Justification to Disregard U-234 Mass in Certain Uranium Systems

Presented at ANS NCSD 2013 Topical Meeting

October 1, 2013

Andrew Wysong

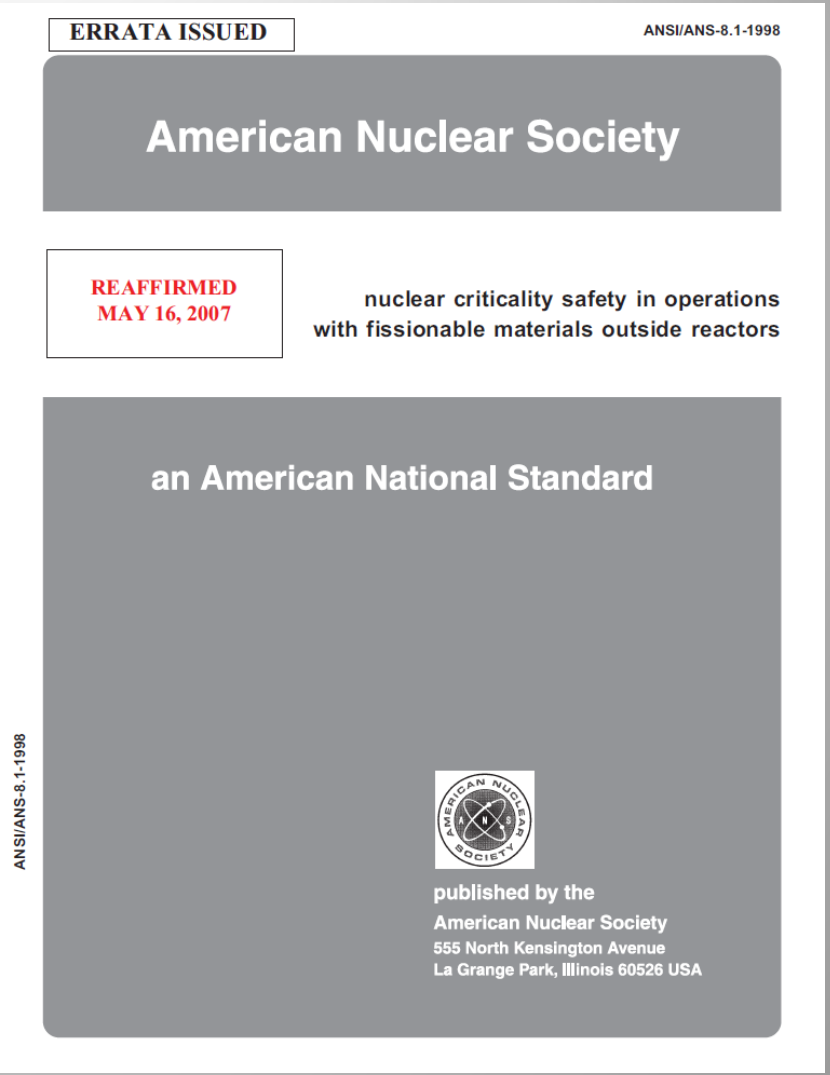
Lawrence Livermore National Laboratory

LLNL-PRES-644185

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344. Lawrence Livermore National Security, LLC

Introduction

- ANSI/ANS-8.1-1998
  - ^{235}U limits apply to mixtures "with ^{234}U, ^{236}U, or ^{238}U, provided that ^{234}U is considered to be ^{235}U" in computing mass
  - Goal is to show an improved way of handling the other common uranium isotopes when setting ^{235}U limits



Background Information

- Published K-Infinity Values*
  - ^{235}U – 2.28
  - ^{234}U – 1.52
  - ^{236}U – 0.74
  - ^{238}U – 0.34

*Lavarenne C. et all, "CSAS1X/SCALE 4.4 Calculation Using ENDF/B-5 238 Group Library as Reported in Table 4 of SEC/T/03.146," *Evaluation of Nuclear Criticality Safety Data and Limits for Actinides in Transport*, IRSN, Fontenay-Aux-Roses, France, May 2003

Argument

- In uranium enriched up to 94% ^{235}U by weight there exist bounding ratios of ^{234}U, ^{236}U, and ^{238}U due to the enrichment process.
- In these systems, using the ratios mentioned above, it is shown that the negative reactivity effects of ^{236}U and ^{238}U always outweigh the positive reactivity effects from ^{234}U.

