Verification of the $^{239}Pu(NO_3)_4$ Solution Fissile Concentration Subcritical Limit in ANSI/ANS-8.1-2014

Doug Bowen
Nuclear Data and Criticality Safety Group Leader
Reactor and Nuclear Systems Division
Oak Ridge National Laboratory

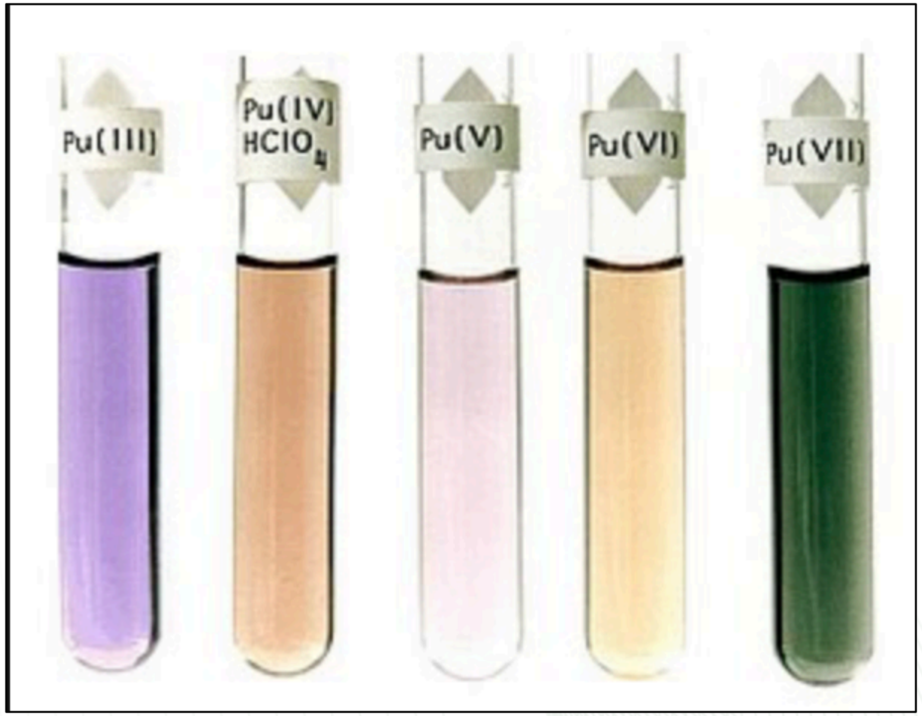
Nick Brown
Nuclear Criticality Safety Manager
Nuclear Fuel Services, Inc.

Travis Greene
Reactor and Nuclear Systems Division
Oak Ridge National Laboratory

2019 Winter ANS Meeting
November 20, 2019

Agenda

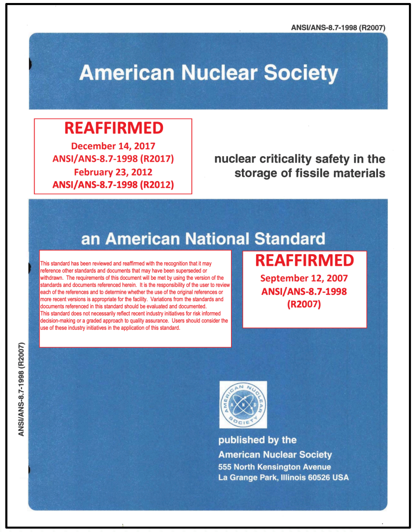
- Introduction
- Development of Pu Nitrate Solution Subcritical Limits (SCLs)
- SCL History
- Fissile Concentration Subcritical Limit (FCSL) Computation Results
- Concluding remarks

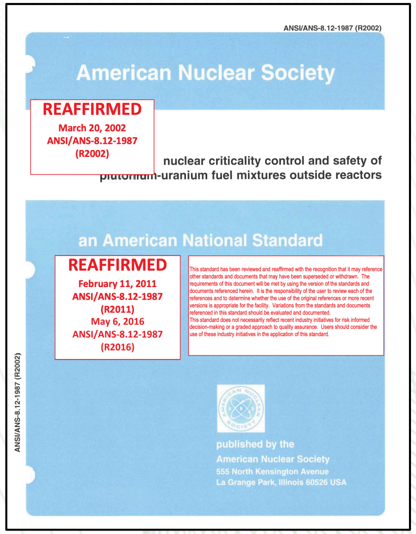


OAK RIDGE National Laboratory

Introduction

- What is a subcritical limit?
  - “The limiting value assigned to a controlled parameter that results in a subcritical system under specified conditions.”
  - “The controlled parameter limit allows for uncertainties in the calculations and experimental data used in its derivation **but not for contingencies…**”
  - Several ANS standards provide subcritical limits: ANS-8.1, ANS-8.7, ANS-8.12, and ANS-8.15
  - Bases are provided in the literature or in the standard appendices
    - Bases are working group consensus values
    - May be based on validated computations or a combination of computations and critical experiments
  - ANS-8.1 Pu SCL bases are provided in a 1981 *Nuclear Science and Engineering* article, “Subcritical Limits for Plutonium Systems” (Hugh Clark, Savannah River Laboratory)



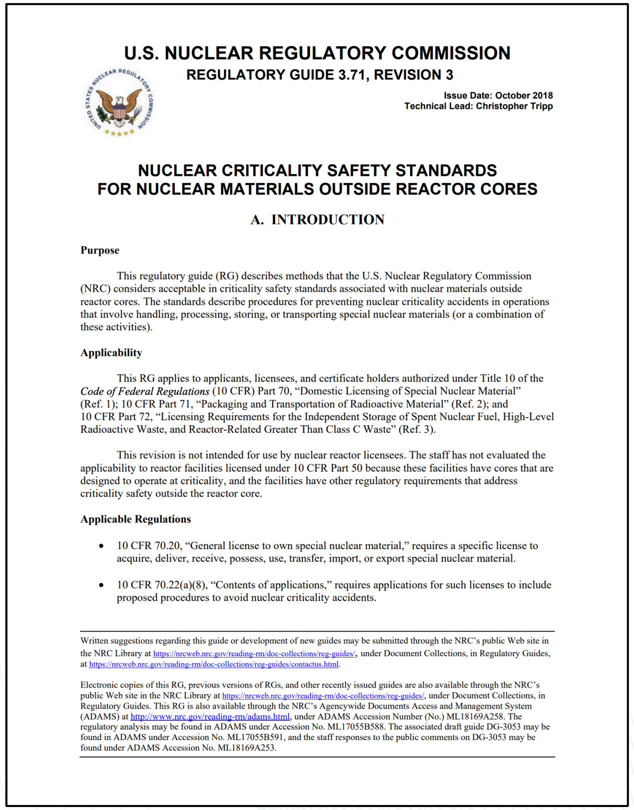


OAK RIDGE National Laboratory

Issue – Fissile Concentration Subcritical Limit for $Pu(NO_3)_2$

- NRC did not endorse the fissile concentration subcritical limit in Table 1, ANS-8.1
  - Guidance to NRC licensees was from Regulatory Guide 3.71, Revision 3 (2018)
  - As stated, the NRC requires a licensee to utilize their license-approved NCS validated computational methods to develop their own SCLs or to ensure the ^{240}Pu content, in this case, is conservative relative to the assumptions used in the SCL rather than use the FCSL in the ANS-8.1 standard

