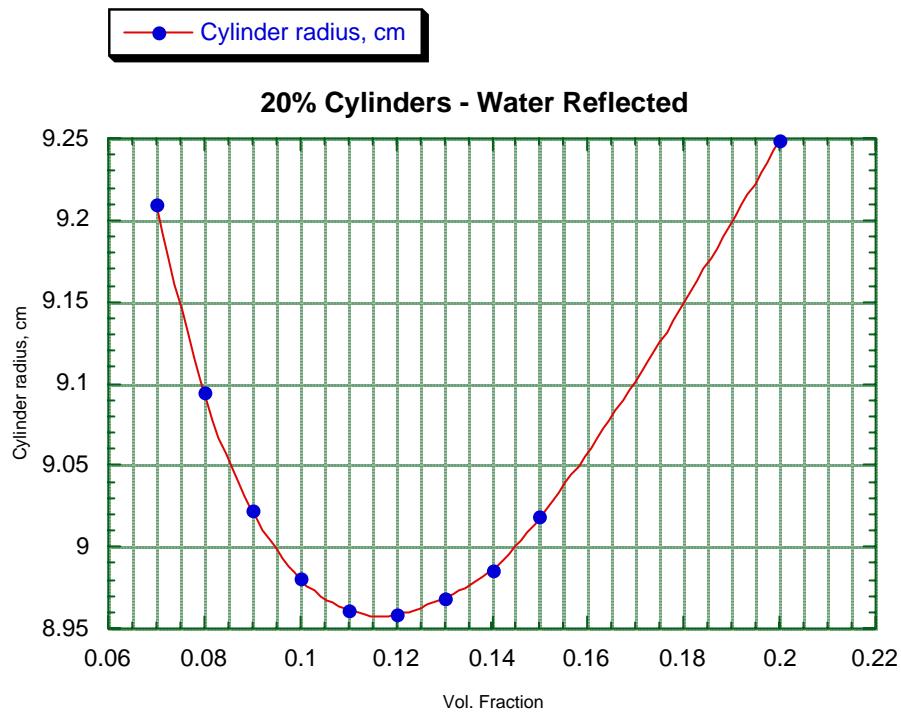


Fig. A.19. Radius vs volume fraction for 20% enriched $\text{UO}_2\text{-H}_2\text{O}$ cylinders.

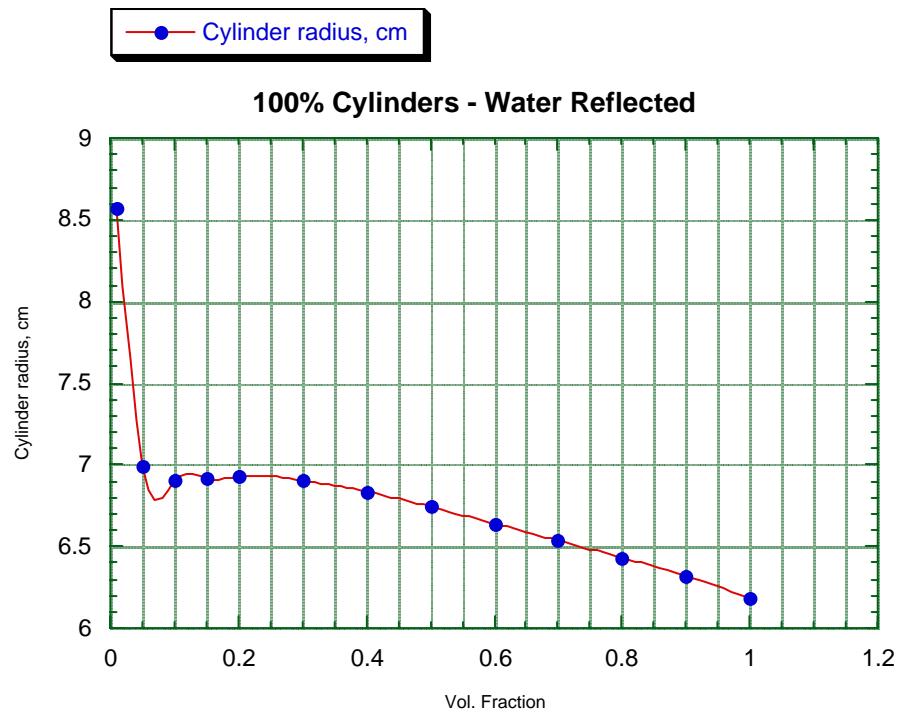


Fig. A.20. Radius vs volume fraction for 100% enriched $\text{UO}_2\text{-H}_2\text{O}$ cylinders.

APPENDIX B

**GRAPHS OF UNH-H₂O, WATER REFLECTED (30 cm):
CRITICAL PARAMETERS vs URANIUM DENSITY**

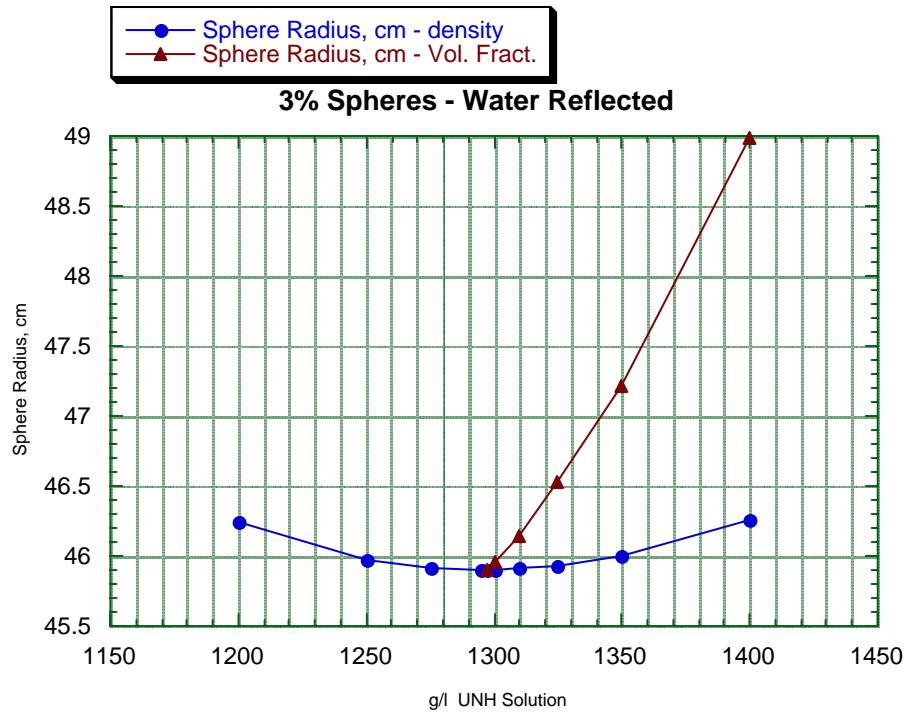


Fig. B.1. Radius vs density and volume fraction for 3% enriched UNH–H₂O spheres.

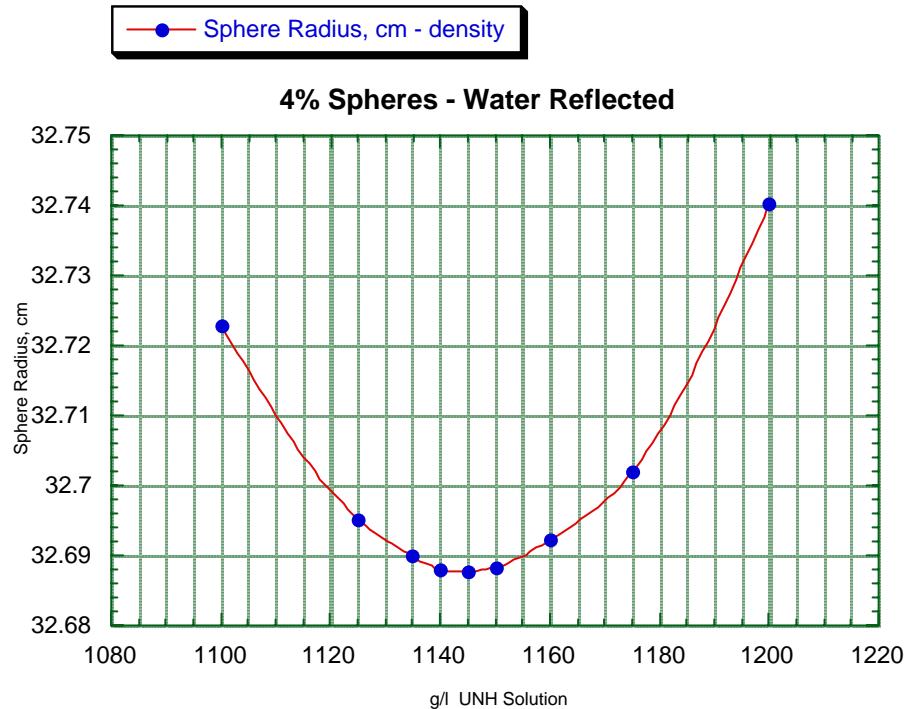


Fig. B.2. Radius vs density for 4% enriched UNH–H₂O spheres.

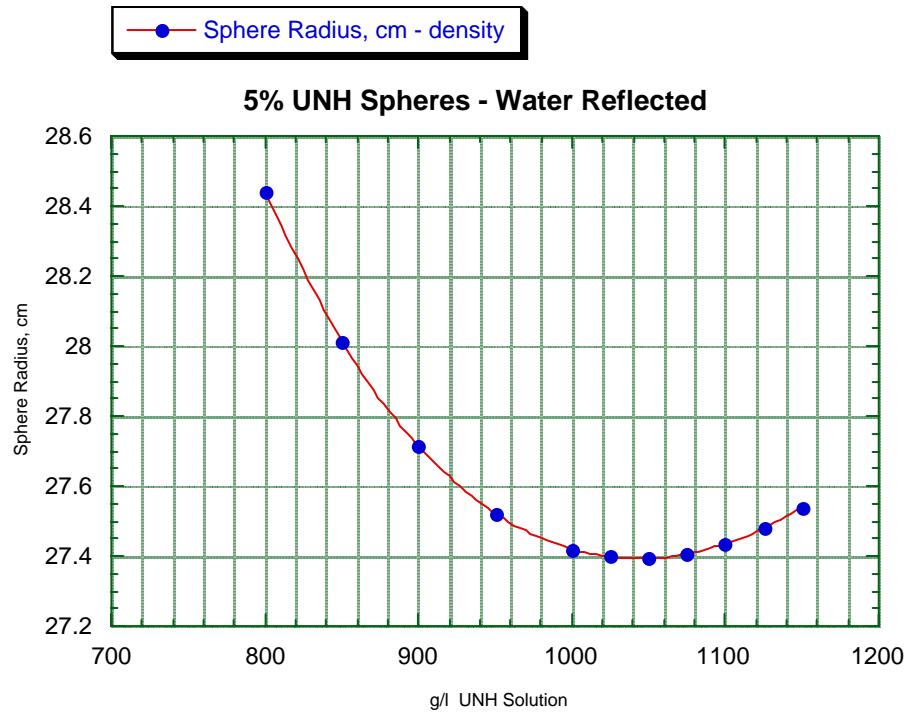


Fig. B.3. Radius vs density for 5% enriched UNH–H₂O spheres.

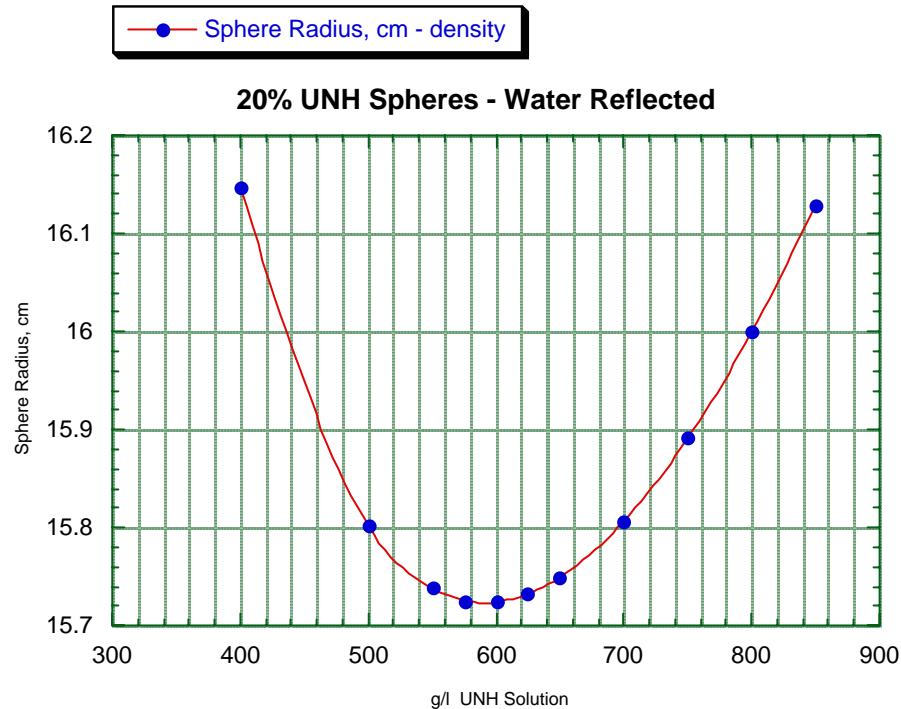


Fig. B.4. Radius vs density for 20% enriched UNH–H₂O spheres.

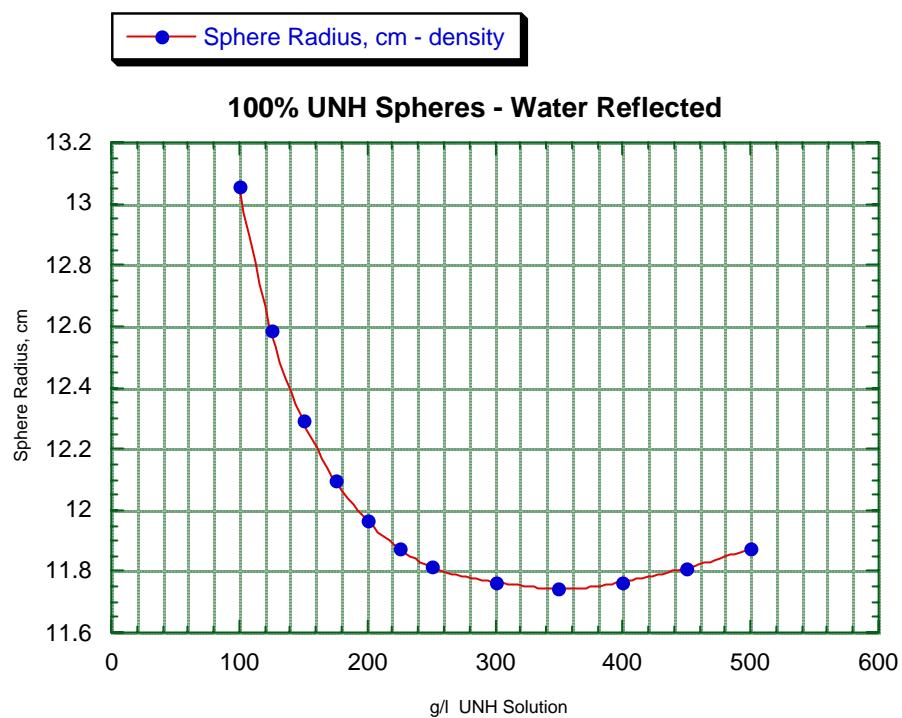


Fig. B.5. Radius vs density for 100% enriched UNH–H₂O spheres.

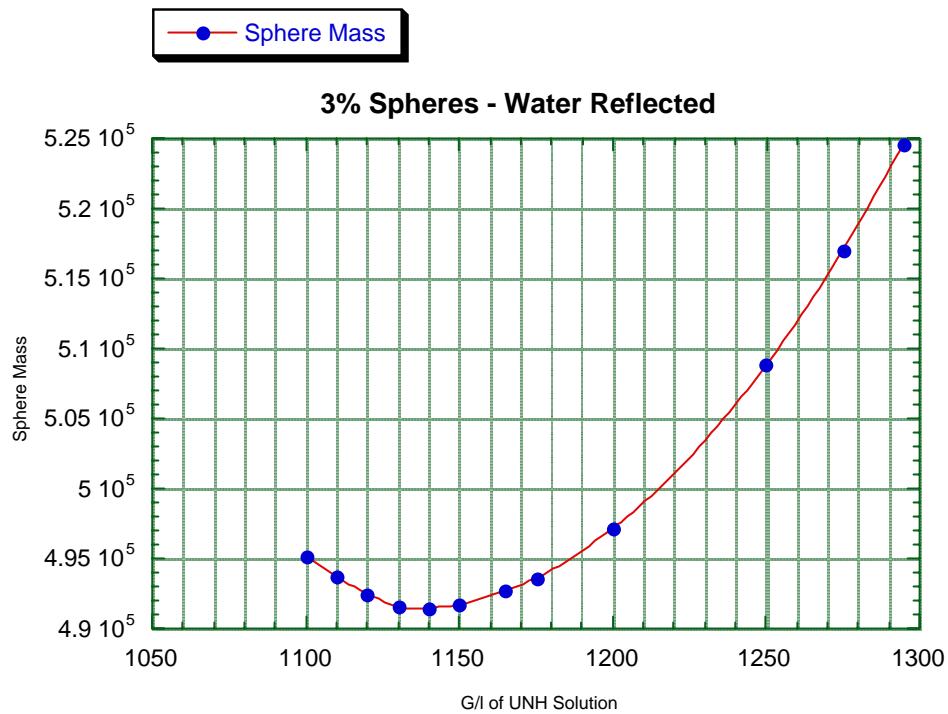


Fig. B.6. Mass vs density for 3% enriched UNH–H₂O spheres.