Methodology

- Calculations with MCNP5
  - Using ENDF/B-VII cross sections
  - Benchmarking
    - 709 HEU cases from ICSBEP*
    - Scherzo-556 International Standard**
      - Calculated keff = 0.99999 ± 0.001
    - Comparison with Published K-Infinity Values

Isotope	Calculated k_∞	Published k_∞'s
^{234}U	1.70139 ± 0.00208	1.52
^{236}U	0.71249 ± 0.00208	0.74
^{238}U	0.30835 ± 0.00127	0.34

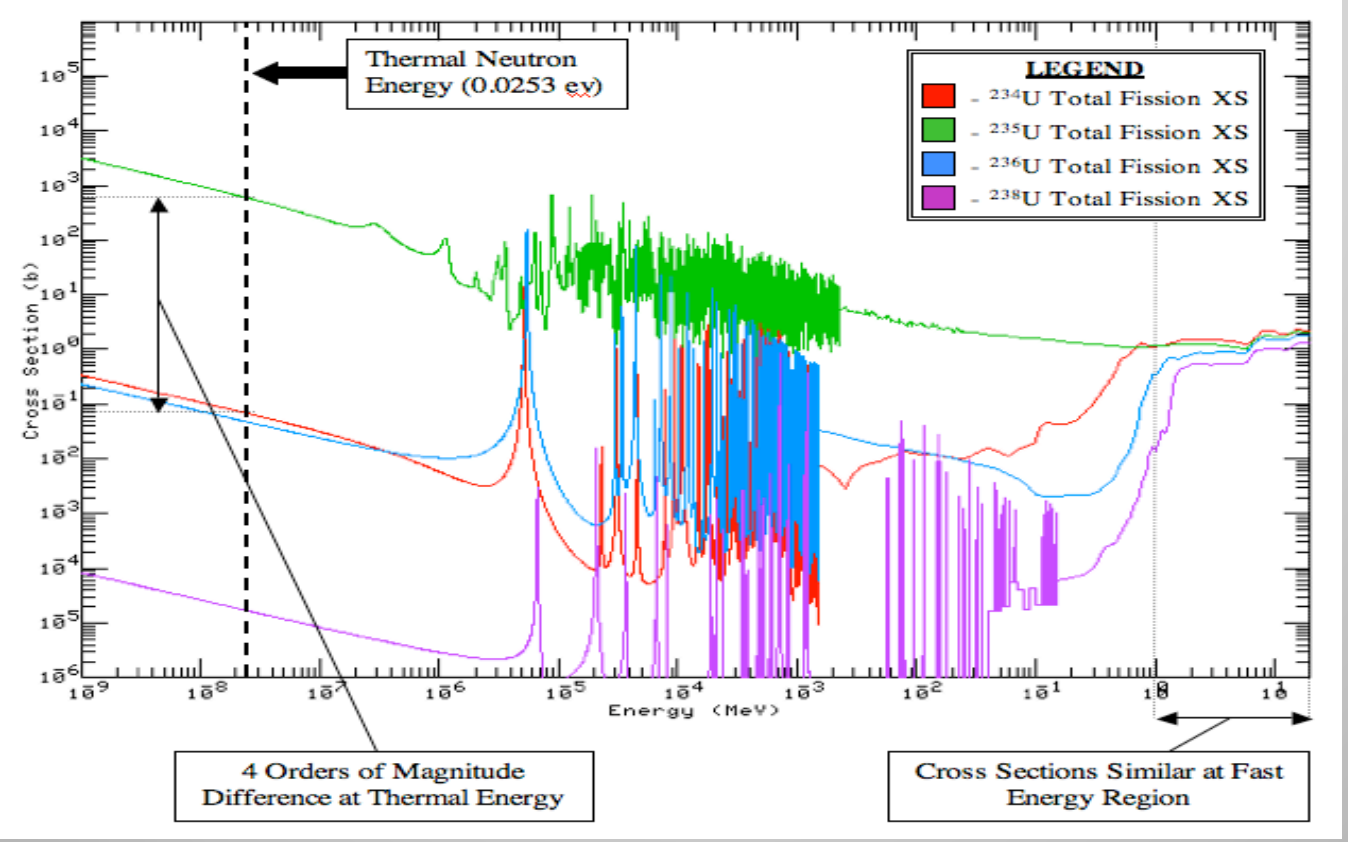
**International Handbook of Evaluated Criticality Safety Benchmark Experiments*, NEA/NSC/DOC(95)03, Organization for Economic Cooperation and Development, Nuclear Energy Agency, Paris, France, September 2007 edition