Parameter	Subcritical Limit for Fissile Solute $^{239}Pu(NO_3)_4$ [16]
Mass of fissile nuclide (kg)	0.48
Diameter of cylinder of solution (cm)	15.4
Thickness of slab of solution (cm)	5.5
Volume of solution (L)	7.3
Concentration of fissile nuclide (g/L)	7.3
Atomic ratio of hydrogen to fissile nuclide[1)]	3630
Areal density of fissile nuclide (g/cm^2)	0.25


U.S. NUCLEAR REGULATORY COMMISSION

REGULATORY GUIDE 3.71, REVISION 3

Issue Date: October 2018
Technical Lead: Christopher Tripp

NUCLEAR CRITICALITY SAFETY STANDARDS
FOR NUCLEAR MATERIALS OUTSIDE REACTOR CORES

A. INTRODUCTION

Purpose

This regulatory guide (RG) describes methods that the U.S. Nuclear Regulatory Commission (NRC) considers acceptable in criticality safety standards associated with nuclear materials outside reactor cores. The standards describe procedures for preventing nuclear criticality accidents in operations that involve handling, processing, storing, or transporting special nuclear materials (or a combination of these activities).

Applicability

This RG applies to applicants, licensees, and certificate holders authorized under Title 10 of the *Code of Federal Regulations* (10 CFR) Part 70, "Domestic Licensing of Special Nuclear Material" (Ref. 1); 10 CFR Part 71, "Packaging and Transportation of Radioactive Material" (Ref. 2); and 10 CFR Part 72, "Licensing Requirements for the Independent Storage of Spent Nuclear Fuel, High-Level Radioactive Waste, and Reactor-Related Greater Than Class C Waste" (Ref. 3).

This revision is not intended for use by nuclear reactor licensees. The staff has not evaluated the applicability to reactor facilities licensed under 10 CFR Part 50 because these facilities have cores that are designed to operate at criticality, and the facilities have other regulatory requirements that address criticality safety outside the reactor core.

Applicable Regulations

- 10 CFR 70.20, "General license to own special nuclear material," requires a specific license to acquire, deliver, receive, possess, use, transfer, import, or export special nuclear material.
- 10 CFR 70.22(a)(8), "Contents of applications," requires applications for such licenses to include proposed procedures to avoid nuclear criticality accidents.

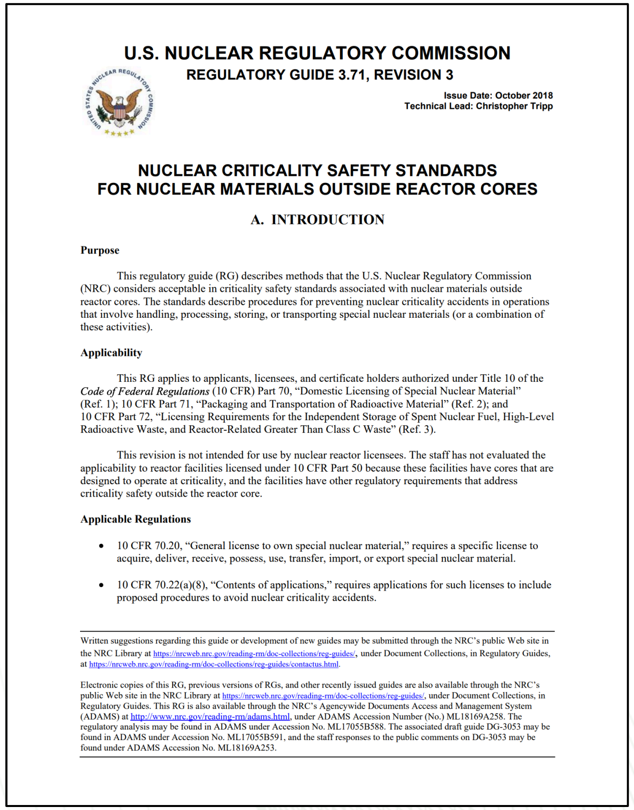
Written suggestions regarding this guide or development of new guides may be submitted through the NRC's public Web site in the NRC Library at https://nrcweb.nrc.gov/reading-rm/doc-collections/reg-guides/, under Document Collections, in Regulatory Guides, at https://nrcweb.nrc.gov/reading-rm/doc-collections/reg-guides/contactus.html.

Electronic copies of this RG, previous versions of RGs, and other recently issued guides are also available through the NRC's public Web site in the NRC Library at https://nrcweb.nrc.gov/reading-rm/doc-collections/reg-guides/, under Document Collections, in Regulatory Guides. This RG is also available through the NRC's Agencywide Documents Access and Management System (ADAMS) at http://www.nrc.gov/reading-rm/adams.html, under ADAMS Accession Number (No.) ML18169A258. The regulatory analysis may be found in ADAMS under Accession No. ML17055B588. The associated draft guide DG-3053 may be found in ADAMS under Accession No. ML17055B591, and the staff responses to the public comments on DG-3053 may be found under ADAMS Accession No. ML18169A253.

Issue – Fissile Concentration Subcritical Limit for $Pu(NO_3)_2$

- NRC did not endorse the fissile concentration subcritical limit in Table 1, ANS-8.1
  - Guidance to NRC licensees was from Regulatory Guide 3.71, Revision 3 (2018)
  - As stated, the NRC requires a licensee to utilize their license-approved NCS validated computational methods to develop their own SCLs or to ensure the ^{240}Pu content, in this case, is conservative relative to the assumptions used in the SCL rather than use the FCSL in the ANS-8.1 standard

Parameter	Subcritical Limit for Fissile Solute $^{239}Pu(NO_3)_4$ [16]
Mass of fissile nuclide (kg)	0.48
Diameter of cylinder of solution (cm)	15.4
Thickness of slab of solution (cm)	5.5
Volume of solution (L)	7.3
Concentration of fissile nuclide (g/L)	7.3
Atomic ratio of hydrogen to fissile nuclide1)	3630
Areal density of fissile nuclide (g/cm^2)	0.25


U.S. NUCLEAR REGULATORY COMMISSION

REGULATORY GUIDE 3.71, REVISION 3

Issue Date: October 2018
Technical Lead: Christopher Tripp

NUCLEAR CRITICALITY SAFETY STANDARDS
FOR NUCLEAR MATERIALS OUTSIDE REACTOR CORES

A. INTRODUCTION

Purpose

This regulatory guide (RG) describes methods that the U.S. Nuclear Regulatory Commission (NRC) considers acceptable in criticality safety standards associated with nuclear materials outside reactor cores. The standards describe procedures for preventing nuclear criticality accidents in operations that involve handling, processing, storing, or transporting special nuclear materials (or a combination of these activities).