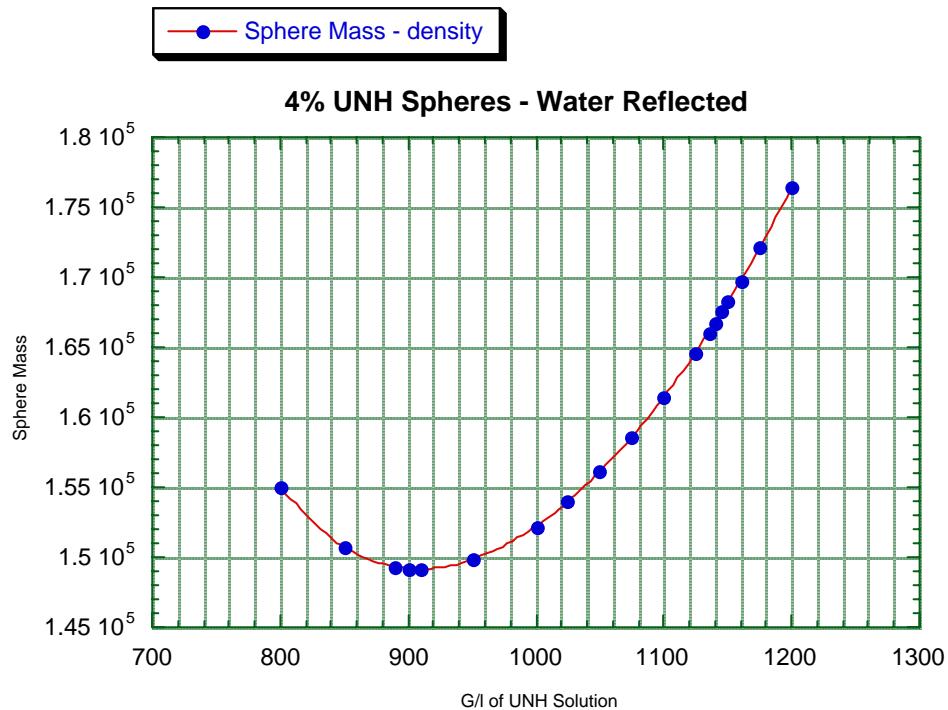


Fig. B.7. Mass vs density for 4% enriched UNH–H₂O spheres.

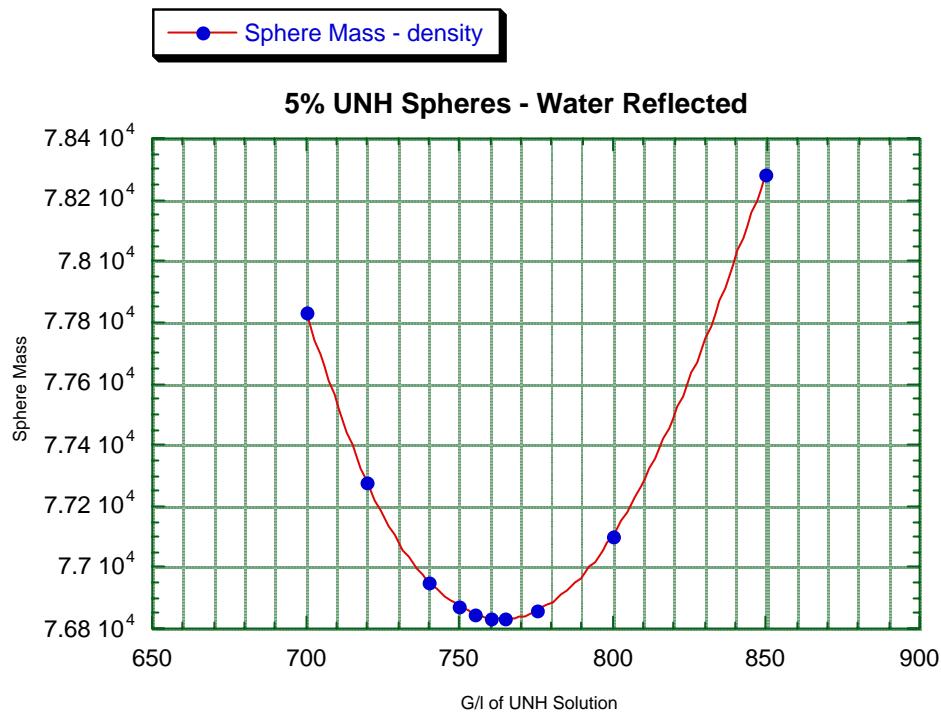


Fig. B.8. Mass vs density for 5% enriched UNH–H₂O spheres.

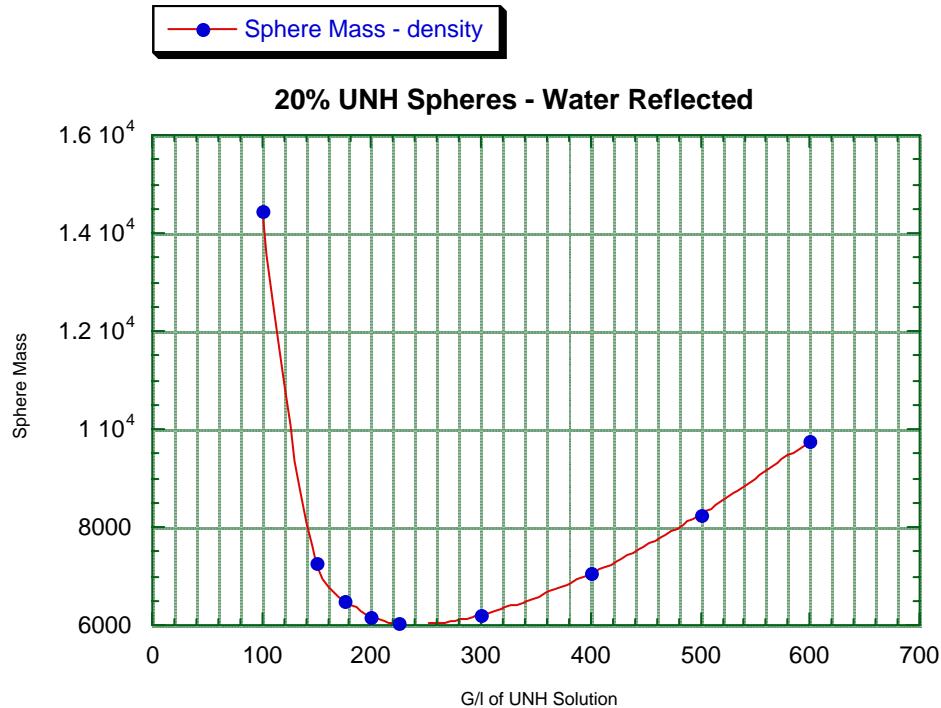


Fig. B.9. Mass vs density for 20% enriched UNH–H₂O spheres.

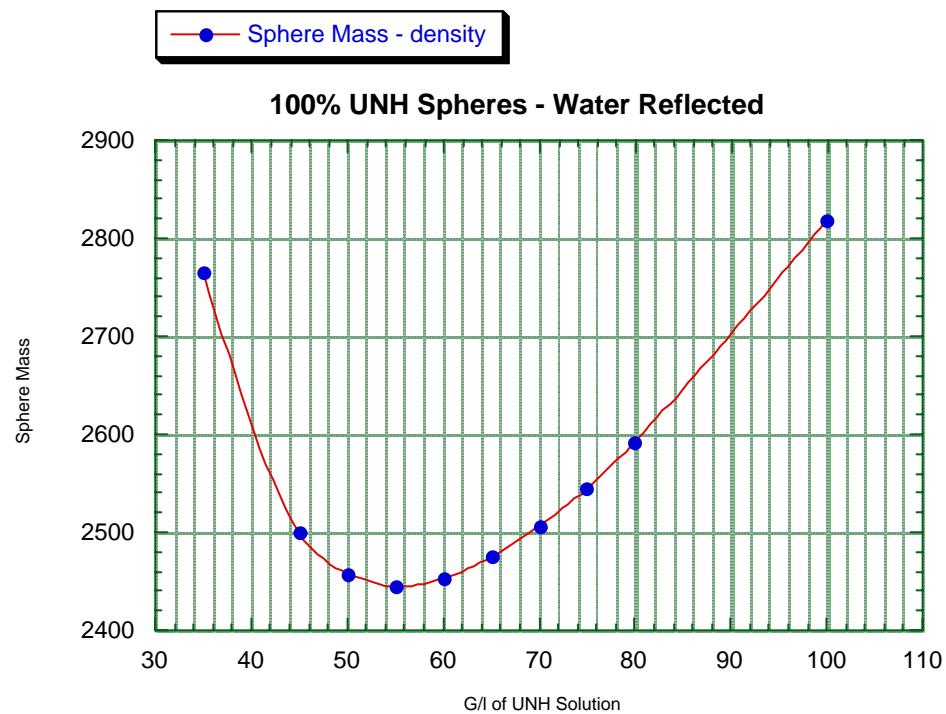


Fig. B.10. Mass vs density for 100% enriched UNH–H₂O spheres.

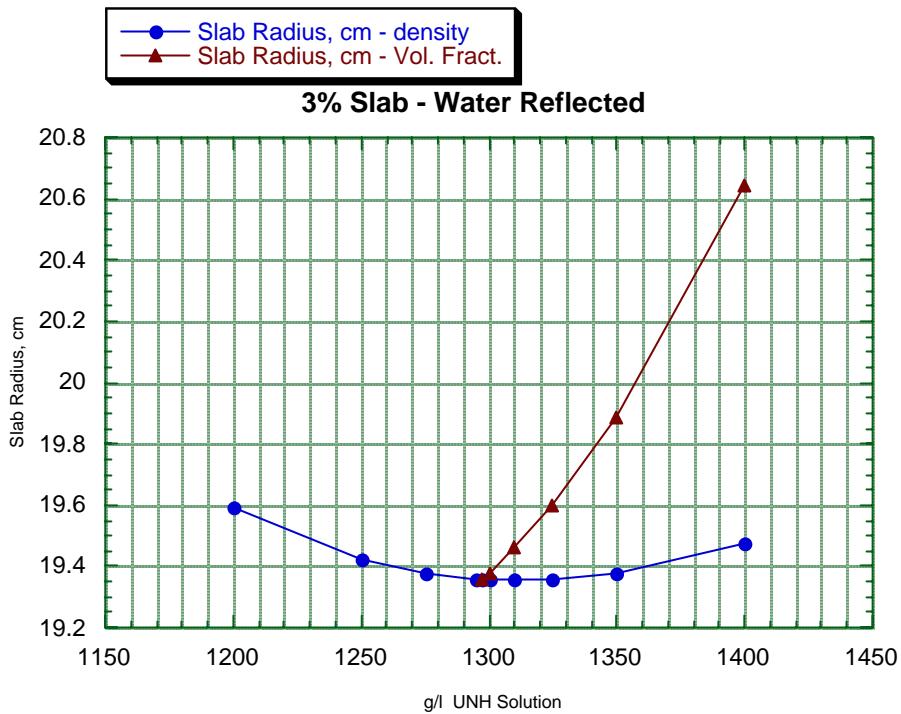


Fig. B.11. Radius vs density and volume fraction for 3% enriched UNH–H₂O slabs.

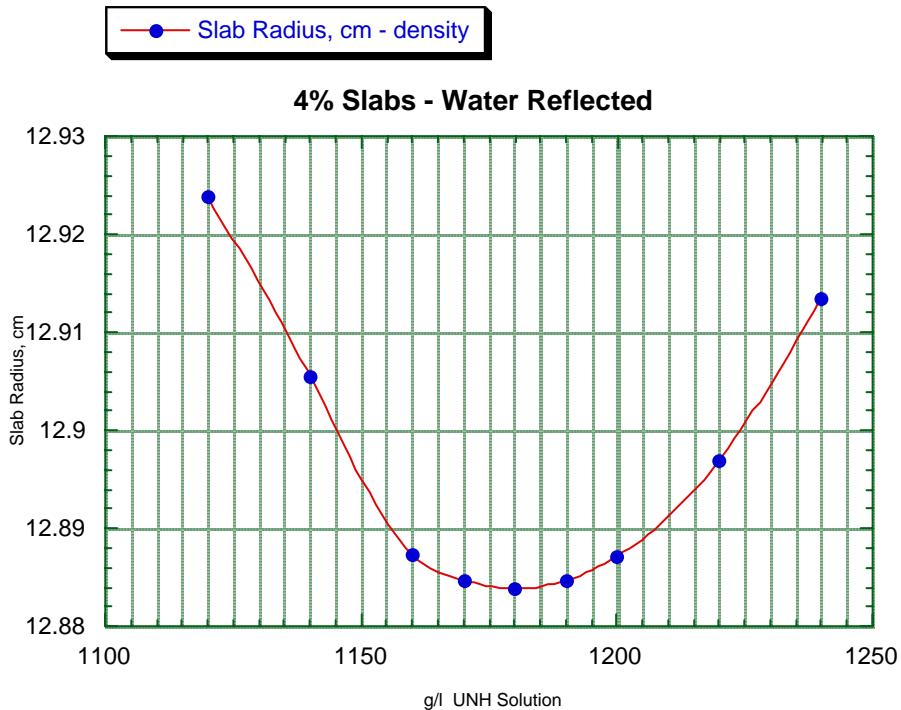


Fig. B.12. Radius vs density for 4% enriched UNH–H₂O slabs.

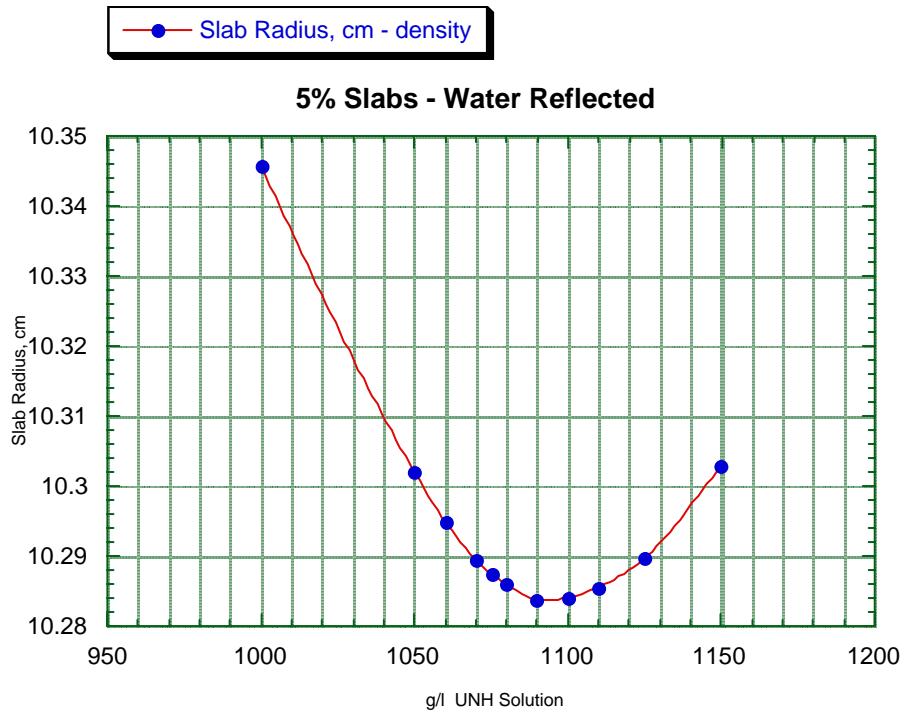


Fig. B.13. Radius vs density for 5% enriched UNH–H₂O slabs.

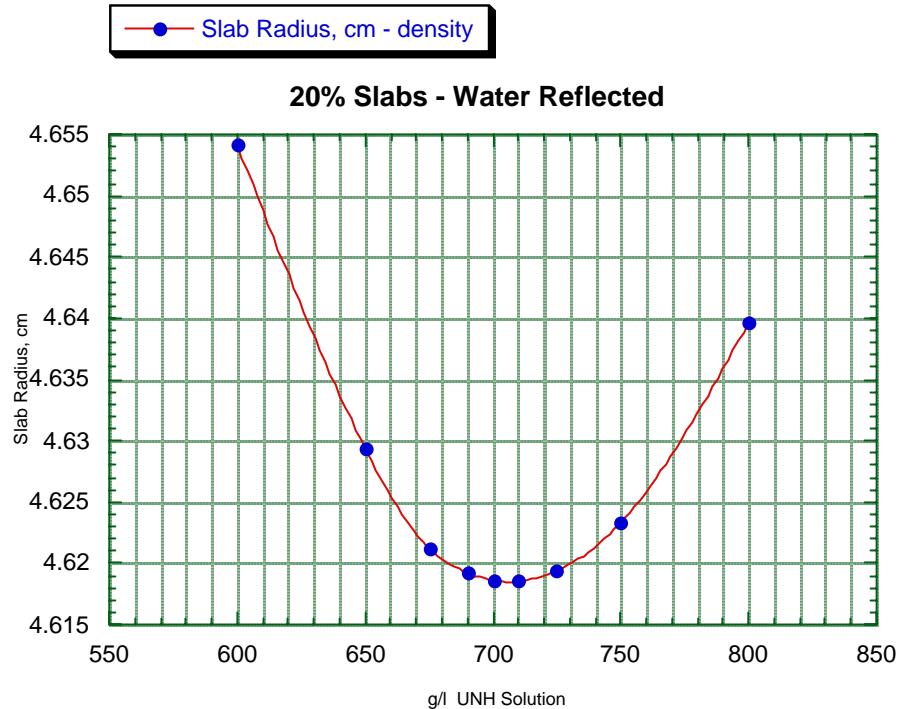


Fig. B.14. Radius vs density for 20% enriched UNH–H₂O slabs.