**Darrouzet, M., Chaudat, J. P., Fischer, E. A., Ingram, G., Sanders, J. E., Scholtyssek, W., "Studies of Unit Kinf Lattices in Metallic Uranium Assemblies Zebra 8H Sneak 8, Ermine, and Harmonie UK," *Proceedings of an International Symposium on Physics of Fast Reactors*, Tokyo, October 16-19, 1973, Committee for the International Symposium on Physics of Fast Reactors, A28, 1973

Methodology

- Benchmark Issues
  - ^{234}U data 11.9% different from Published Data
    - Calculated with ENDF/B-VII versus ENDF/B-V
    - Any differences are exaggerated by extreme geometry
      - Modeled in infinite system
  - No published benchmarks involving significant quantities of separated ^{234}U

Analysis – Fission Cross Sections



- Energy range of interest is fast neutrons
  - ^{235}U dominates at all energies except fast neutrons
  - Investigate fast energy range where ^{234}U, ^{236}U, and ^{238}U have cross sections on the same order of magnitude
  - In a thermal or epithermal system reactivity effects from ^{234}U will be smaller and bounded by a fast system.

Analysis – Isotopic Compositions

- Examined 16 Uranium Compositions

Reference	Isotopic Weight Percentage			
	^{234}U	^{235}U	^{236}U	^{238}U
J. Korean Nucl. Soc. V14, #3, p103 (1982).	0.0015	0.2360	0.0053	99.7572
Chart of the Nuclides	0.0056	0.7110	0.00	99.2834
ORNL-CDC-5	0.011	2.140	0.017	97.832
ORNL-CDC-5	0.017	3.043	0.012	96.928
ORNL-2968	0.02	4.89	-	95.09
JBB-05-94	0.026	4.946	0.050	94.978
ORNL-TM-1195	0.03	4.98	0.04	94.95
NSE, v72, p230 (1979)	0.1035	10.0591	-	89.8374
AHSB (RP) R 58	0.13	20.00	0.08	79.79
ORNL-2968	0.2	37.5	0.2	62.1
AWRE NR 1/66	0.70	37.51	0.18	61.62
AERE R/R 2703	0.74	45.01	-	54.24
ORNL/ENG-2	0.42	62.40	0.29	36.89
AHSB(RP) R 58	0.87	90.00	0.17	8.96
TrANS, v14, #2 (1971)	0.97	93.206	0.24	5.584
LA-1614	1.05	93.71	--	5.24
Bounding Reactivity Weight % Limit	**1.20**	**94.00**	**0.30**	**4.50**

Analysis – Isotopic Compositions

- Determine the ratios of non ^{235}U isotopes for fresh uranium:

Isotopes	^{234}U	^{236}U	^{238}U
Bounding Reactivity Weight % Limit	1.20%	0.30%	4.50%
Ratio Integer	4.00	1.00	15.00

  - Enrichments lower than 94% are bound due to the lower mass fraction of ^{234}U and ^{235}U

*Pellarin, D.J., Jarriel, J.L., "Pulsed Reactivity Measurements of Large ^{235}U-Al Castings in H_2O," *Transactions of the American Nuclear Society*, 27, 413-415 (1977).

Analysis – Isotopic Compositions

- In reprocessed fuel the ^{234}U mass percentage becomes smaller due to reduced ^{233}U content

Reference	Isotopic Weight Percentage			
	^{234}U	^{235}U	^{236}U	^{238}U
TrANS, v27, p414 (1977)*	1.00	76.97	10.5	11.49

- Determine the ratios of non ^{235}U isotopes for reprocessed uranium:

Isotopes	^{234}U	^{236}U	^{238}U
Ratio Integer	1.00	11.00	11.00

*Pellarin, D.J., Jarriel, J.L., "Pulsed Reactivity Measurements of Large ^{235}U-Al Castings in H_2O," *Transactions of the American Nuclear Society*, 27, 413-415 (1977).

Analysis – Isotopic Ratio Assertion 1

- Fresh Uranium Ratios:

Isotopes	^{234}U	^{236}U	^{238}U
Ratio Integer	4.00	1.00	15.00

- Reprocessed Uranium Ratios:

Isotopes	^{234}U	^{236}U	^{238}U
Ratio Integer	1.00	11.00	11.00

- Assertion #1
  - Maximum weight fraction of ^{234}U among the lesser U isotopes (^{234}U, ^{236}U, ^{238}U) is 0.20 for uranium enriched up to 94%

$$^{234}\text{U Weight Fraction} = \frac{^{234}U}{^{234}U + ^{236}U + ^{238}U} = \frac{4.00}{4.00 + 1.00 + 15.00} = 0.20$$

*Pellarin, D.J., Jarriel, J.L., "Pulsed Reactivity Measurements of Large ^{235}U-Al Castings in H_2O," *Transactions of the American Nuclear Society*, 27, 413-415 (1977).