Applicability

This RG applies to applicants, licensees, and certificate holders authorized under Title 10 of the *Code of Federal Regulations* (10 CFR) Part 70, "Domestic Licensing of Special Nuclear Material" (Ref. 1); 10 CFR Part 71, "Packaging and Transportation of Radioactive Material" (Ref. 2); and 10 CFR Part 72, "Licensing Requirements for the Independent Storage of Spent Nuclear Fuel, High-Level Radioactive Waste, and Reactor-Related Greater Than Class C Waste" (Ref. 3).

This revision is not intended for use by nuclear reactor licensees. The staff has not evaluated the applicability to reactor facilities licensed under 10 CFR Part 50 because these facilities have cores that are designed to operate at criticality, and the facilities have other regulatory requirements that address criticality safety outside the reactor core.

Applicable Regulations

- 10 CFR 70.20, "General license to own special nuclear material," requires a specific license to acquire, deliver, receive, possess, use, transfer, import, or export special nuclear material.
- 10 CFR 70.22(a)(8), "Contents of applications," requires applications for such licenses to include proposed procedures to avoid nuclear criticality accidents.

Written suggestions regarding this guide or development of new guides may be submitted through the NRC's public Web site in the NRC Library at https://nrcweb.nrc.gov/reading-rm/doc-collections/reg-guides/, under Document Collections, in Regulatory Guides, at https://nrcweb.nrc.gov/reading-rm/doc-collections/reg-guides/contactus.html.

Electronic copies of this RG, previous versions of RGs, and other recently issued guides are also available through the NRC's public Web site in the NRC Library at https://nrcweb.nrc.gov/reading-rm/doc-collections/reg-guides/, under Document Collections, in Regulatory Guides. This RG is also available through the NRC's Agencywide Documents Access and Management System (ADAMS) at http://www.nrc.gov/reading-rm/adams.html, under ADAMS Accession Number (No.) ML18169A258. The regulatory analysis may be found in ADAMS under Accession No. ML17055B588. The associated draft guide DG-3053 may be found in ADAMS under Accession No. ML17055B591, and the staff responses to the public comments on DG-3053 may be found under ADAMS Accession No. ML18169A253.

OAK RIDGE National Laboratory

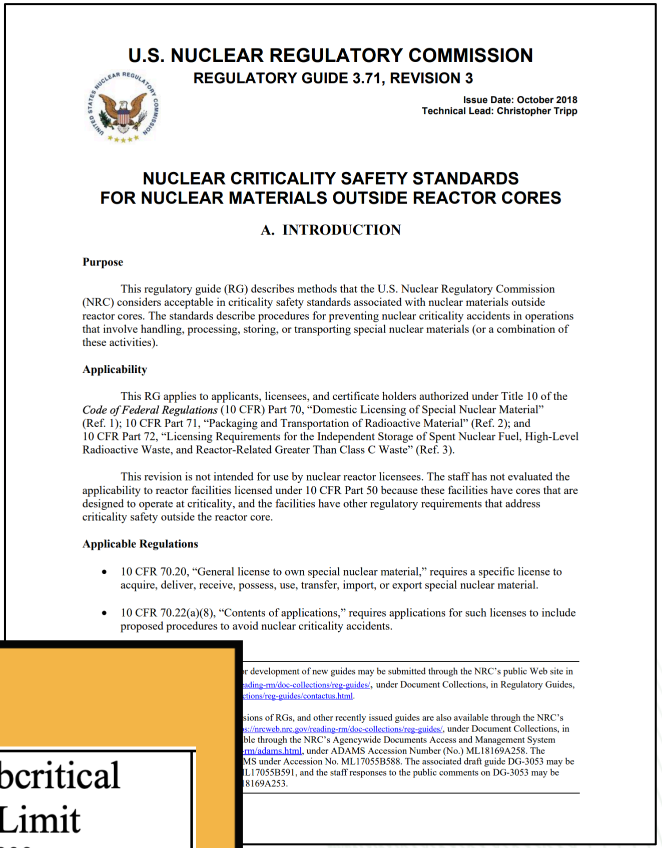
Issue – Fissile Concentration Subcritical Limit for $Pu(NO_3)_2$

- NRC did not endorse the fissile concentration subcritical limit in Table 1, ANS-8.1
  - Guidance to NRC licensees was from Regulatory Guide 3.71, Revision 3 (2018)
  - As stated, the NRC requires a licensee to utilize their license-approved NCS validated computational methods to develop their own SCLs or to ensure the ^{240}Pu content, in this case, is conservative relative to the assumptions used in the SCL rather than use the FCSL in the ANS-8.1 standard

Parameter	Subcritical Limit for Fissile Solute
	$^{239}Pu(NO_3)_4$ [16]
Mass of fissile nuclide (kg)	0.48
Diameter of cylinder of solution (cm)	15.4
Thickness of slab of solution (cm)	5.5
Volume of solution (L)	7.3
Concentration of fissile nuclide (g/L)	7.3
Atomic ratio of hydrogen to fissile nuclide[1)]	3630
Areal density of fissile nuclide (g/cm^2)	0.25

History of the FCSL:

Standard	Year	Subcritical Limit (g ^{239}Pu/L)
ANSI N16.1	1969	7.3
	1975	7.0
ANSI/ANS-8.1	1983	7.3
	1998	7.3
	2014	7.3



U.S. NUCLEAR REGULATORY COMMISSION
REGULATORY GUIDE 3.71, REVISION 3

Issue Date: October 2018
Technical Lead: Christopher Tripp

NUCLEAR CRITICALITY SAFETY STANDARDS
FOR NUCLEAR MATERIALS OUTSIDE REACTOR CORES

A. INTRODUCTION

Purpose

This regulatory guide (RG) describes methods that the U.S. Nuclear Regulatory Commission (NRC) considers acceptable in criticality safety standards associated with nuclear materials outside reactor cores. The standards describe procedures for preventing nuclear criticality accidents in operations that involve handling, processing, storing, or transporting special nuclear materials (or a combination of these activities).

Applicability

This RG applies to applicants, licensees, and certificate holders authorized under Title 10 of the *Code of Federal Regulations* (10 CFR) Part 70, "Domestic Licensing of Special Nuclear Material" (Ref. 1); 10 CFR Part 71, "Packaging and Transportation of Radioactive Material" (Ref. 2); and 10 CFR Part 72, "Licensing Requirements for the Independent Storage of Spent Nuclear Fuel, High-Level Radioactive Waste, and Reactor-Related Greater Than Class C Waste" (Ref. 3).