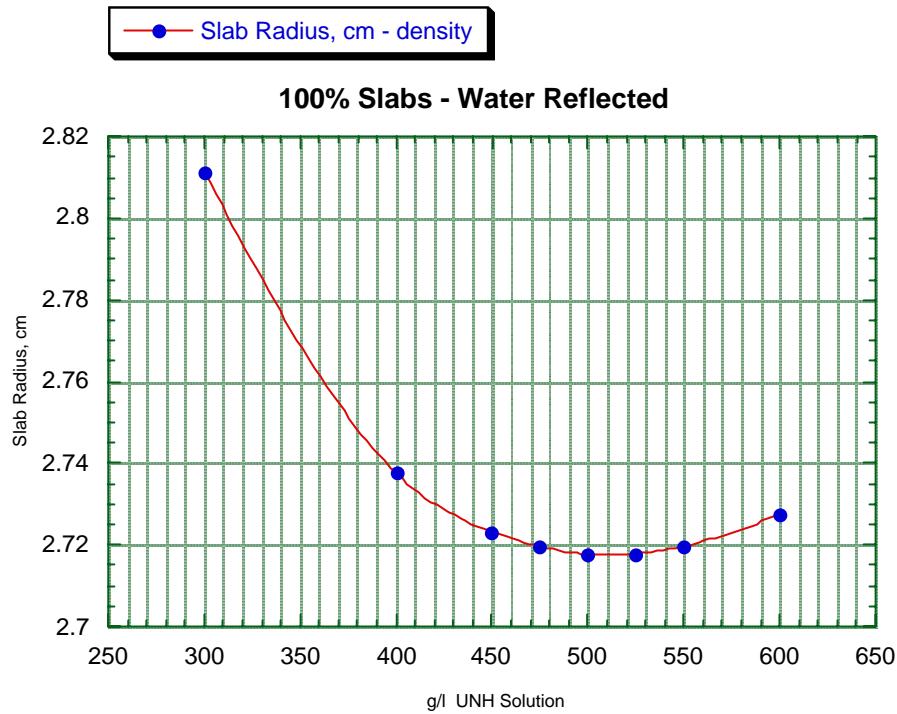


Fig. B.15. Radius vs density for 100% enriched UNH–H₂O slabs.

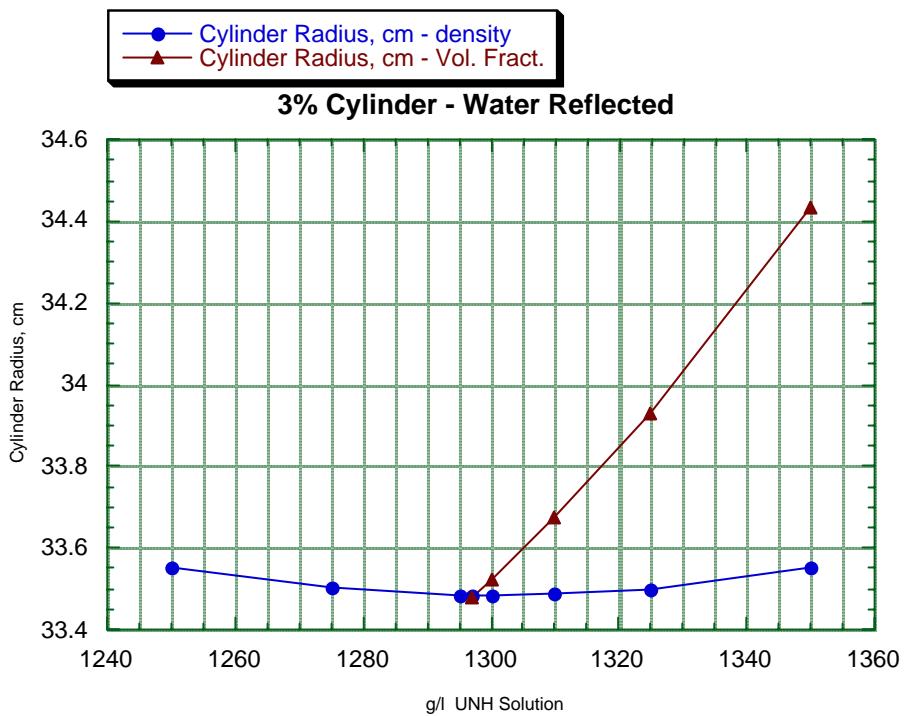


Fig. B.16. Radius vs density and volume fraction for 3% enriched UNH–H₂O cylinders.

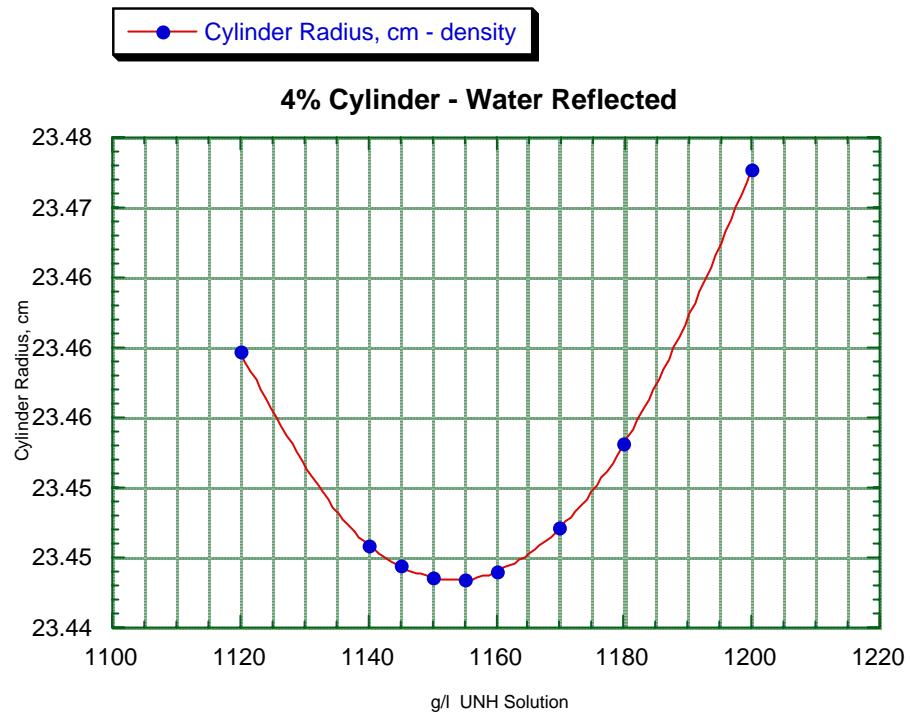


Fig. B.17. Radius vs density for 4% enriched UNH–H₂O cylinders.

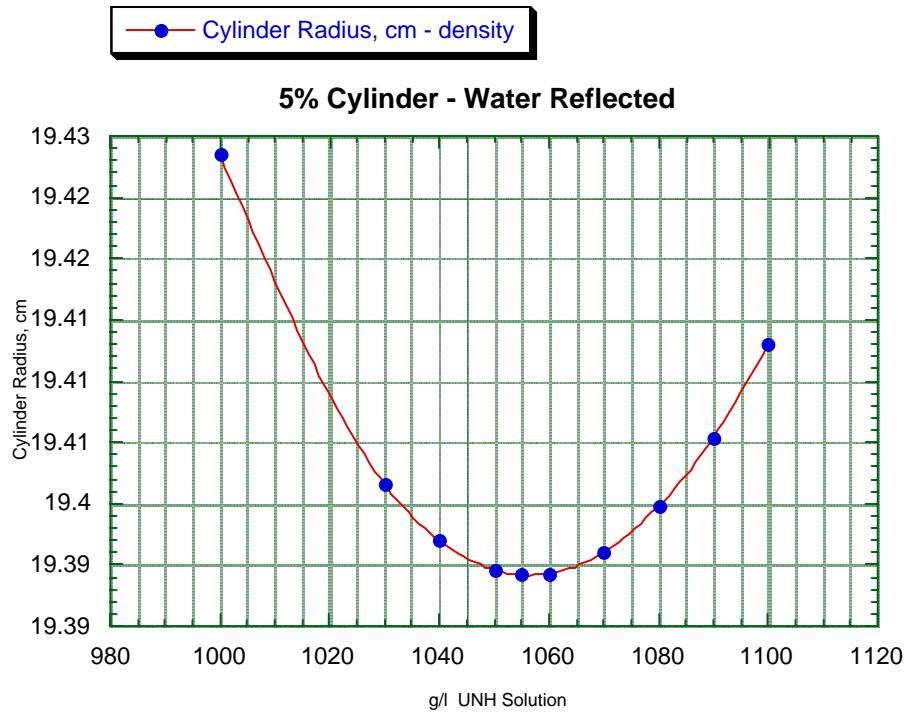


Fig. B.18. Radius vs density for 5% enriched UNH–H₂O cylinders.

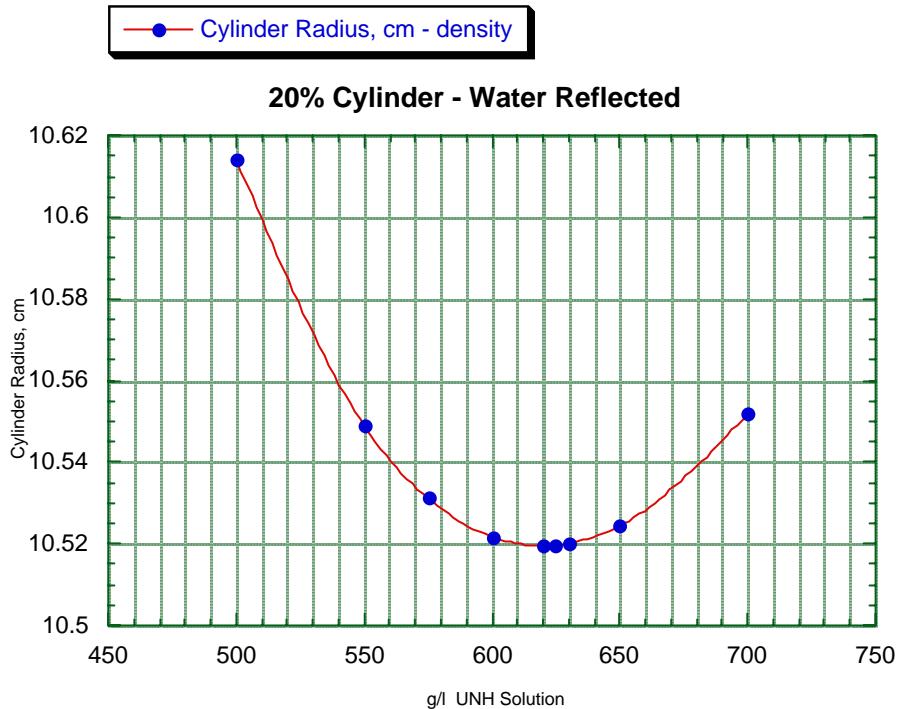


Fig. B.19. Radius vs density for 20% enriched UNH–H₂O cylinders.

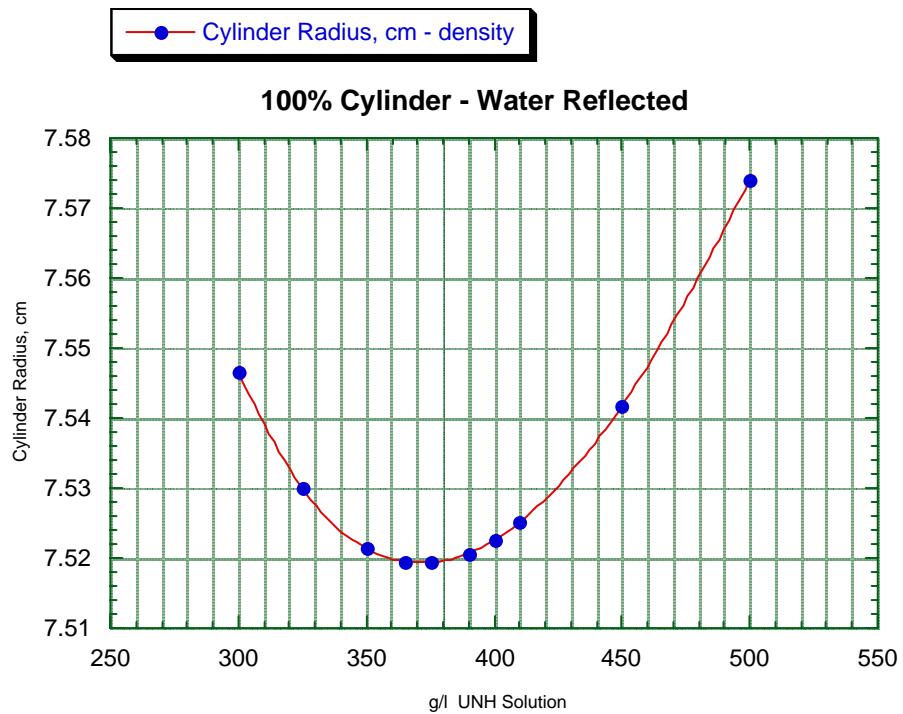


Fig. B.20. Radius vs density for 100% enriched UNH–H₂O cylinders.

APPENDIX C

**GRAPHS OF $\text{PuO}_2\text{-H}_2\text{O}$, WATER REFLECTED (30 cm):
CRITICAL PARAMETERS vs FUEL VOLUME FRACTIONS**

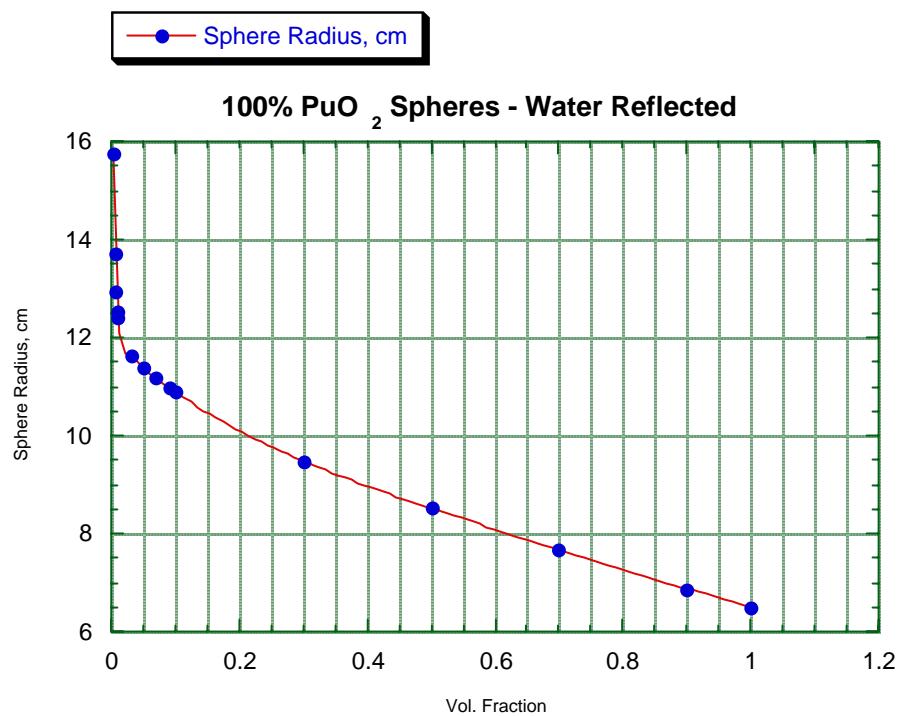


Fig. C.1. Radius vs volume fraction for 100% enriched PuO₂-H₂O spheres.