Analysis – Isotopic Ratio Assertion 2

- Fresh Uranium Ratios:

Isotopes	^{234}U	^{236}U	^{238}U
Ratio Integer	4.00	1.00	15.00

- Reprocessed Uranium Ratios:

Isotopes	^{234}U	^{236}U	^{238}U
Ratio Integer	1.00	11.00	11.00

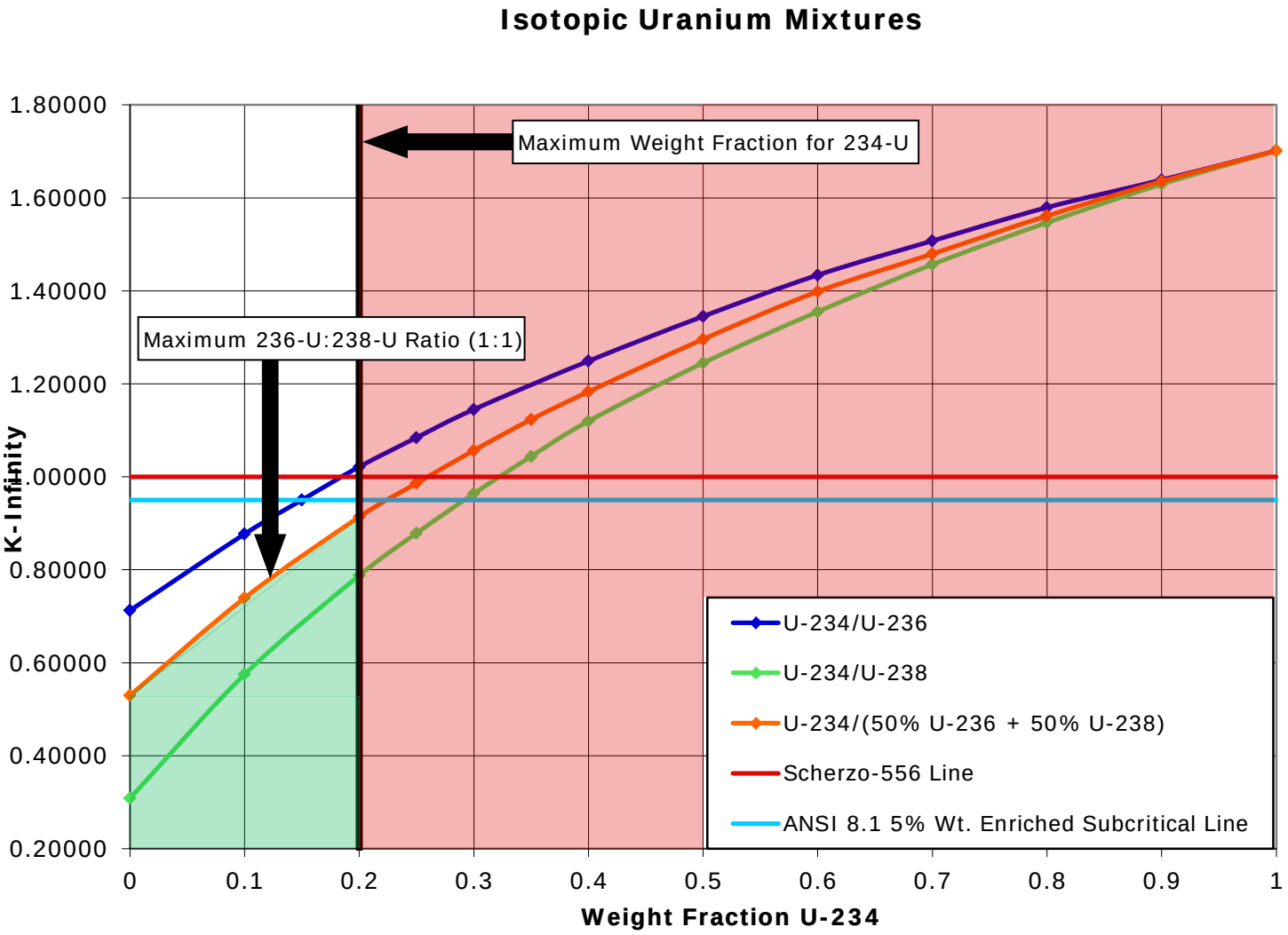
- Assertion #2
  - Minimum ratio of ^{238}U to ^{236}U is 1:1 for uranium enriched up to 94%

*Pellarin, D.J., Jarriel, J.L., "Pulsed Reactivity Measurements of Large ^{235}U-Al Castings in H_2O," *Transactions of the American Nuclear Society*, 27, 413-415 (1977).