This revision is not intended for use by nuclear reactor licensees. The staff has not evaluated the applicability to reactor facilities licensed under 10 CFR Part 50 because these facilities have cores that are designed to operate at criticality, and the facilities have other regulatory requirements that address criticality safety outside the reactor core.

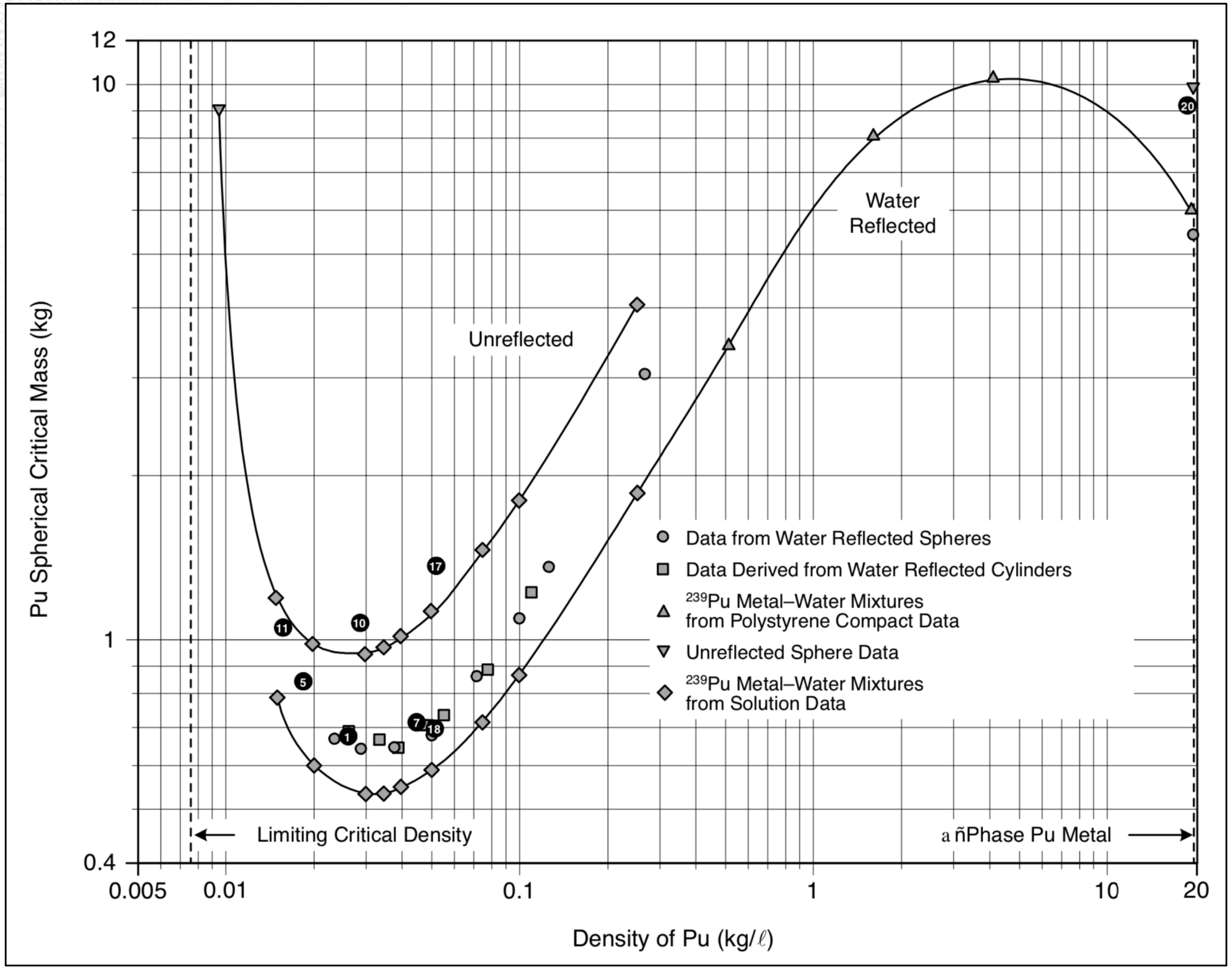
Applicable Regulations

- 10 CFR 70.20, "General license to own special nuclear material," requires a specific license to acquire, deliver, receive, possess, use, transfer, import, or export special nuclear material.
- 10 CFR 70.22(a)(8), "Contents of applications," requires applications for such licenses to include proposed procedures to avoid nuclear criticality accidents.

…r development of new guides may be submitted through the NRC's public Web site in …eading-rm/doc-collections/reg-guides/, under Document Collections, in Regulatory Guides, …ctions/reg-guides/contactus.html.

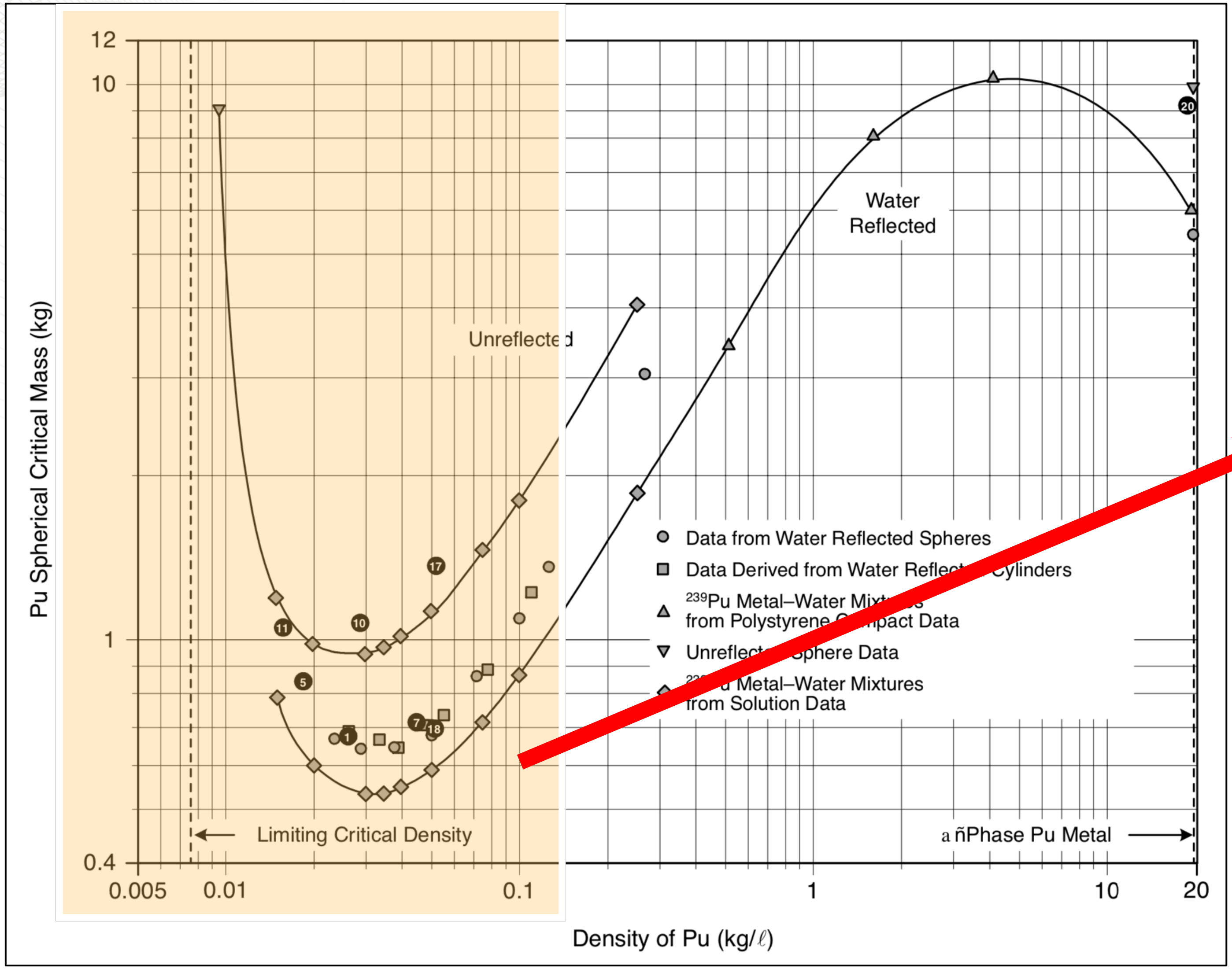
…sions of RGs, and other recently issued guides are also available through the NRC's …s://nrcweb.nrc.gov/reading-rm/doc-collections/reg-guides/, under Document Collections, in …ble through the NRC's Agencywide Documents Access and Management System …-rm/adams.html, under ADAMS Accession Number (No.) ML18169A258. The …MS under Accession No. ML17055B588. The associated draft guide DG-3053 may be …L17055B591, and the staff responses to the public comments on DG-3053 may be …18169A253.