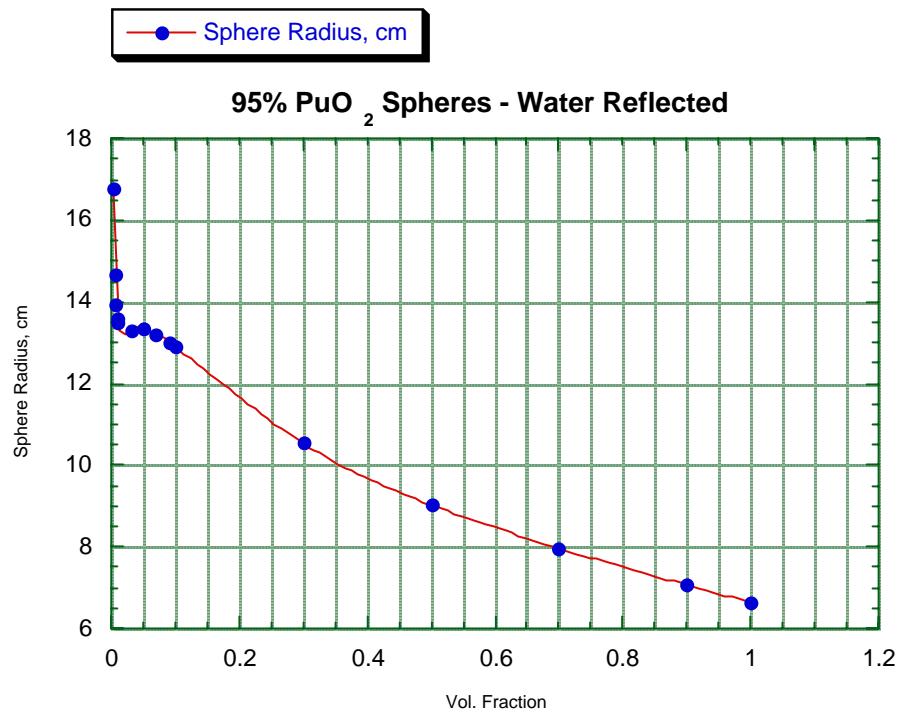


Fig. C.2. Radius vs volume fraction for 95% enriched PuO₂-H₂O spheres.

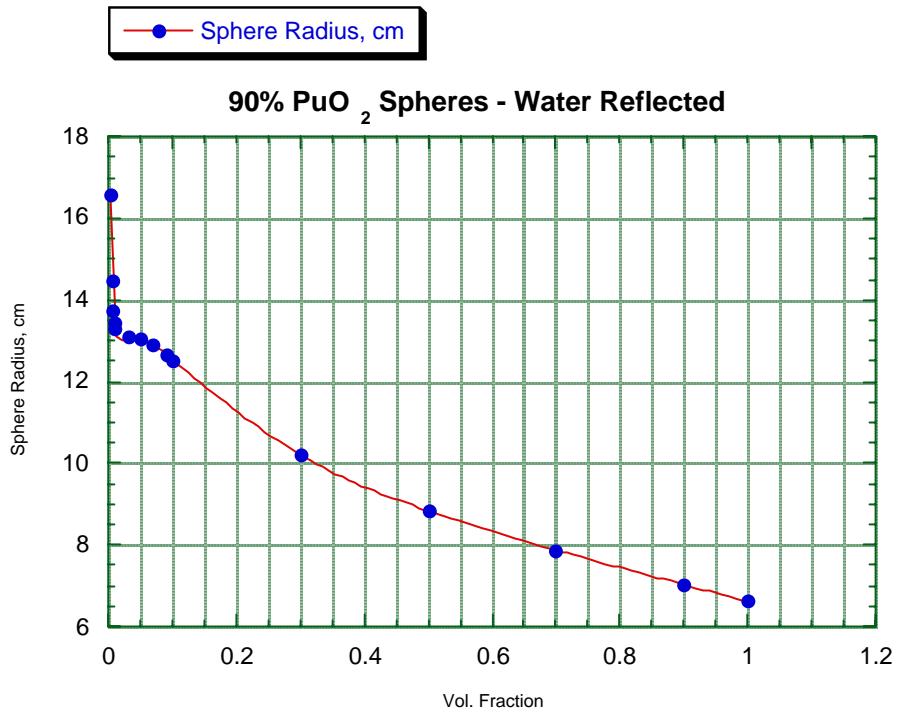


Fig. C.3. Radius vs volume fraction for 90% enriched $\text{PuO}_2\text{-H}_2\text{O}$ spheres.

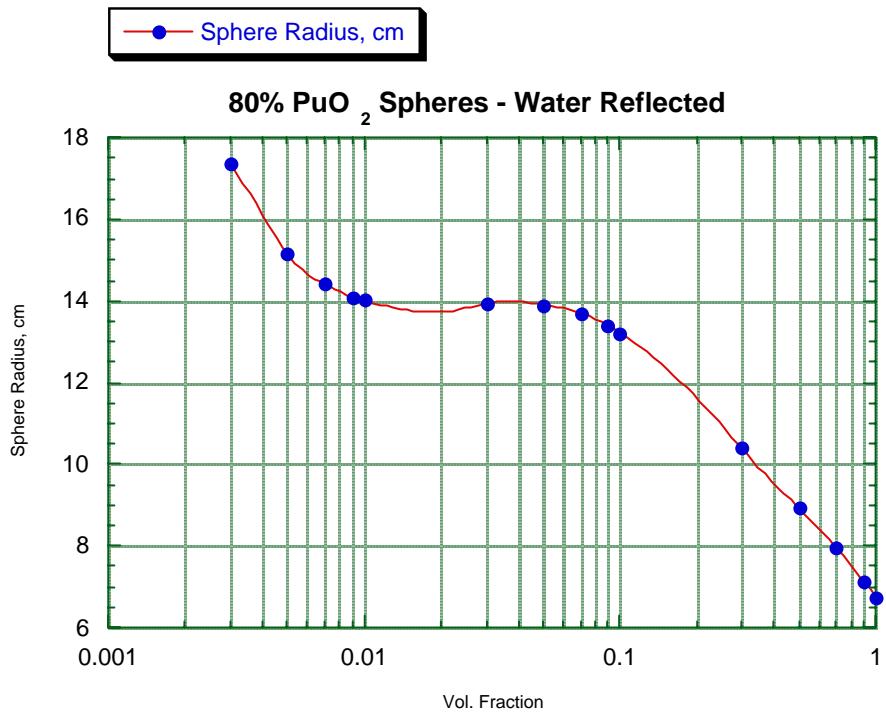


Fig. C.4. Radius vs volume fraction for 80% enriched $\text{PuO}_2\text{-H}_2\text{O}$ spheres.

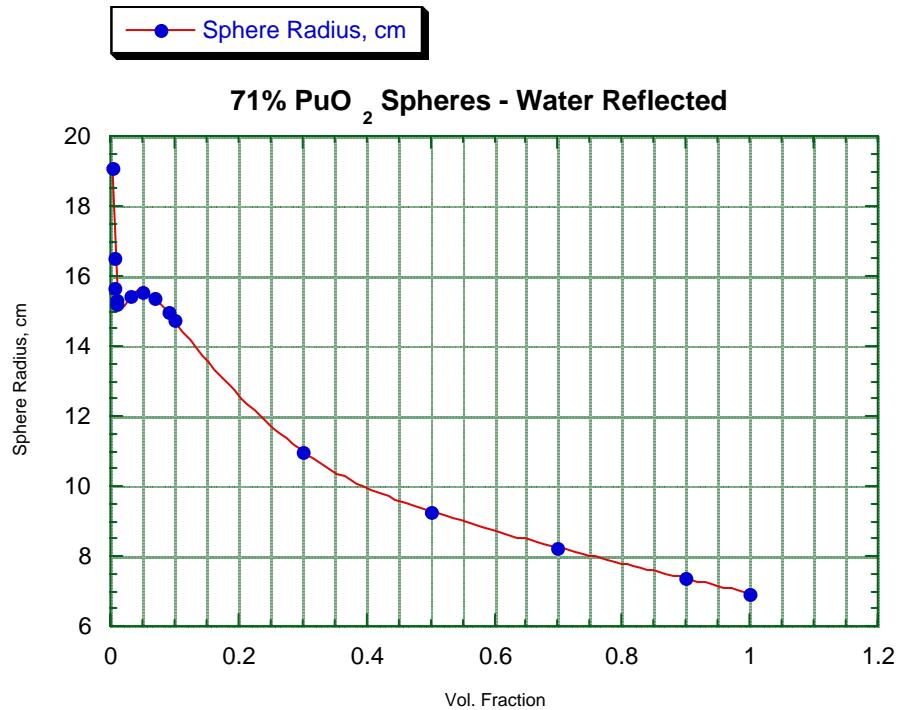


Fig. C.5. Radius vs volume fraction for 71% enriched PuO₂-H₂O spheres.

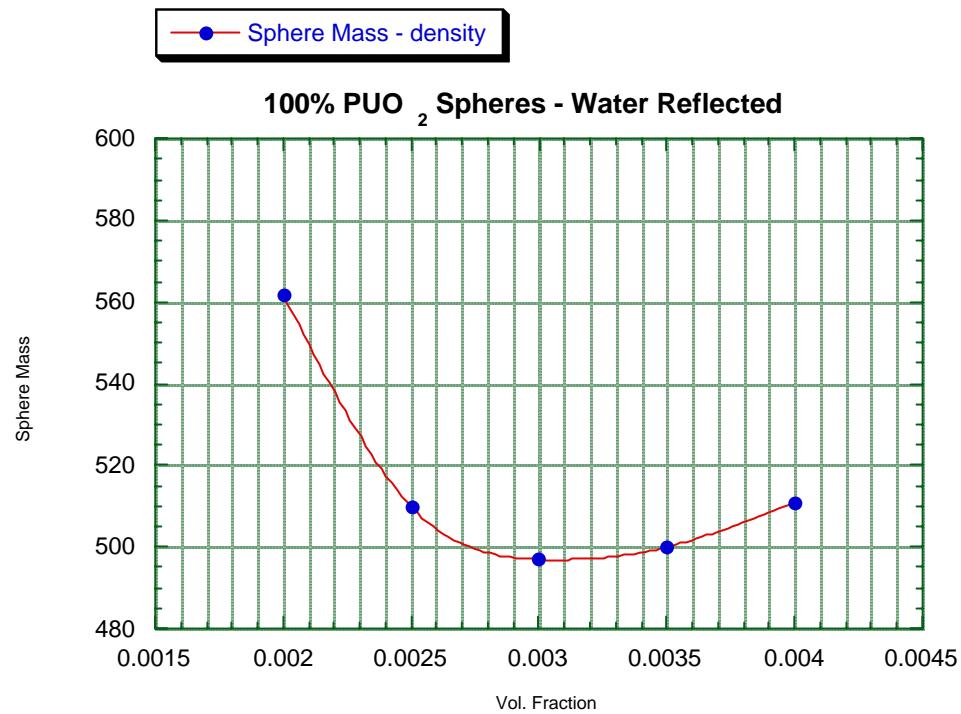


Fig. C.6. Mass vs volume fraction for 100% enriched $\text{PuO}_2\text{-H}_2\text{O}$ spheres.

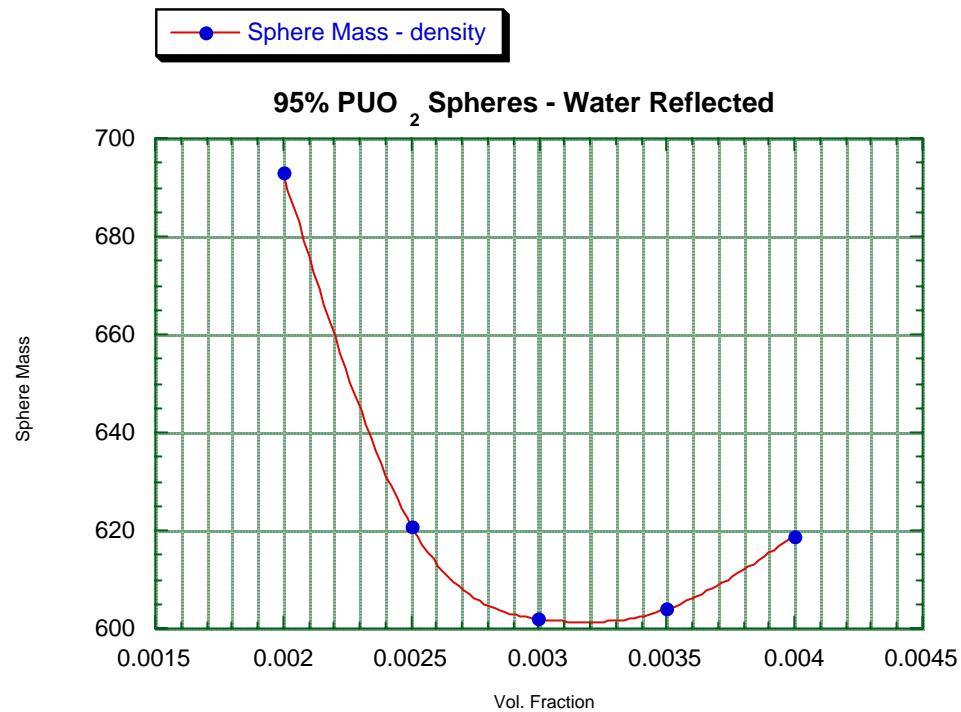


Fig. C.7. Mass vs volume fraction for 95% enriched $\text{PuO}_2\text{-H}_2\text{O}$ spheres.

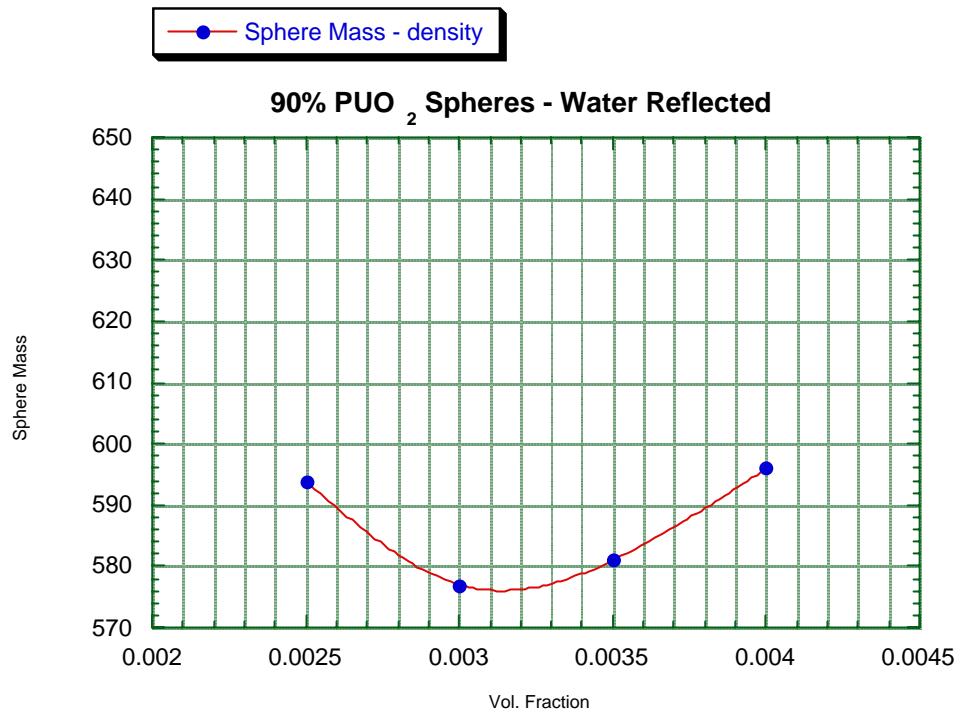


Fig. C.8. Mass vs volume fraction for 90% enriched $\text{PuO}_2\text{-H}_2\text{O}$ spheres.

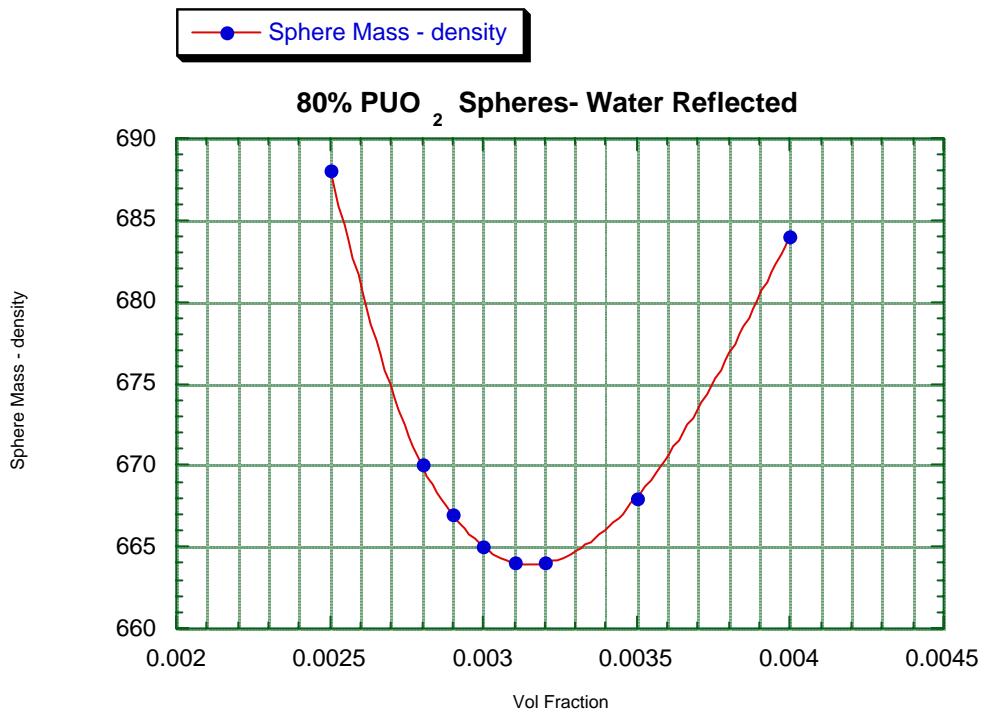


Fig. C.9. Mass vs volume fraction for 80% enriched $\text{PuO}_2\text{-H}_2\text{O}$ spheres.