Analysis – Bounding Model

- Infinite Homogenous Uranium System
  - U Density 18.81 g/cm^3 (ANSI/ANS-8.1)
  - Three Isotopic Mixtures
    - ^{234}U mixed into ^{236}U
      - Weight fraction from 0.0 to 1.0 of ^{234}U
    - ^{234}U mixed into ^{238}U
      - Weight fraction from 0.0 to 1.0 of ^{234}U
    - ^{234}U mixed into ratio of ^{238}U:^{236}U of 1:1
      - Weight fraction from 0.0 to 1.0 of ^{234}U

Results – Bounding Model



Results – Bounding Model

- Vertical Line at X=0.20
  - Marks the maximum weight fraction of ^{234}U at 0.20 (Assertion 1)
- Orange Plotted Line
  - Data points for 1:1 ratio of ^{236}U and ^{238}U (Assertion 2) mixed with ^{234}U
- Established subcritical limit using ANSI/ANS-8.1* 5% enriched ^{235}U
  - Modeled in infinite geometry – resulting k_{eff} = 0.95006 ± 0.00110
- At max weight fraction of ^{234}U mixed with 1:1 ratio ^{236}U and ^{238}U
  - k_{∞} = 0.91367 ± 0.00208
  - Value less than subcritical limit (ANSI/ANS-8.1-1998[1] 5% ^{235}U line)
- Illustrates that for any uranium enrichment level up to 94% ^{235}U by weight ^{234}U **does not** need to be considered as the mixture $k_{\infty} < 1.0$

**American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors*, ANSI/ANS-8.1-1998, American Nuclear Society, LaGrange Park, IL, September 9, 1998

Conclusion

- The analysis in this evaluation
  - Proves that bounding isotopic ratios for ^{234}U, ^{236}U, and ^{238}U exist in enrichment up to 94% ^{235}U
  - These ratios effectively bound any reactivity increase from ^{234}U
  - Thus mass limits only need to be placed on ^{235}U

References

1. *American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors*, ANSI/ANS-8.1-1998, American Nuclear Society, LaGrange Park, IL, September 9, 1998.

2. Chou, S. P., "Verification of the Installation of SCALE5, MCNP5 Versions 1.20 and 1.30, and MCNP4C2," CSAM06-031, Nuclear Criticality Safety Division, LLNL, Livermore, CA, February 14, 2006.

3. Krass, A. K., "Validation of MCNP5 (Version 1.30) on Surya for General Application to Highly Enriched Uranium Systems," CSM 1512, Nuclear Criticality Safety Division, LLNL, Livermore, CA, March 4, 2008.

4. *International Handbook of Evaluated Criticality Safety Benchmark Experiments*, NEA/NSC/DOC(95)03, Organization for Economic Cooperation and Development, Nuclear Energy Agency, Paris, France, September 2007 edition.

5. Darrouzet, M., Chaudat, J. P., Fischer, E. A., Ingram, G., Sanders, J. E., Scholtyssek, W., "Studies of Unit Kinf Lattices in Metallic Uranium Assemblies Zebra 8H Sneak 8, Ermine, and Harmonie UK," *Proceedings of an International Symposium on Physics of Fast Reactors*, Tokyo, October 16-19, 1973, Committee for the International Symposium on Physics of Fast Reactors, A28, 1973.

6. Lavarenne C. et all, "CSAS1X/SCALE 4.4 Calculation Using ENDF/B-5 238 Group Library as Reported in Table 4 of SEC/T/03.146," *Evaluation of Nuclear Criticality Safety Data and Limits for Actinides in Transport*, IRSN, Fontenay-Aux-Roses, France, May 2003.

7. Heinrichs, D. P., "Typical Uranium Isotopics," CSAM-97-261, Criticality Safety Group, LLNL, Livermore, CA, October 1, 1997.

8. Pellarin, D.J., Jarriel, J.L., "Pulsed Reactivity Measurements of Large ^{235}U-Al Castings in H_2O," *Transactions of the American Nuclear Society*, 27, 413-415 (1977).

9. Mosteller, R. D., "Water-Reflected, Highly Enriched Uranium Sphere," HEU-MET-FAST-004, International Handbook of Evaluated Criticality Safety Benchmark Experiments, NEA Nuclear Science Committee, Idaho Falls, ID, September, 2007.

10. Thomas, J. T., "Critical Experiments with UF_6 Cylinder Model 8A Containers," Union Carbide Corporation, Y-12 Plate Report Y-DR-128, Oak Ridge, TN, September 1974.