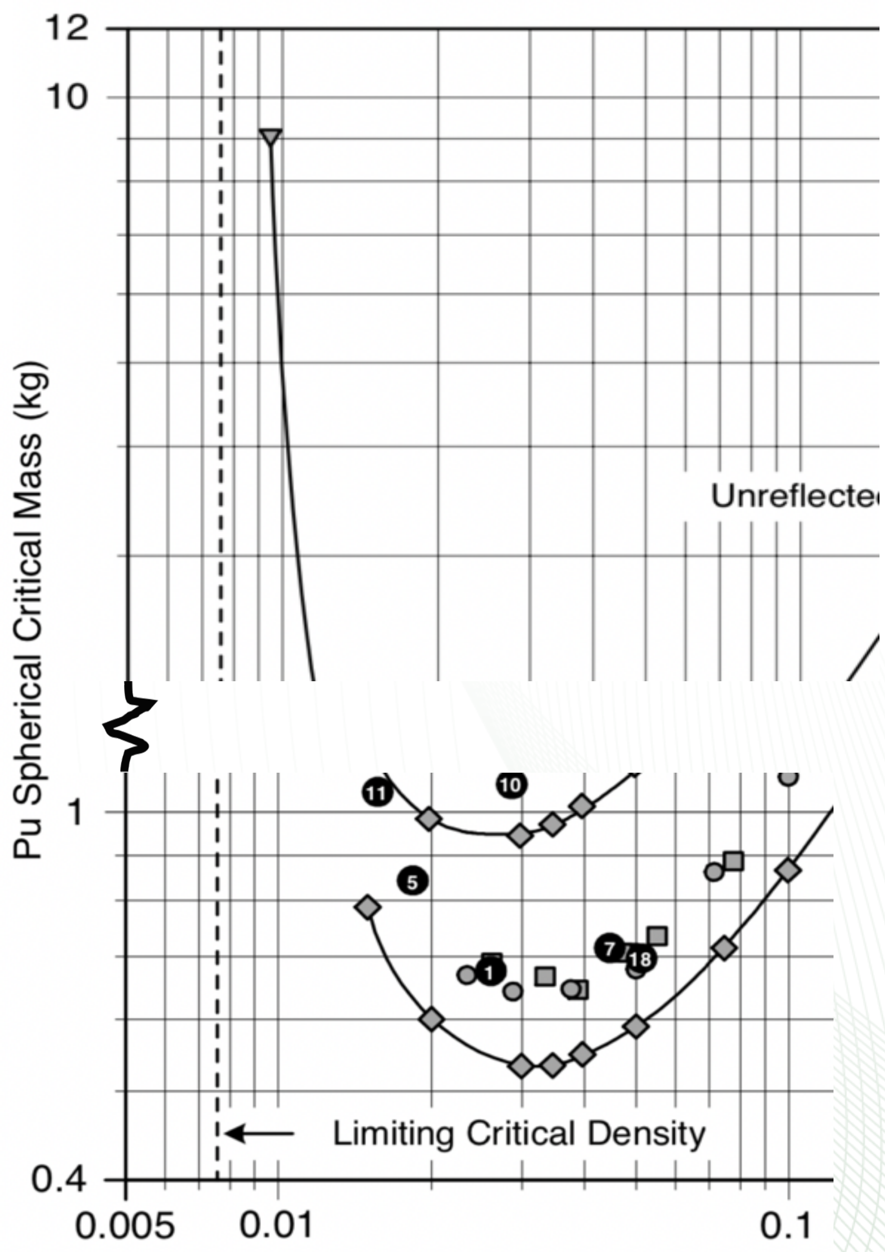
Plutonium Critical Mass Curve (LA-13638)