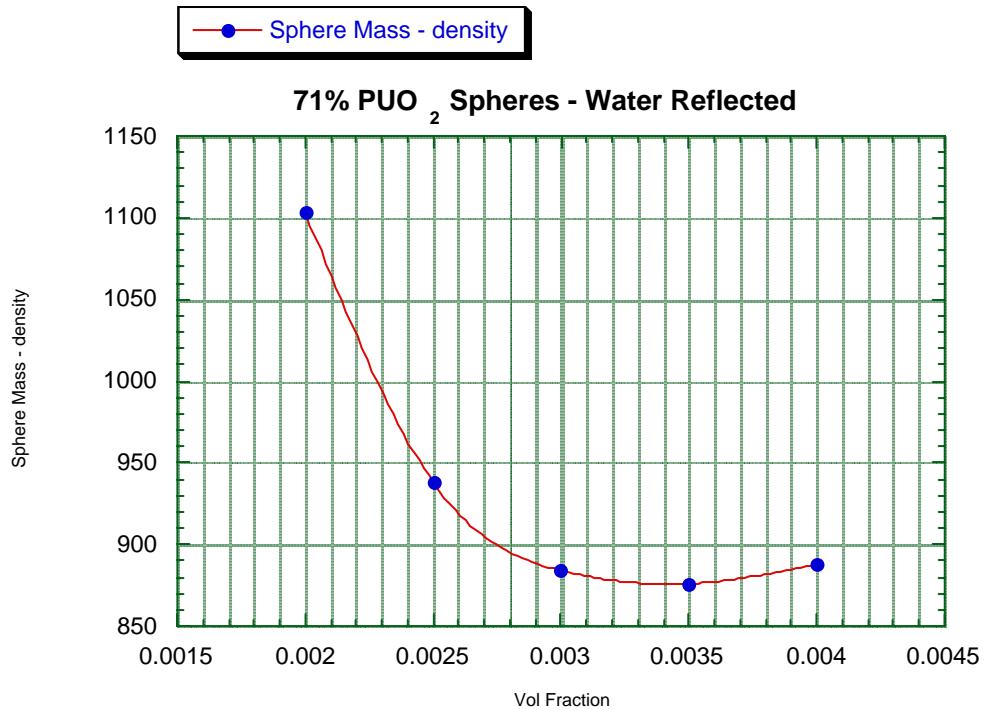


Fig. C.10. Mass vs volume fraction for 71% enriched PuO₂-H₂O spheres.

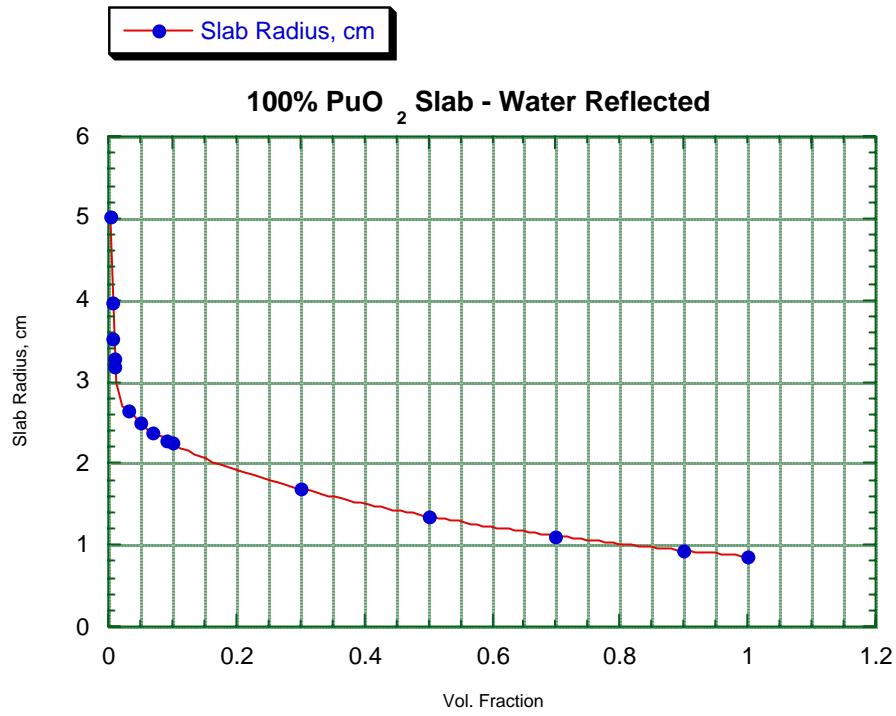


Fig. C.11. Radius vs volume fraction for 100% enriched $\text{PuO}_2\text{-H}_2\text{O}$ slabs.

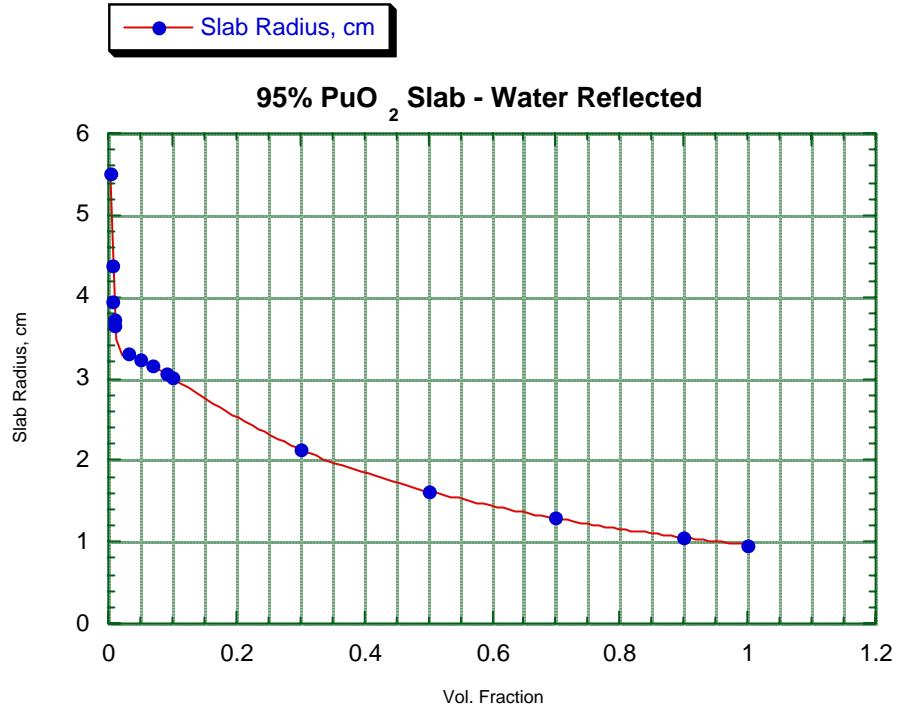


Fig. C.12. Radius vs volume fraction for 95% enriched $\text{PuO}_2\text{-H}_2\text{O}$ slabs.

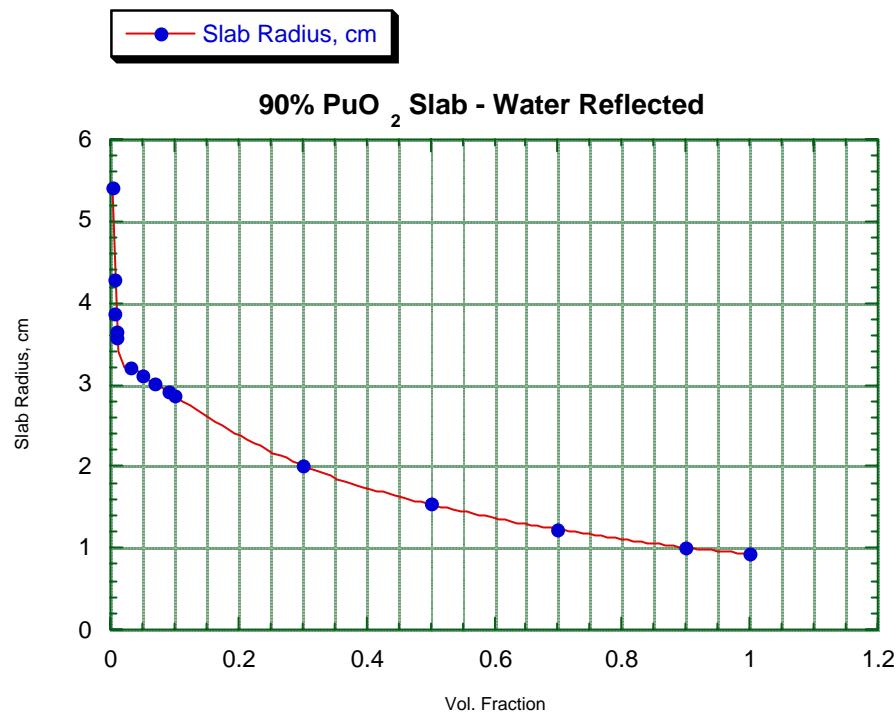


Fig. C.13. Radius vs volume fraction for 90% enriched $\text{PuO}_2\text{-H}_2\text{O}$ slabs.

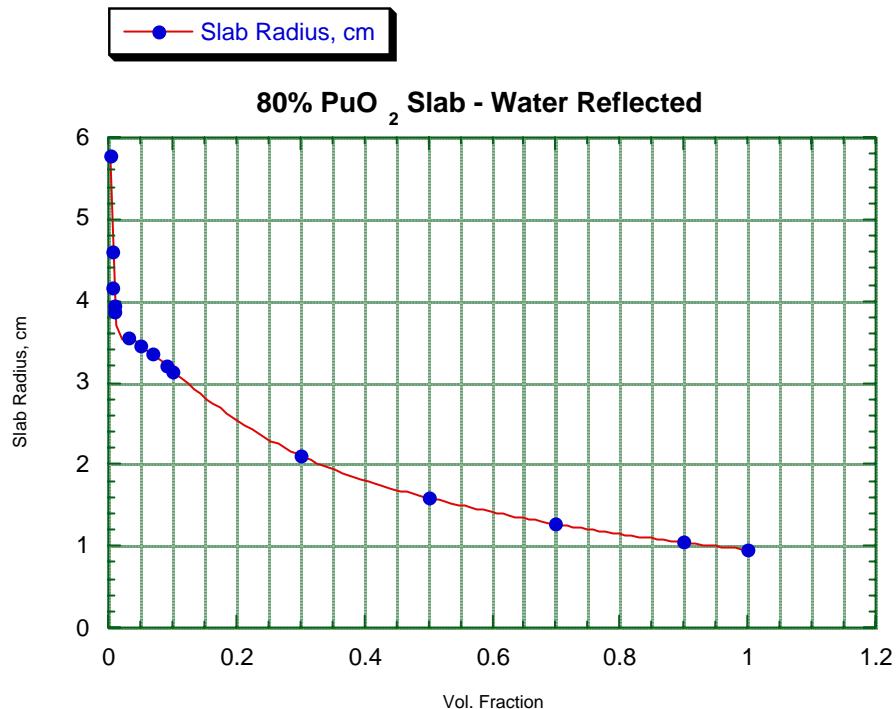


Fig. C.14. Radius vs volume fraction for 80% enriched $\text{PuO}_2\text{-H}_2\text{O}$ slabs.

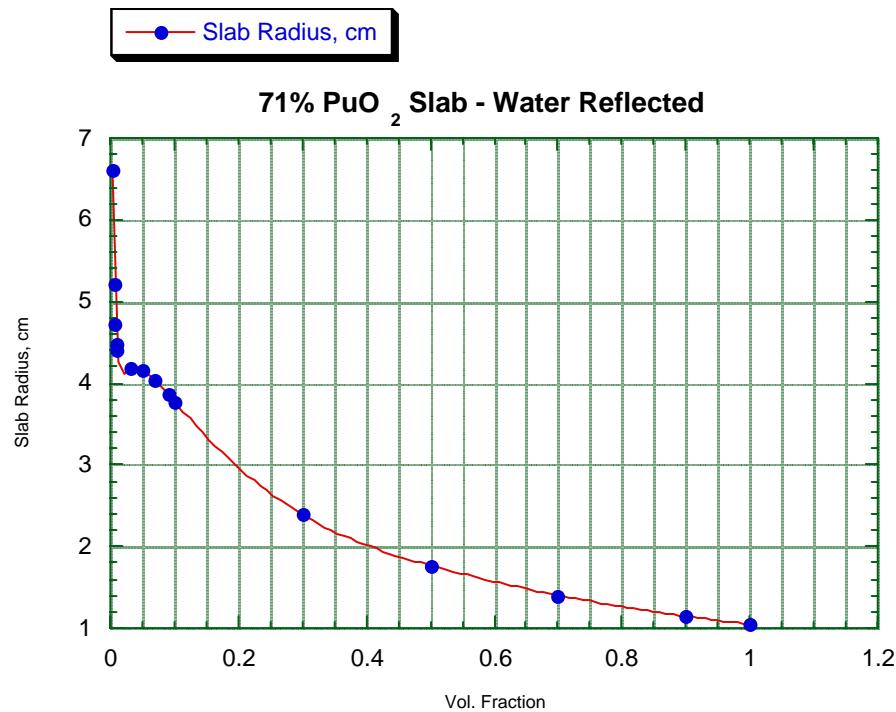


Fig. C.15. Radius vs volume fraction for 71% enriched $\text{PuO}_2\text{-H}_2\text{O}$ slabs.

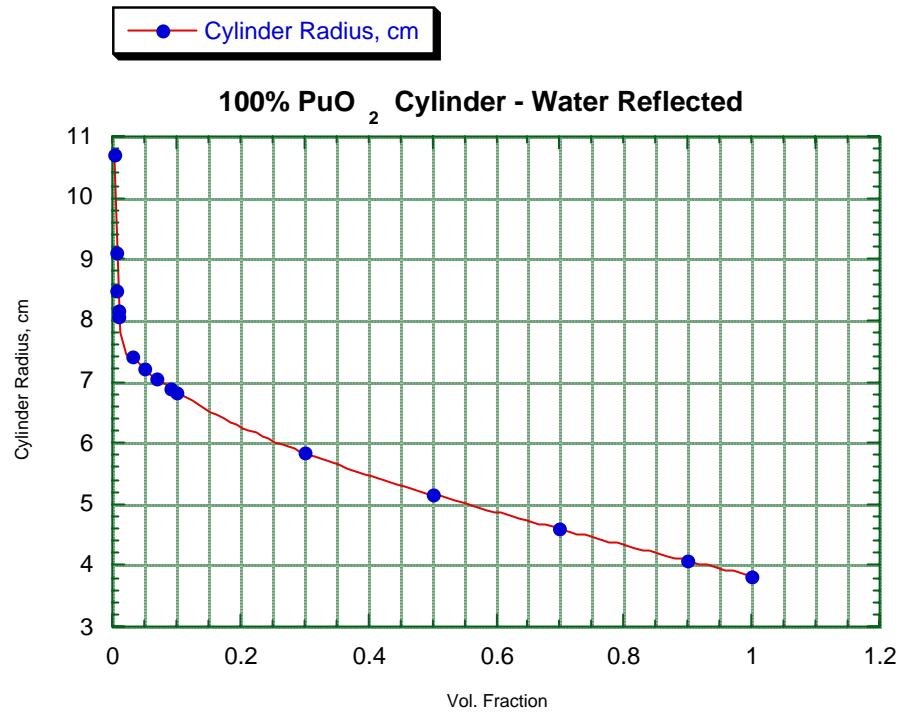


Fig. C.16. Radius vs volume fraction for 100% enriched $\text{PuO}_2\text{-H}_2\text{O}$ cylinders.

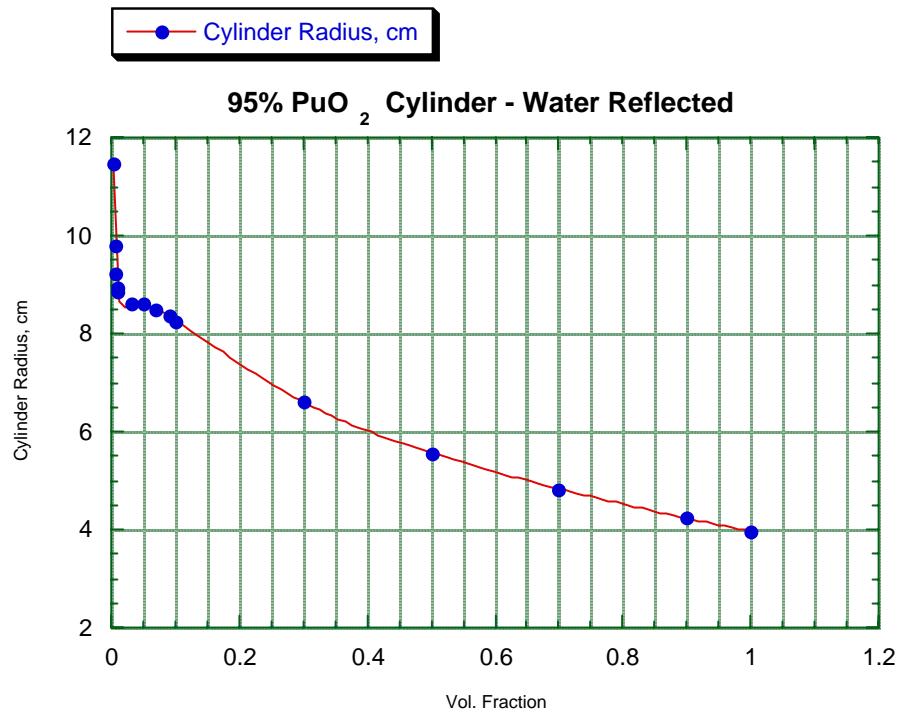


Fig. C.17. Radius vs volume fraction for 95% enriched $\text{PuO}_2\text{-H}_2\text{O}$ cylinders.

