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JUSTIFICATION TO DISREGARD U-234 MASS IN CERTAIN URANIUM SYSTEMS

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ABSTRACT

The ANSI/ANS-8.1-1998;R2007 standard states ^{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. Analysis of historical data and calculations coupled with benchmark data show a superior way of handling the other common uranium isotopes when setting limits for a certain ^{235}U systems. Currently ^{234}U mass is required to be considered as ^{235}U mass in order to account for the fact ^{234}U has a k_∞ = 1.70 meaning criticality can be reached whereas ^{236}U and ^{238}U have k_∞'s equal to 0.71 and 0.31 meaning they remain subcritical in infinite amounts. Multiple isotopic compositions of enriched uranium from published experiments and reference documents were analyzed. Results of the analysis indicate that in uranium enriched to 94% ^{235}U by weight, bounding ratios of ^{234}U, ^{236}U, and ^{238}U exist due to the enrichment process. The established material ratios are utilized in Monte Carlo calculations of three different isotopic parameters in infinite metal systems. The results indicate that a subcritical k_∞ value will always be achieved given the isotopic ratios determined by analysis of historical sample data. The subcritical value is due to the ^{236}U and ^{238}U isotopes having a greater negative effect on reactivity than the increase from an increased fission rate in ^{234}U. The conclusion is that for uranium systems with enrichment below 94% by weight the ^{234}U mass can be ignored due to the associated dilution effect from the ^{236}U and ^{238}U isotopes.

Key Words: Uranium-234, ANS-8.1, Isotopics, k-infinity

1 INTRODUCTION

The ANSI/ANS-8.1-1998 Standard[1] regarding nuclear criticality safety in operations with fissionable material outside reactors states certain guidelines for dealing with uranium isotopes when setting mass limits. In ANSI/ANS-8.1-1998[1] ^{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. In this evaluation calculations coupled with benchmark data show a superior way of handling the other common uranium isotopes when setting limits for ^{235}U.

Currently ^{234}U mass must be considered as ^{235}U mass in order to account for the fact ^{234}U has a k_∞ = 1.70 meaning criticality can be reached with a finite amount of ^{234}U mass. ^{236}U and ^{238}U have k_∞'s equal to 0.71 and 0.31 meaning they remain subcritical in infinite amounts and thus no mass limits are required. In uranium enriched to 94% ^{235}U by weight, bounding ratios of ^{234}U, ^{236}U, and ^{238}U exist due to the enrichment process. Utilizing these ratios it is shown that ^{234}U accounts for less than 0.20 of the total mass fraction of ^{234}U, ^{236}U, and ^{238}U and the remaining 0.80 mass fraction is in a ratio of at least 1:1 for ^{238}U:^{236}U. Creating models conservatively based on these two material limits, the resulting $k_\infty = 0.91367 \pm 0.00208$ suggests a mass limit for ^{234}U is no longer required.

2 METHODOLOGY

All calculations documented in this report were performed using MCNP5 Version 1.30 on SURYA Sun workstation. The SURYA workstation is running on Sun Solaris 5.8 operating system, and MCNP5 was compiled with Sun Workshop 6 Update 2 C and Sun Workshop 6 Update 2 F90 Compilers. Verification of the installation of MCNP5 has been previously documented[2].

The uranium isotopes used in the MCNP5 calculations employed the ENDF/B-VII neutron cross-section data at 300 K. Extensive uranium benchmarking has been completed for MCNP5 on the SURYA workstation at LLNL[3] and includes 709 cases taken from the *International Handbook of Evaluated Criticality Safety Benchmark Experiments*.[4] The total uncertainty determined from the combination of the bias estimate and the average of the combined uncertainties of the individual benchmark results from the benchmark set is 0.087.

A calculation using the Scherzo-556 International Standard[5] acted as a more specific benchmark for the code. The Scherzo-556 International Standard describes the required enrichment (5.56 at% ^{235}U) for an infinite amount of uranium to be exactly critical (k_{eff} = 1.000). Building an infinite model in MCNP5 Version 1.30 on Surya and executing the program resulted in a value of $k_{eff} = 0.99999 \pm 0.00104$. The tiny difference is statistically insignificant and the matching values act as a successful benchmark for application of the code in this evaluation.

Another specific benchmark is the comparison of calculated k_∞ values with published k_∞ values from other published sources. Table I lists these values and their sources.

Table I. Calculated and Published k_∞ Values

Isotope	Calculated k_∞	Published k_∞'s
^{234}U	1.70139 ± 0.00208	1.52[6]
^{236}U	0.71249 ± 0.00208	0.74[6]
^{238}U	0.30835 ± 0.00127	0.34[6]

As evidenced by Table I, the calculations done using MCNP5 Version 1.30 on the Surya workstation are in agreement for ^{236}U and ^{238}U with the calculations from other sources. However, the value for ^{234}U differs by 11.9%. The published of k_∞=1.52[6] for ^{234}U was calculated using ENDF/B-V. The difference in calculated k_∞ of ^{234}U is likely due to changes (additional resonances, etc.) in the updated cross section library ENDF/B-VII. These differences are exaggerated by the extreme geometry of and infinite system. Unfortunately, there are no published benchmarks involving significant quantities of separated ^{234}U that can be applied for this evaluation.