OAK RIDGE National Laboratory

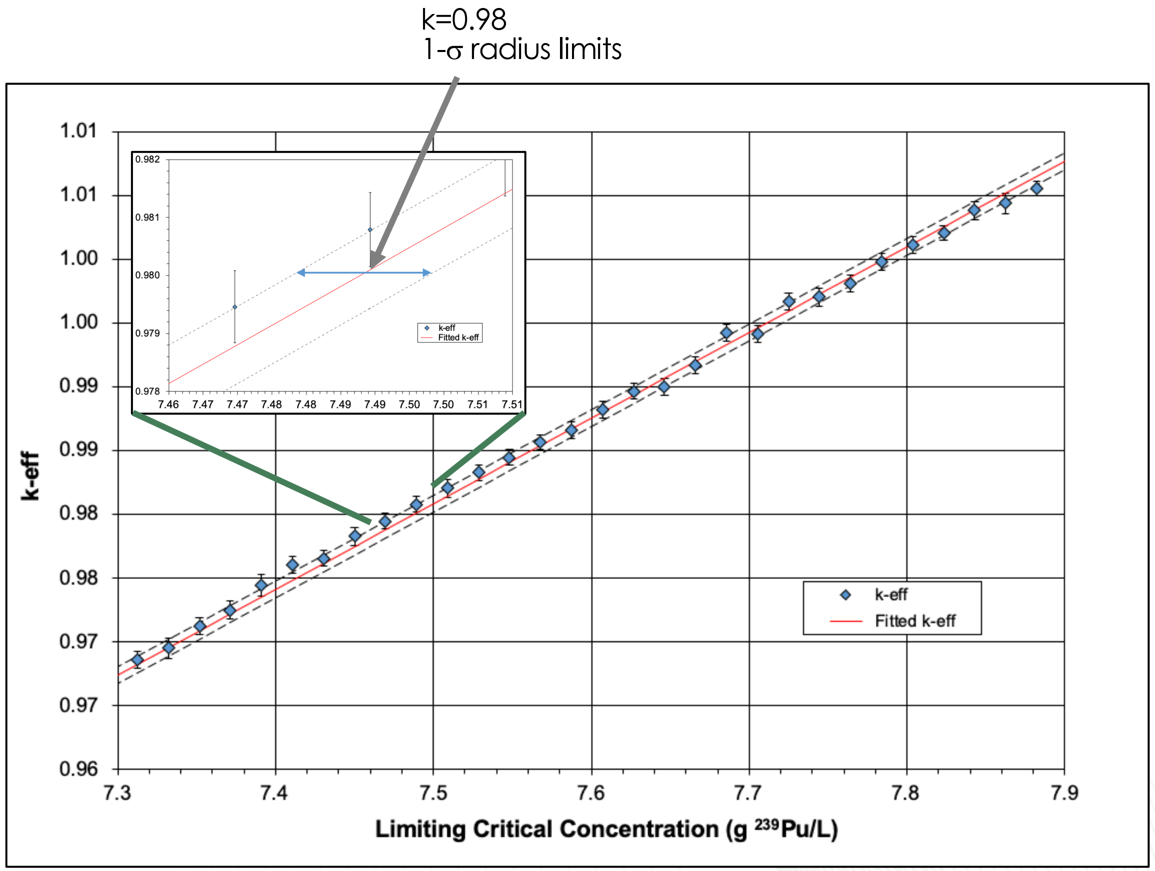
Plutonium Critical Mass Curve





SCL Comparison Calculations

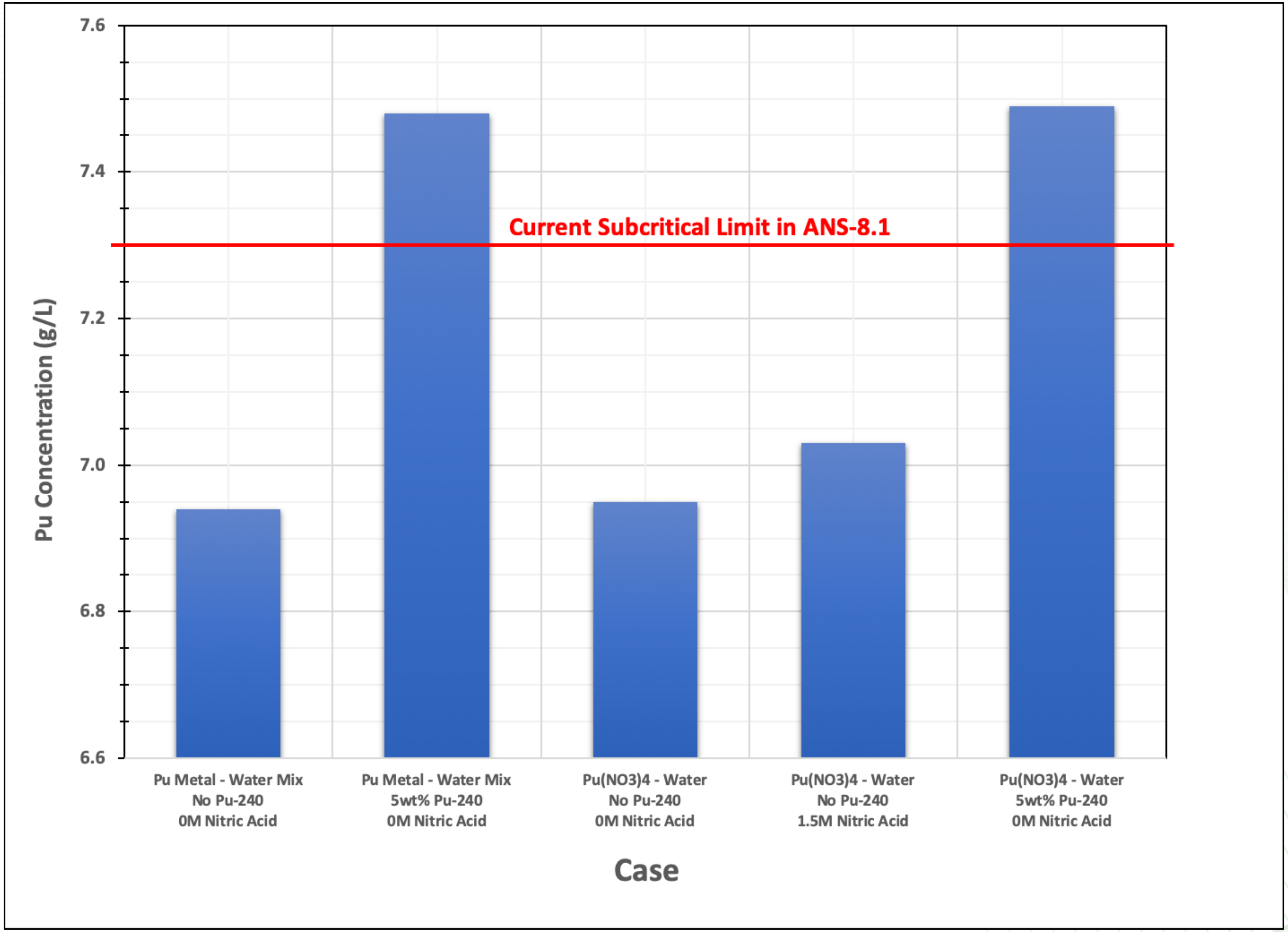
- Both finite and infinite systems were considered
  - Finite
    - Spherical, fully water-reflected, configurations with volumes from 100 L to 1.0E+9 L over a range of Pu concentrations
    - Smaller volumes (100 L to 1000 L systems) are similar to the volumes used in experimental and process facilities, i.e., more realistic configurations, that can be compared to experimental data
  - Infinite
    - Mirror-reflected cuboid configuration over a range of Pu concentration values
- Calculations performed using SCALE version 6.2 (Keno V.a) using ENDF/B-VII.1 cross sections
- The methodology developed for new ANS-8.1 SCL work was used for these calculations
  - Developed by Argonne (Lell, Morman) circa 2001
  - A computational method validation that meets the requirements of ANSI/ANS-8.24-2017 was not performed for this comparison study