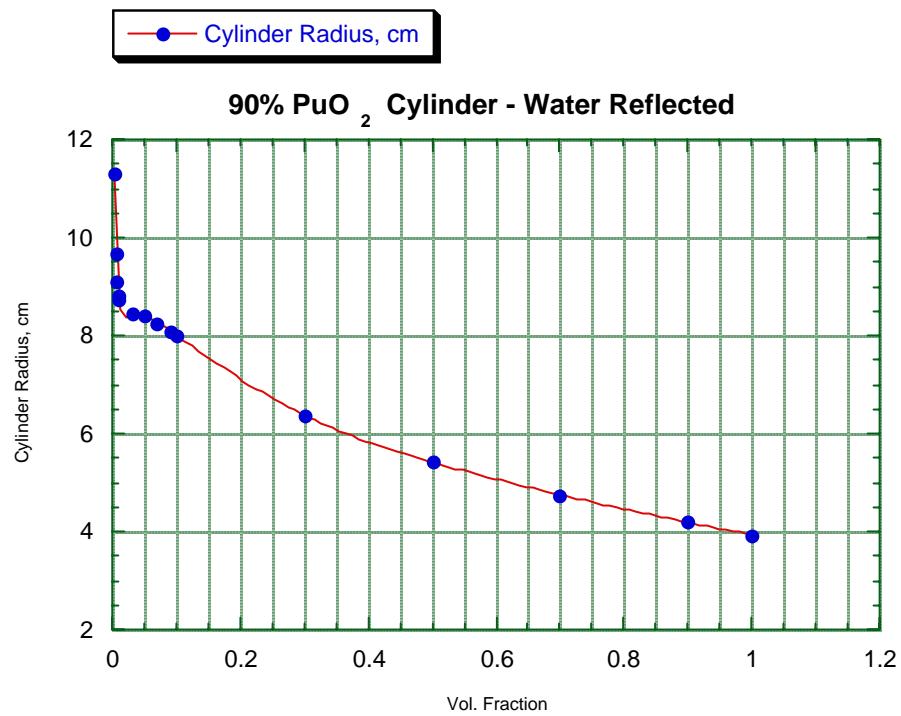


Fig. C.18. Radius vs volume fraction for 90% enriched PuO_2 - H_2O cylinders.

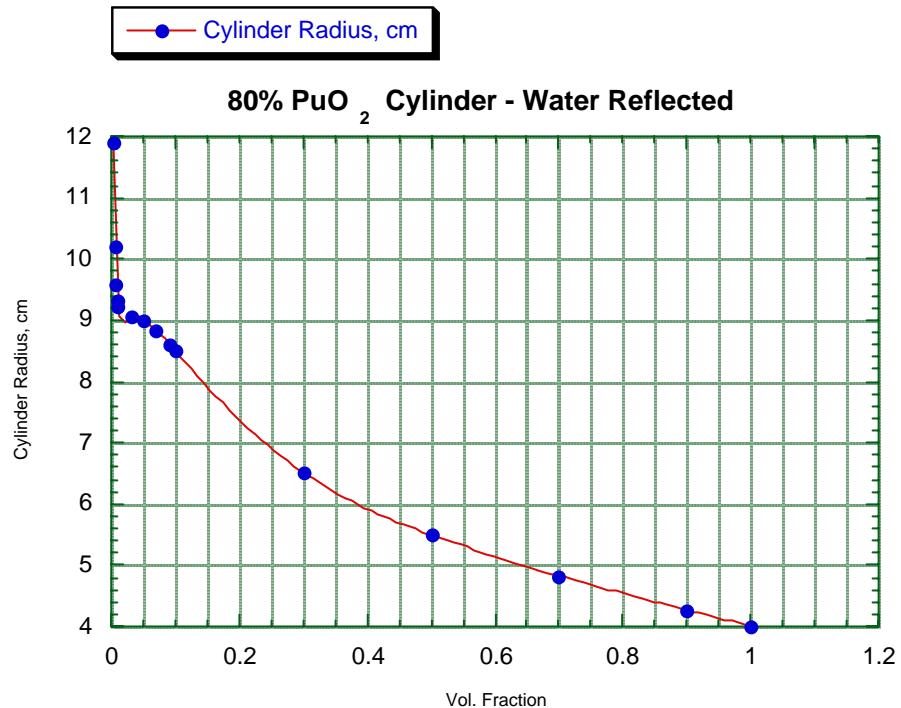


Fig. C.19. Radius vs volume fraction for 80% enriched PuO_2 - H_2O cylinders.

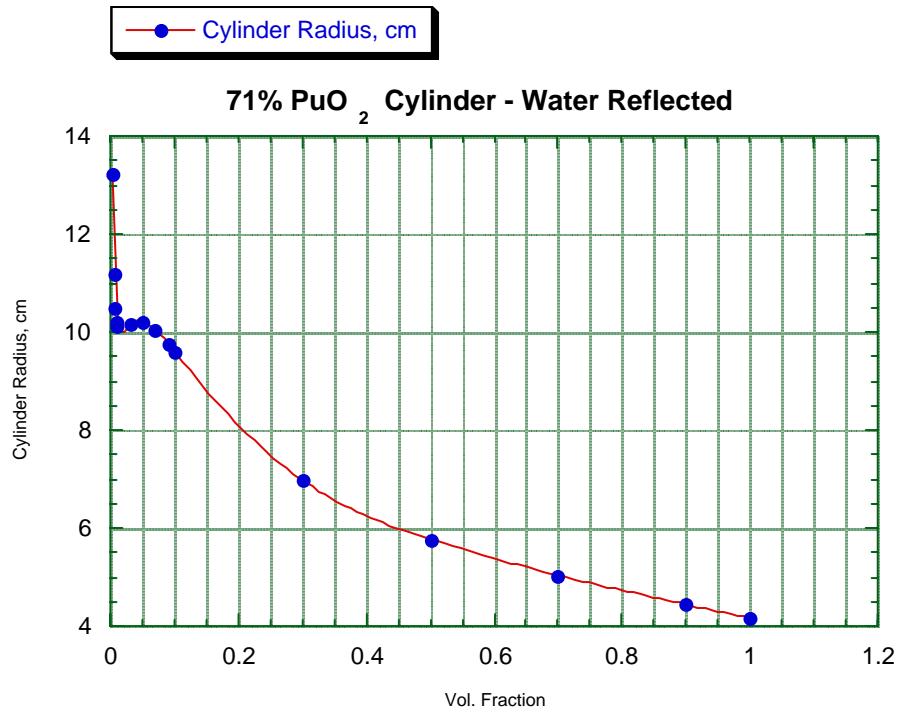


Fig. C.20. Radius vs volume fraction for 71% enriched PuO₂-H₂O cylinders.

APPENDIX D

**GRAPHS OF PuNH-H₂O, WATER REFLECTED (30 cm):
CRITICAL PARAMETERS vs PLUTONIUM DENSITY**

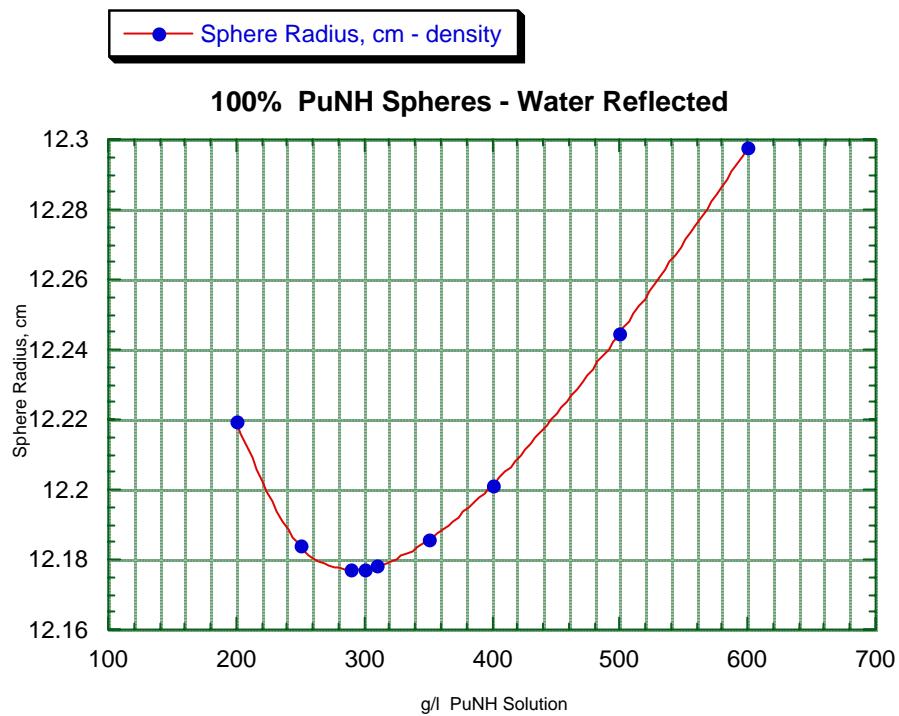


Fig. D.1. Radius vs density for 100% enriched PuNH–H₂O spheres.

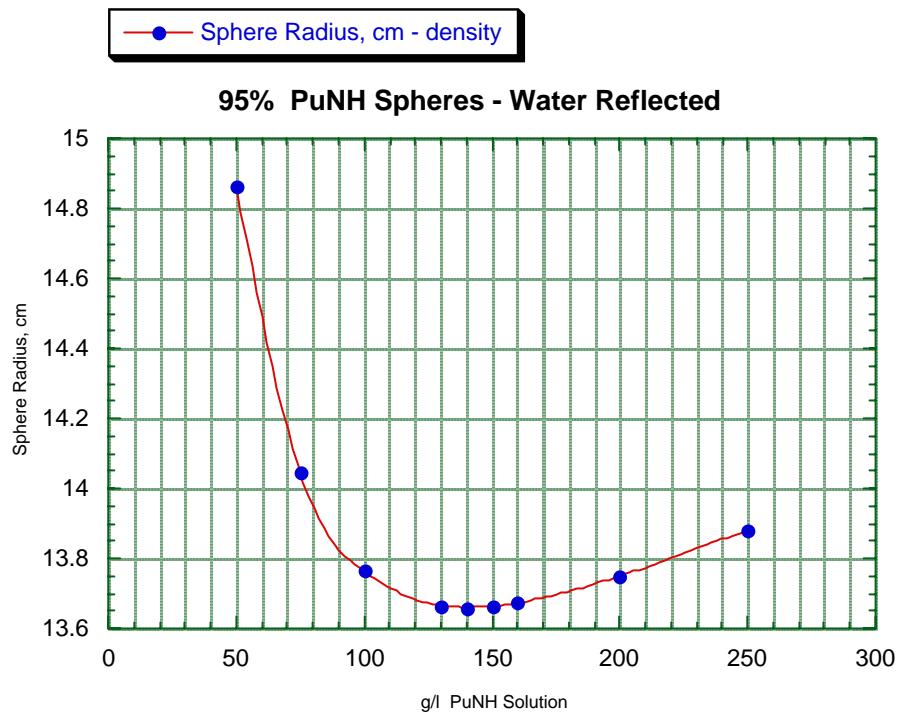


Fig. D.2. Radius vs density for 95% enriched PuNH–H₂O spheres.

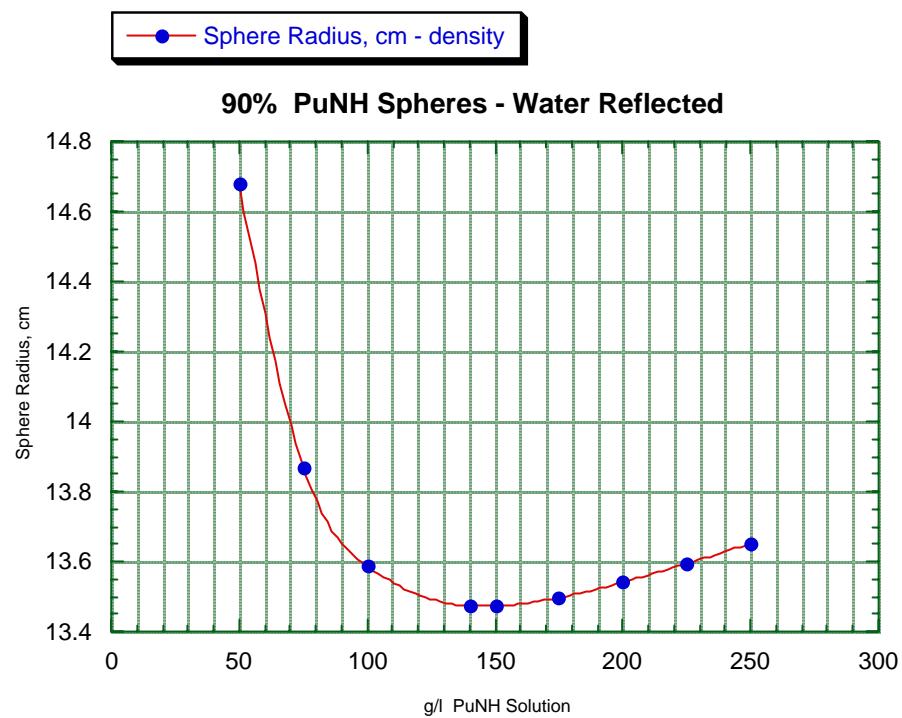


Fig. D.3. Radius vs density for 90% enriched PuNH–H₂O spheres.

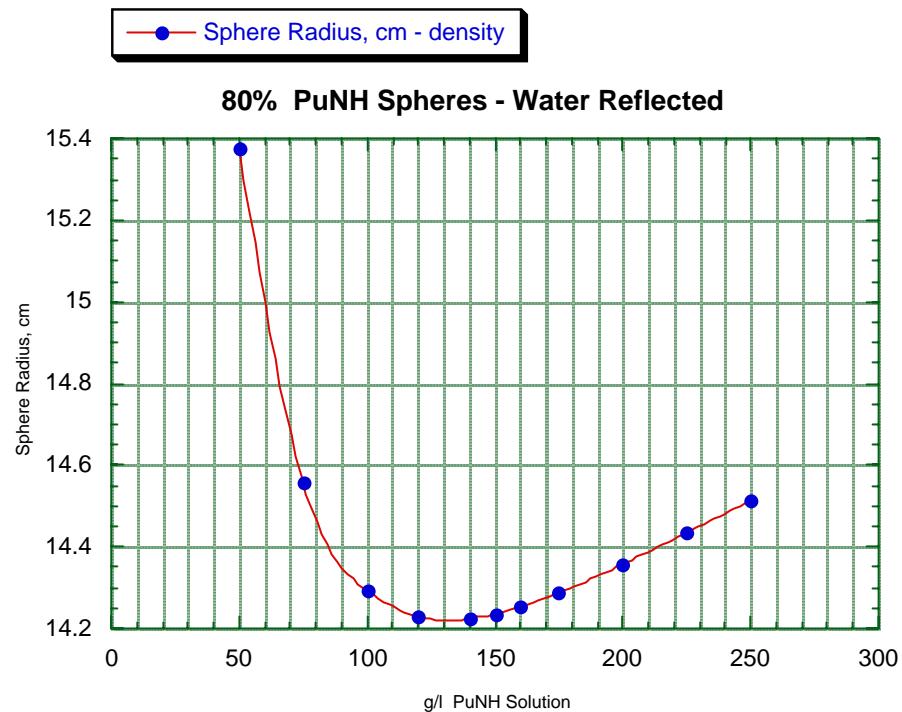


Fig. D.4. Radius vs density for 80% enriched PuNH–H₂O spheres.

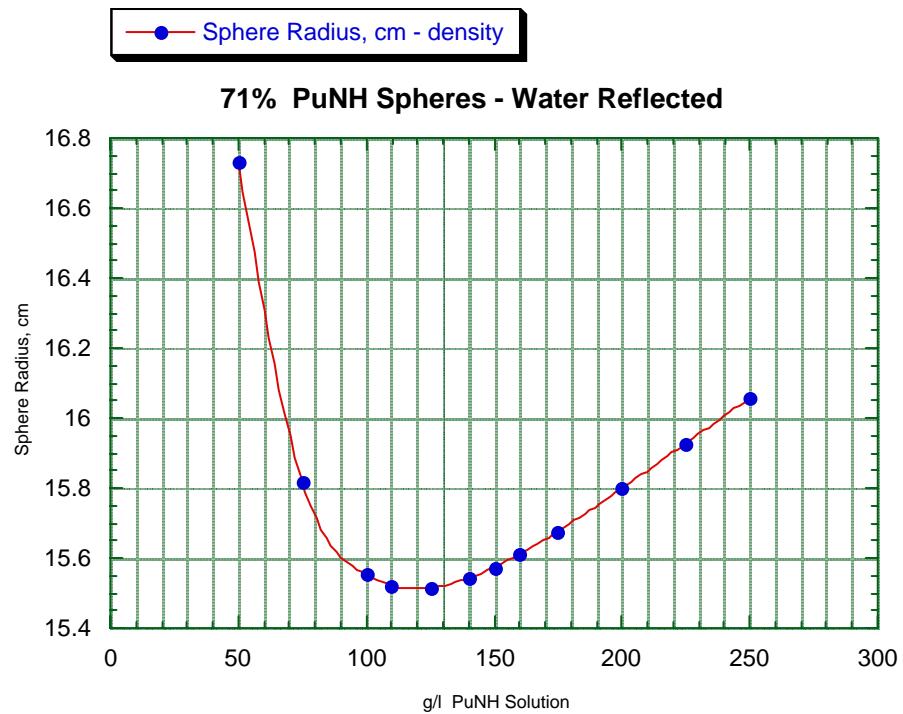


Fig. D.5. Radius vs density for 71% enriched PuNH–H₂O spheres.

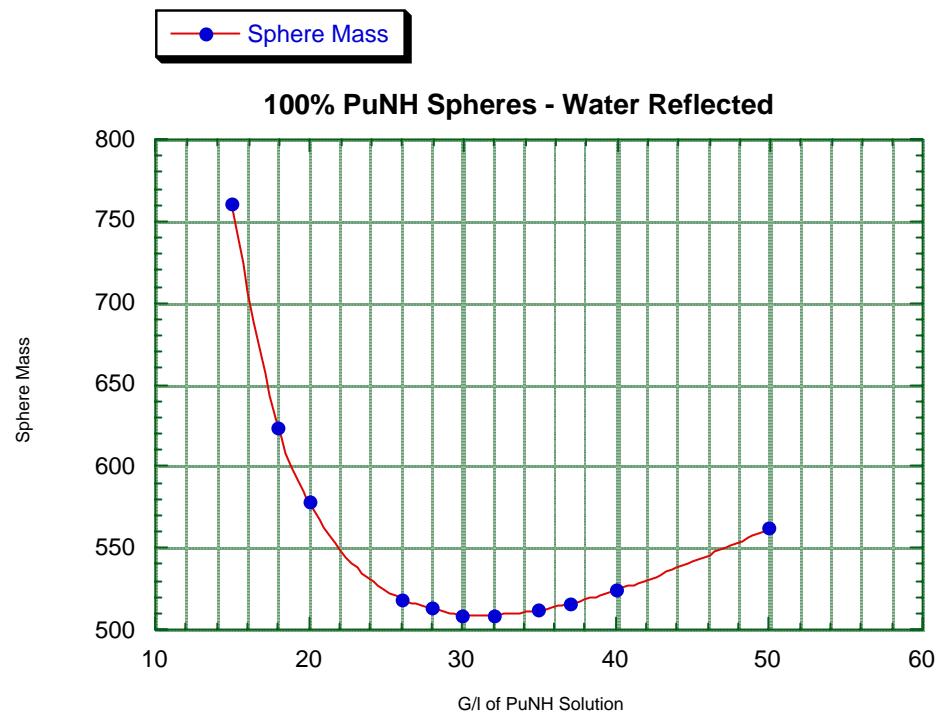


Fig. D.6. Mass vs density for 100% enriched PuNH–H₂O spheres.

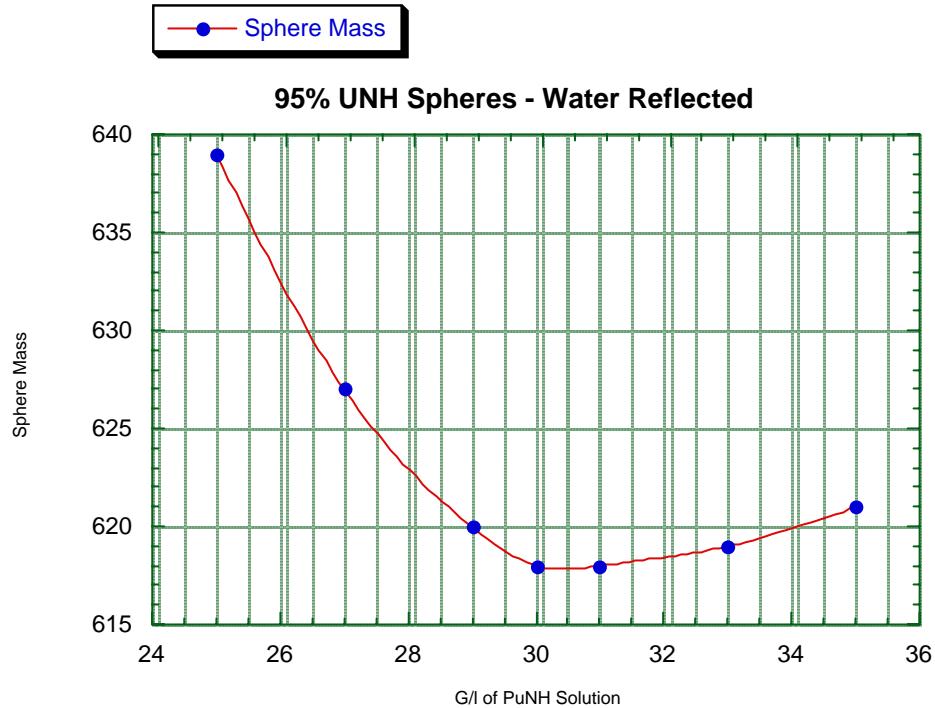


Fig. D.7. Mass vs density for 95% enriched PuNH–H₂O spheres.

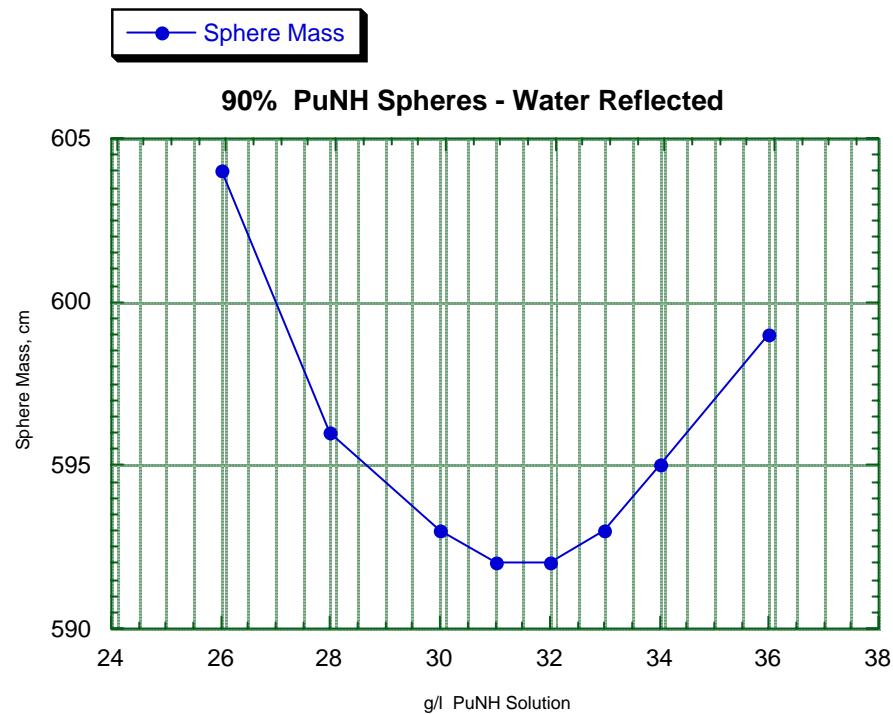


Fig. D.8. Mass vs density for 90% enriched PuNH–H₂O spheres.

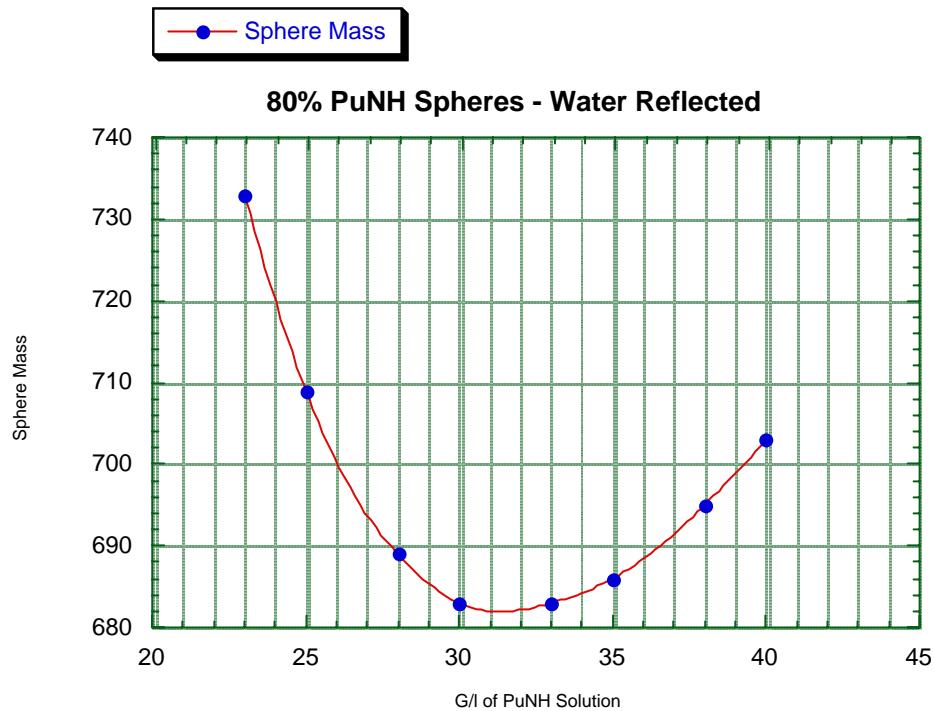


Fig. D.9. Mass vs density for 80% enriched PuNH–H₂O spheres.

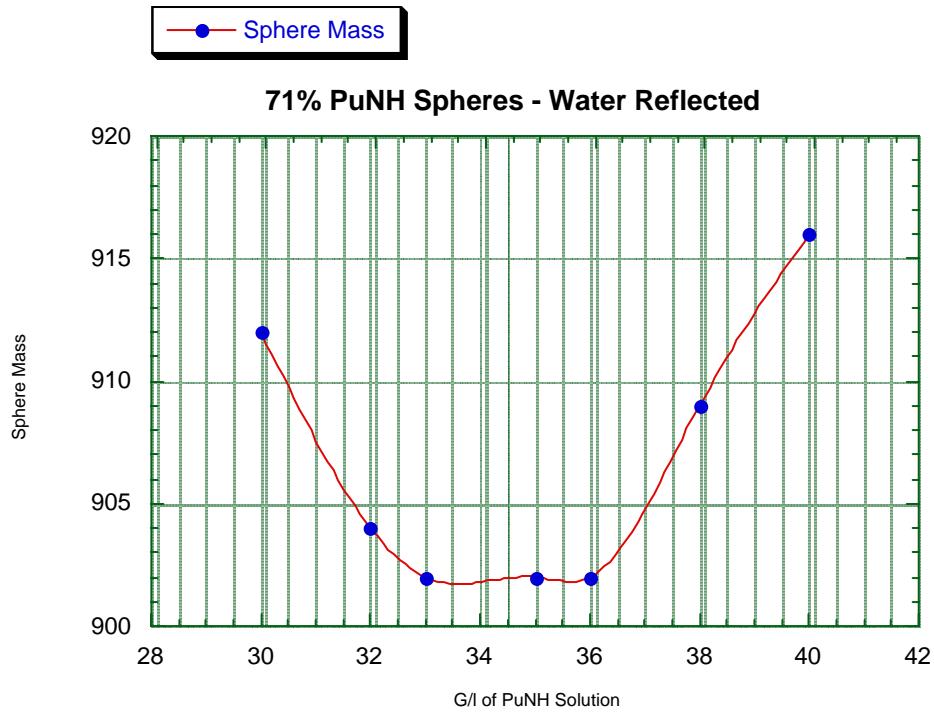


Fig. D.10. Mass vs density for 71% enriched PuNH–H₂O spheres.

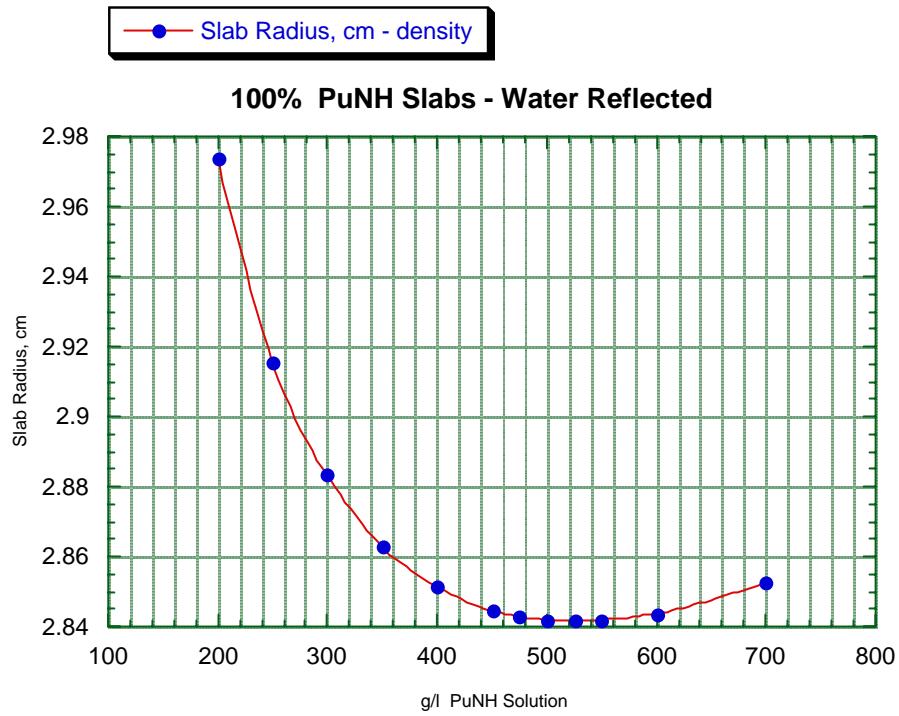


Fig. D.11. Radius vs density for 100% enriched PuNH–H₂O slabs.

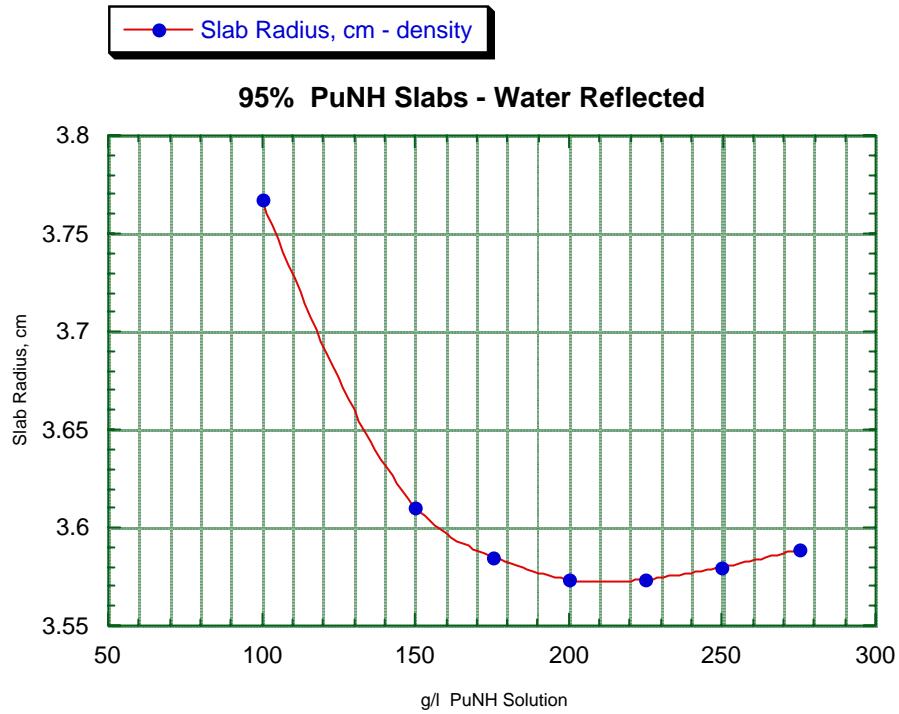


Fig. D.12. Radius vs density for 95% enriched PuNH–H₂O slabs.