- It is generally understood that a 2% keff computational margin was assumed for most, if not all, SCLs in ANS-8.1
- These calculations reported the SCL results corresponding to a k_{eff} of 0.98

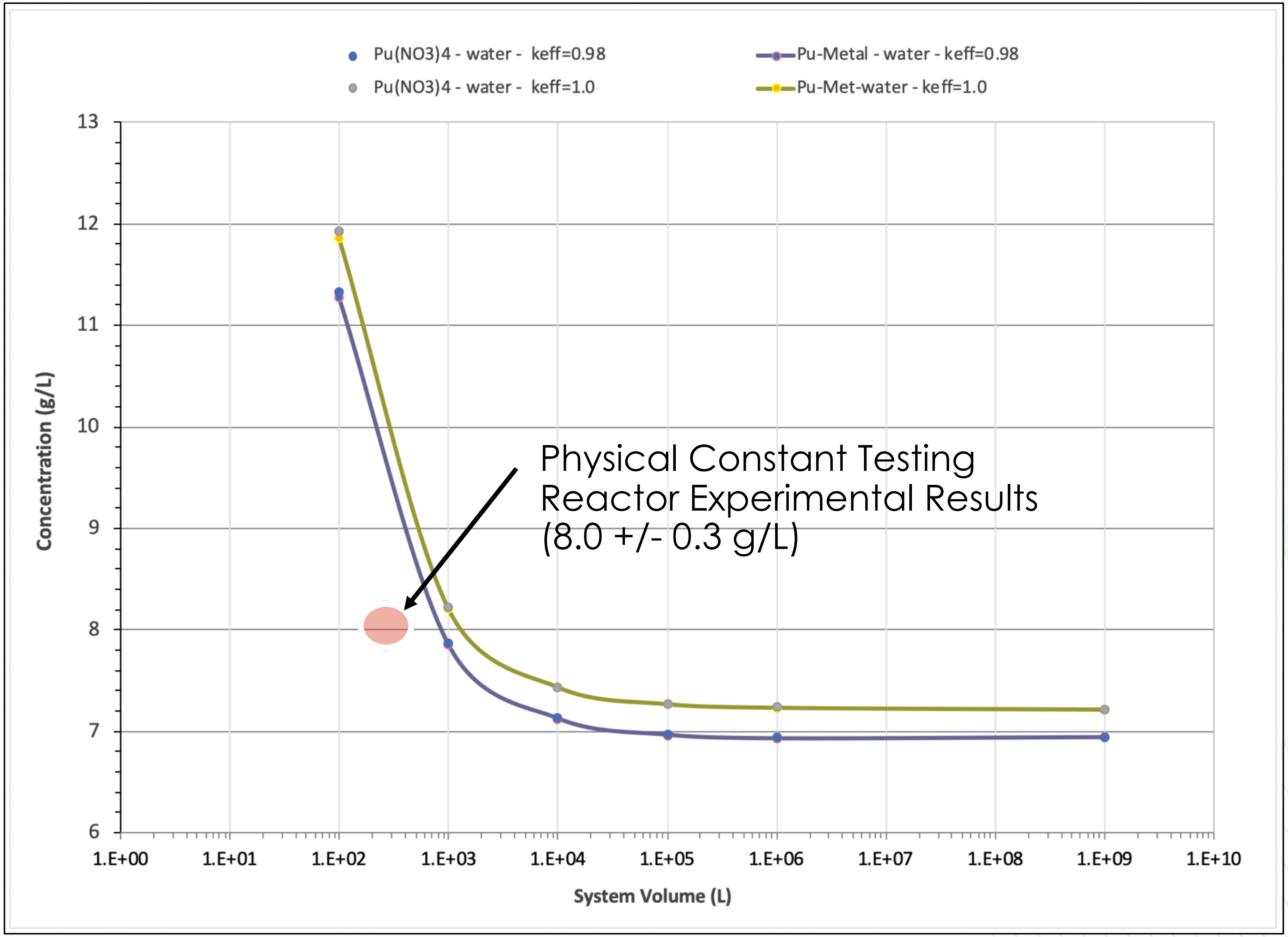
OAK RIDGE National Laboratory

SCL Comparison Calculation Mean Results – Infinite Case Results

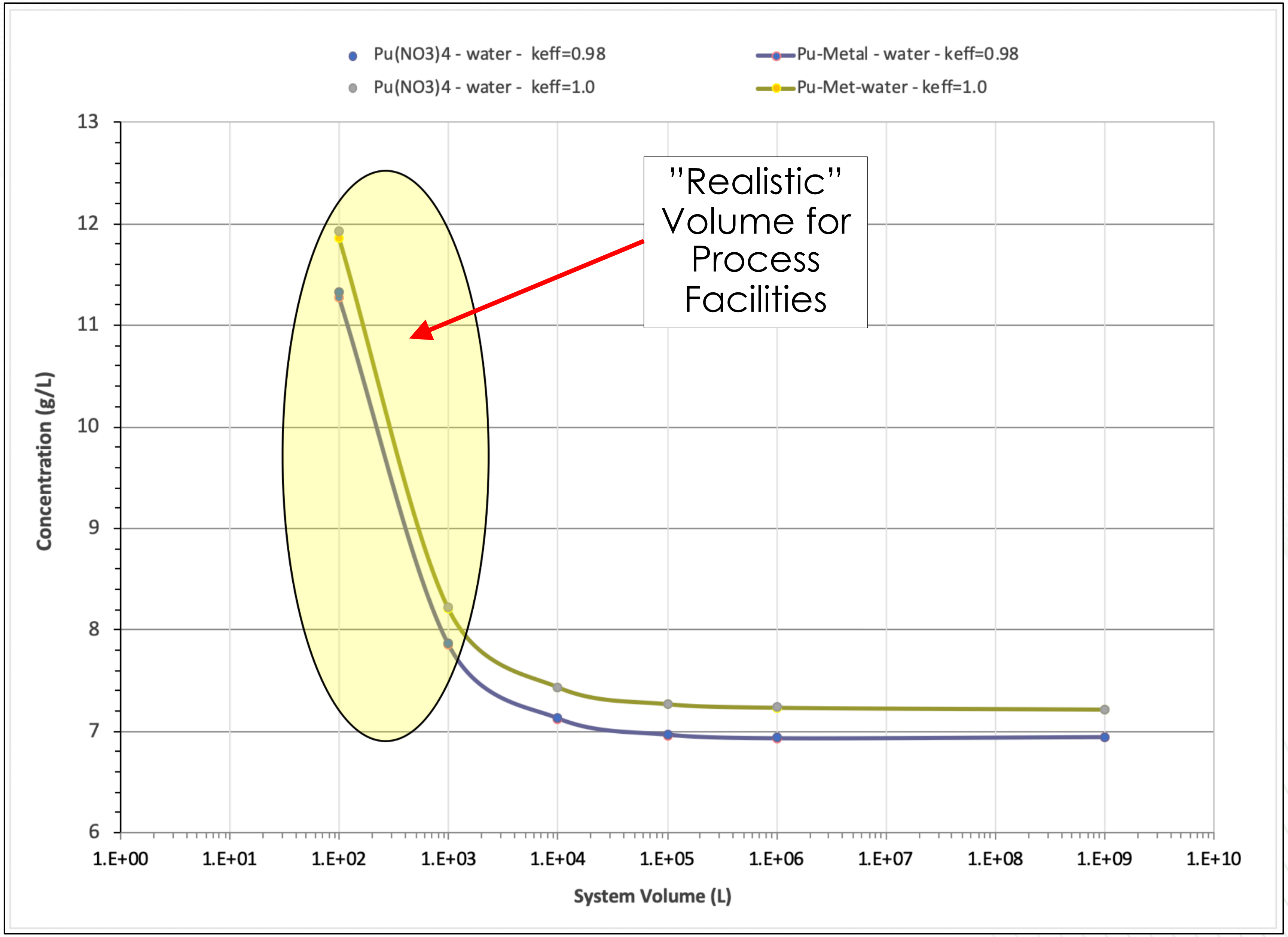


OAK RIDGE National Laboratory

SCL Comparison Calculation Mean Results – Finite Case Results



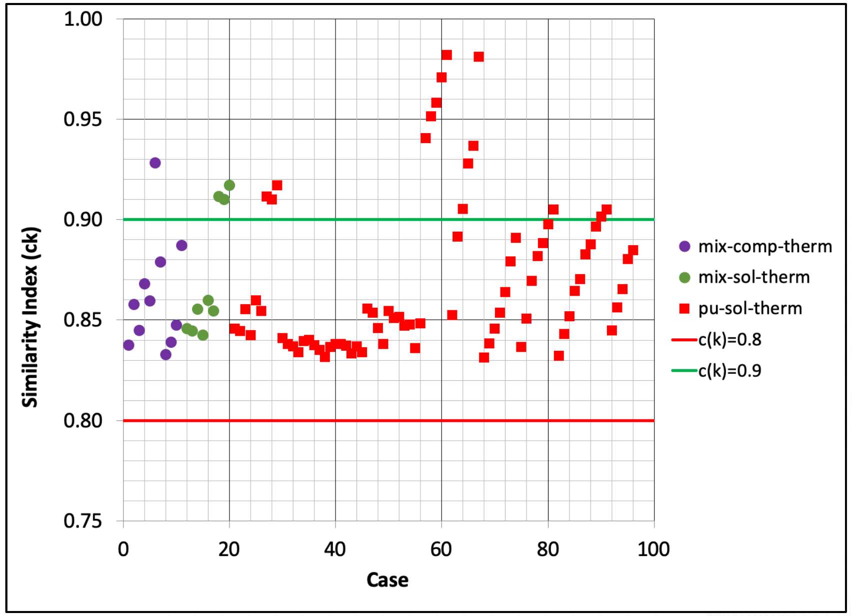
SCL Comparison Calculation Mean Results – Finite Case Results



Similarity Assessment & USL Results – Whisper/TSUNAMI

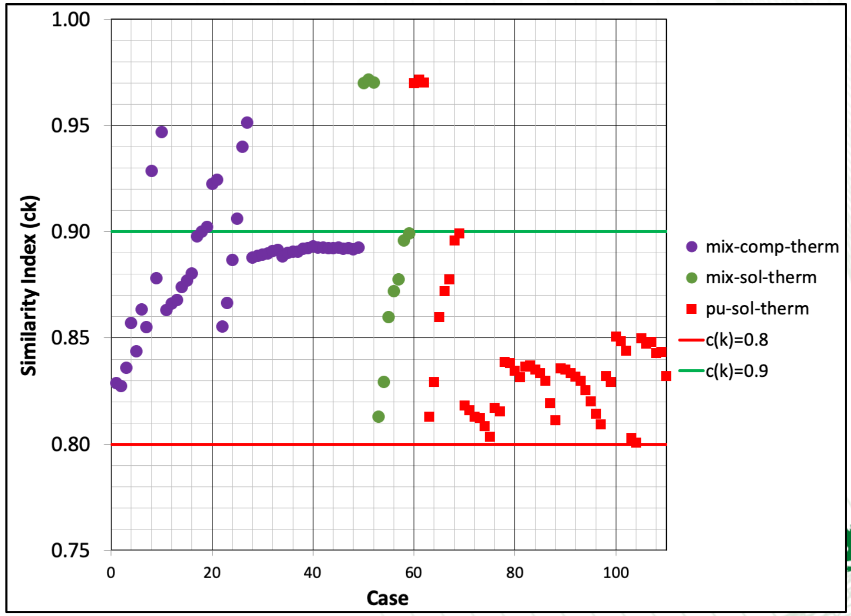
- Whisper Results
  - 96 experiments were identified
  - Mix-Comp-Therm experiments
    - (11 total, 1 exp. with c_k > 0.9)
  - Mix-Sol-Therm experiments
    - (9, 3 exp. with c_k > 0.9)
  - Pu-Sol-Therm experiments
    - (76, 17 exp. with c_k > 0.9)
  - Calculated USL of 0.97674
- TSUNAMI(CE) Results
  - 110 experiments were identified
  - Mix-Comp-Therm experiments
    - (49 total, 9 exp. with c_k > 0.9)
  - Mix-Sol-Therm experiments
    - (10, 3 exp. with c_k > 0.9)
  - Pu-Sol-Therm experiments
    - (61, 3 exp. with c_k > 0.9)
  - Calculated USL of 0.9669

Whisper



44-group covariance data (ENDF/B-VII.1 library) used for all calculations

TSUNAMI



Conclusions

- The ANS-8.1 SCLs were introduced 50 years ago
  - The SCLs have been introduced and modified as necessary to support NCS in operations with fissionable materials outside of reactors
- Currently, the NRC does not endorse the ANS-8.1 FCSL because of concerns about the bases for the computational margin
  - The use of radiation transport codes is allowed for licensees to perform validated computations to generate SCLs or to ensure the ^{240}Pu content, in this case, is conservative relative to the assumptions used in the SCL
- Comparison calculations suggest the current SCL for FCSL may be too large at 7.3 g ^{239}Pu/L
  - Experimental data are sparse at this concentration
  - TSUNAMI/Whisper results were provided that indicate a USL lower than that assumed by ANS-8.1 lore
- The ANS-8.1 working group is currently revising the standard and may consider revising this SCL based on a combination of available experimental data and additional SCL calculations