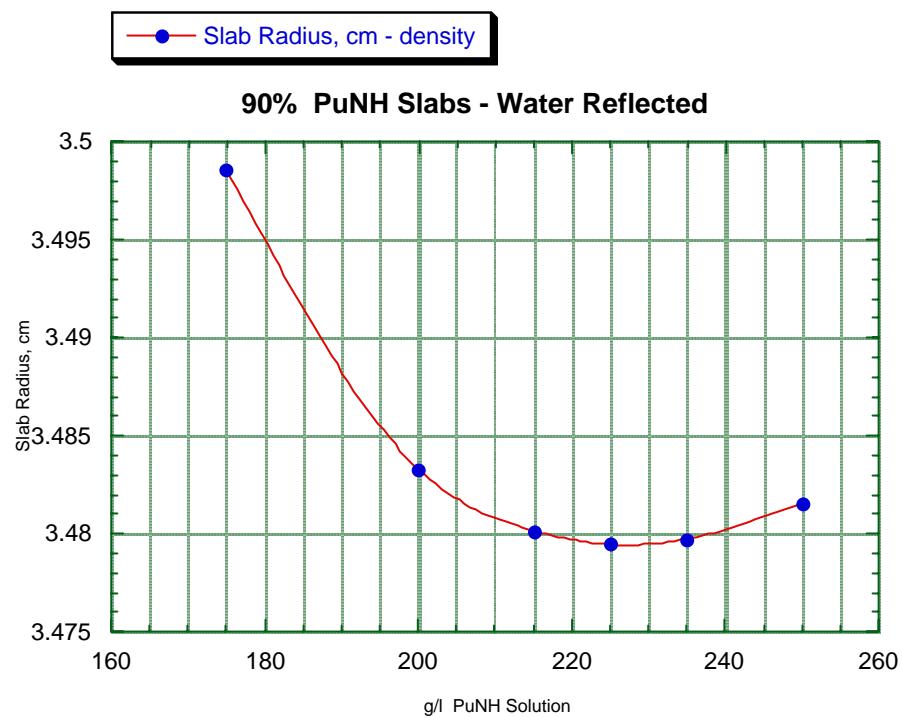


Fig. D.13. Radius vs density for 90% enriched PuNH–H₂O slabs.

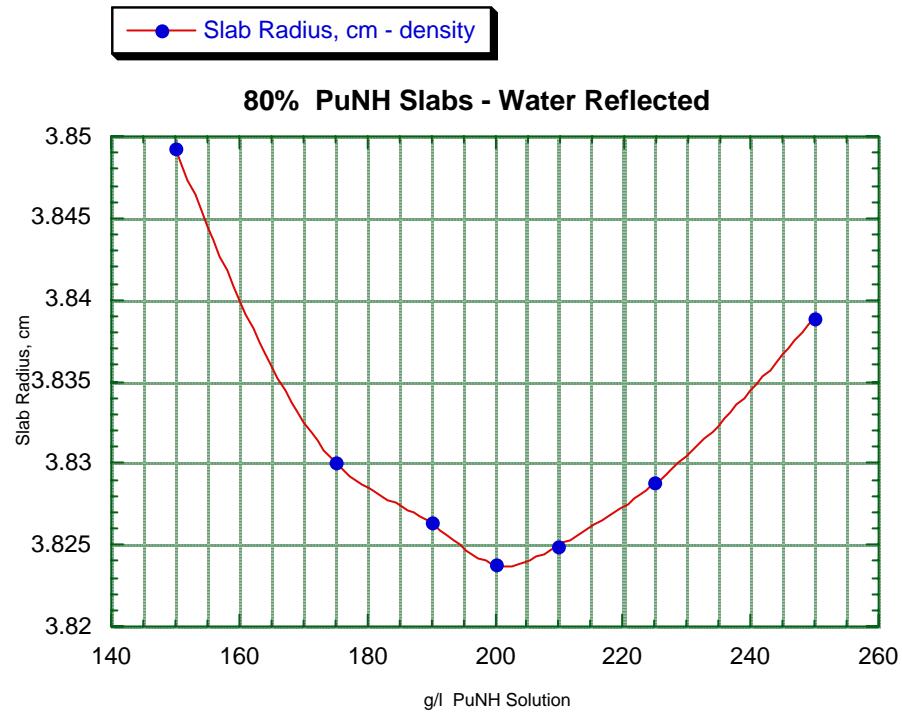


Fig. D.14. Radius vs density for 80% enriched PuNH–H₂O slabs.

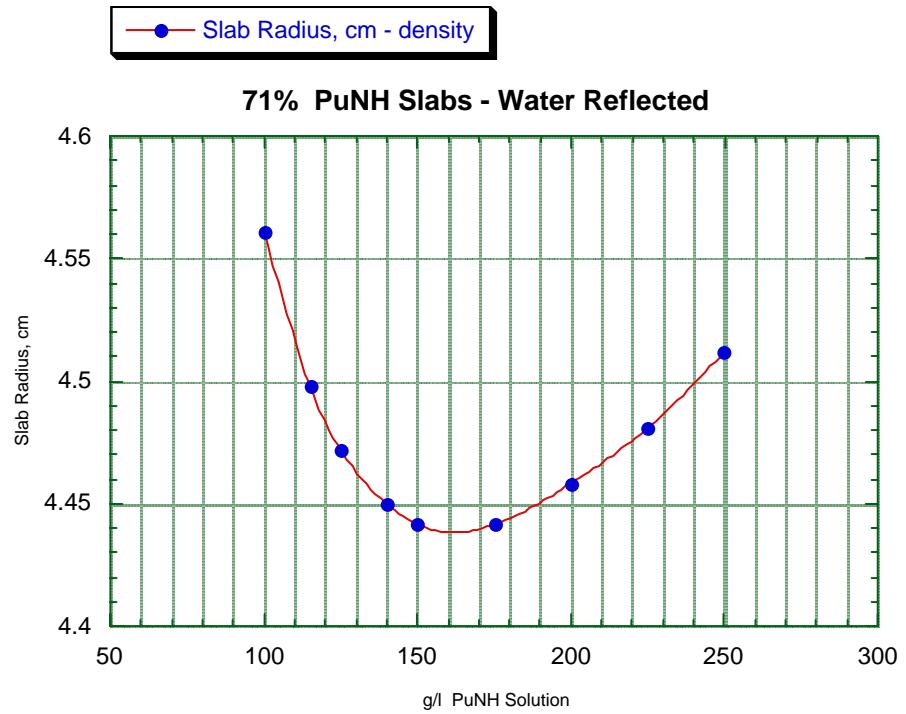


Fig. D.15. Radius vs density for 71% enriched PuNH–H₂O slabs.

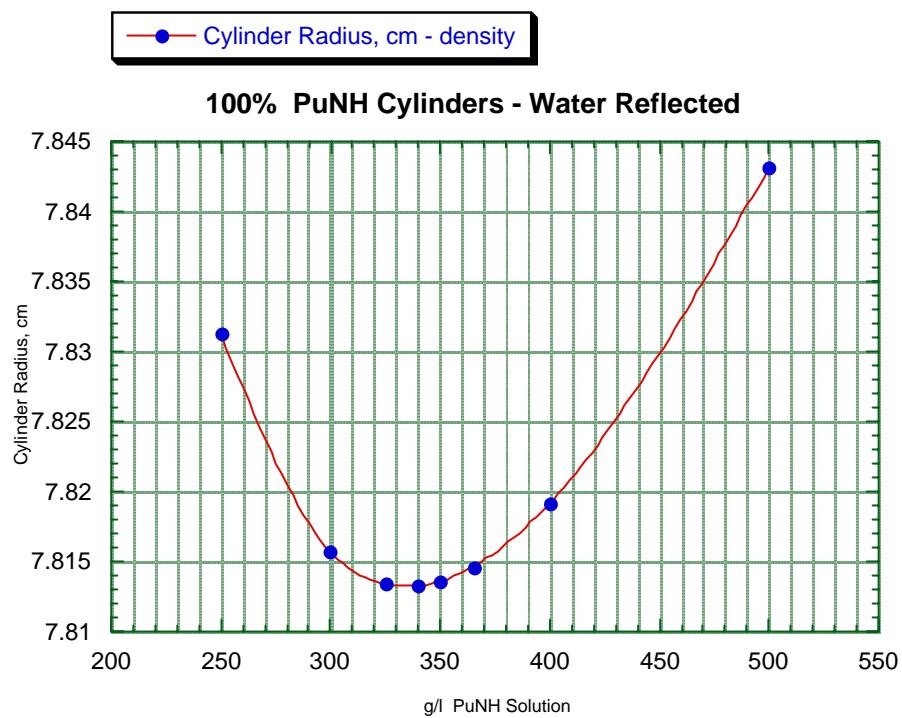


Fig. D.16. Radius vs density for 100% enriched PuNH–H₂O cylinders.

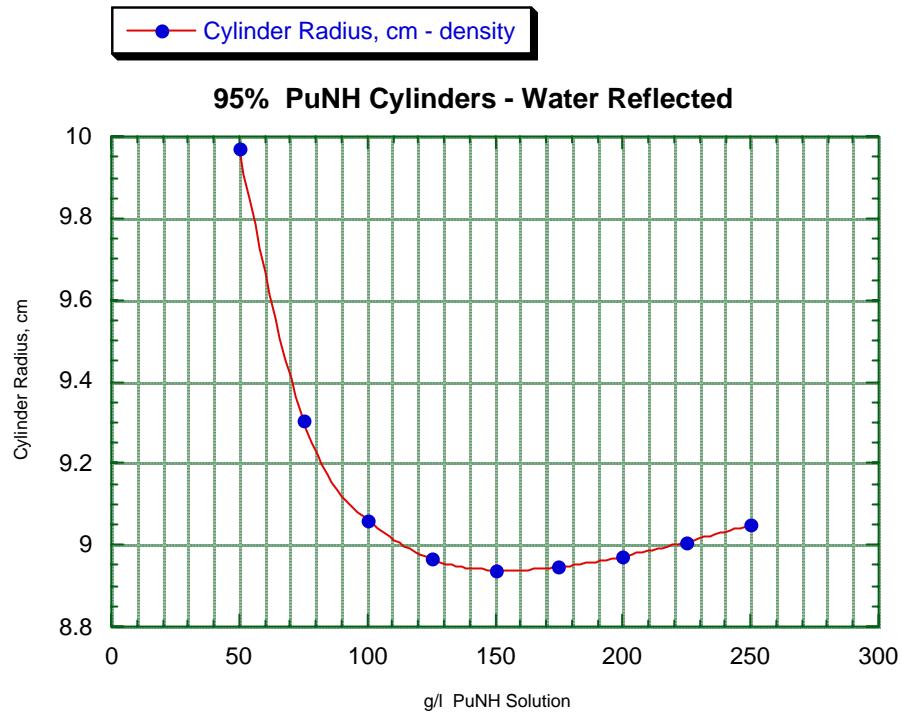


Fig. D.17. Radius vs density for 95% enriched PuNH–H₂O cylinders.

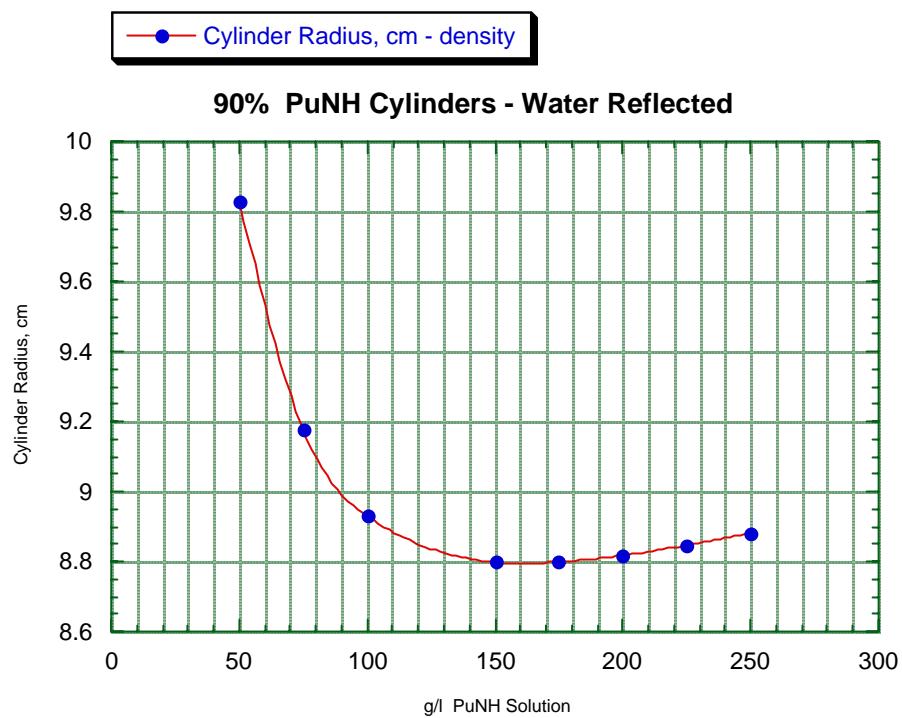


Fig. D.18. Radius vs density for 90% enriched PuNH–H₂O cylinders.

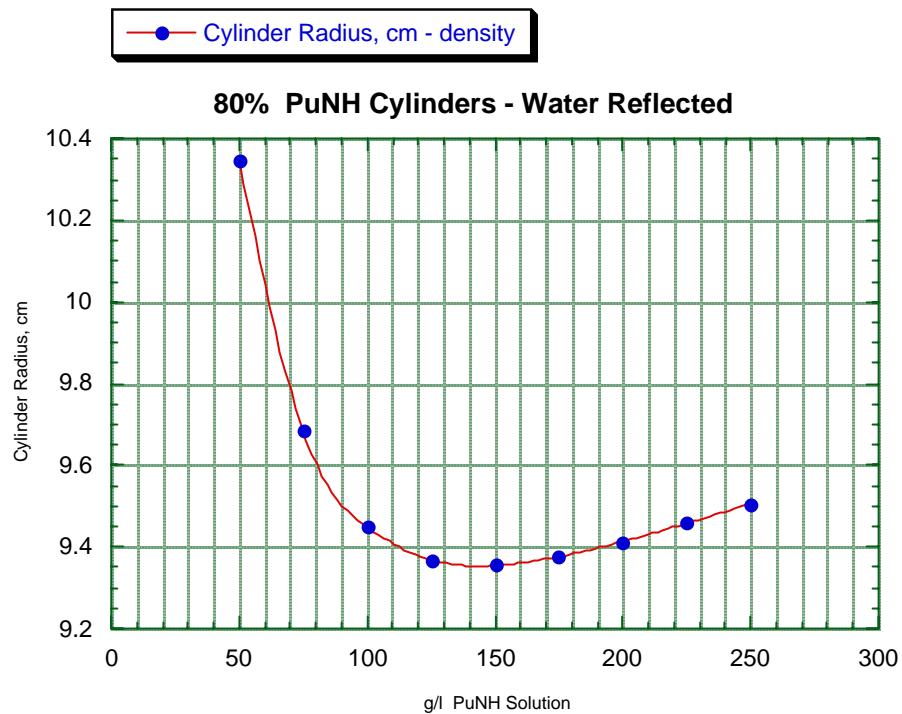


Fig. D.19. Radius vs density for 80% enriched PuNH-H₂O cylinders.

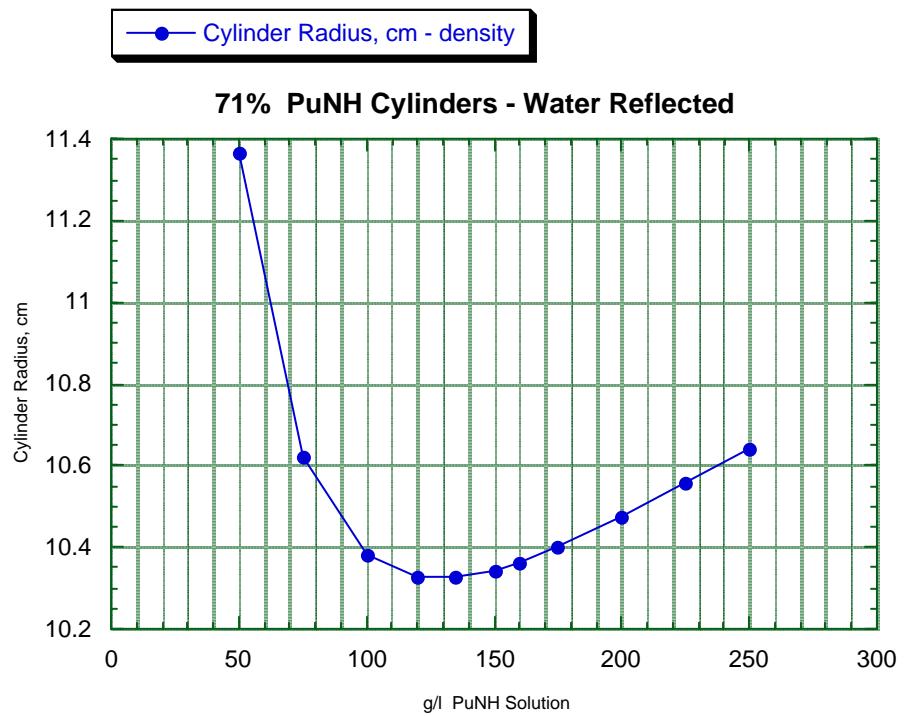


Fig. D.20. Radius vs density for 71% enriched PuNH–H₂O cylinders.

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