3 ANALYSIS

Since the analysis focuses on fission characteristics of different uranium isotopes, Figure 1 shows a plot of the total fission cross sections for ^{234}U, ^{235}U, ^{236}U, and ^{238}U. Notice that the only energy range not completely dominated by the ^{235}U isotope is the fast energy range. Thus, it is only necessary to model a fast metal system because it is in the fast energy region where ^{234}U,

^{236}U, and ^{238}U have cross sections on the same order of magnitude as ^{235}U. In a system with moderating material and thus thermal neutrons, any reactivity effects from ^{234}U will be smaller and thus bounded by those observed in a fast metal system.

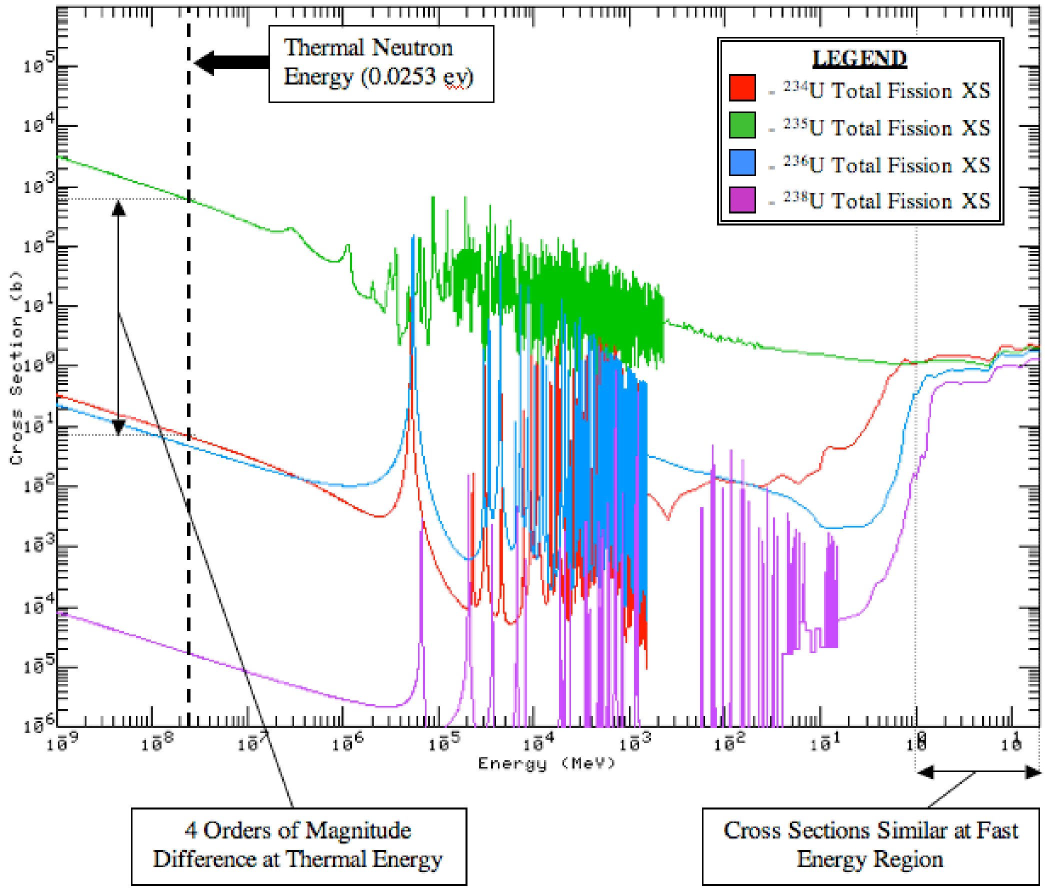

Figure 1. Total Fission Cross Section for ^{234}U, ^{235}U, ^{236}U, and ^{238}U

In order to bound ^{234}U using the mass of ^{236}U and ^{238}U, a bounding isotopic composition for fresh uranium (un-irradiated or reprocessed) enriched up to 94% ^{235}U by weight, must be established. Using data from CSAM97-267[7] *Typical Uranium Isotopics* Table II was created.

Table II. Isotopic Composition of Uranium

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**

Now excluding ^{235}U, we determine ratios between ^{234}U, ^{236}U, and ^{238}U, which are shown in Table III.

Table III. Ratios of Non ^{235}U Isotopes

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% ^{235}U by weight are bound and covered in this analysis due to the lower mass fraction of ^{234}U and ^{235}U, which is illustrated by Table II.

For uranium that has been irradiated and reprocessed, the isotopic compositions and ratios listed in Table II and III are not valid. In reprocessed fuel, there is an elevated isotopic percentage of ^{236}U due to neutron absorption without fission in the ^{235}U atom. This changes the ratio of ^{238}U to ^{236}U to values that often approach 1:1.

The ^{234}U mass percentage becomes smaller in reprocessed fuel because ^{233}U is present in such small amounts that negligible amounts of ^{233}U atoms absorb neutrons without fission and thus become ^{234}U. An isotopic composition taken from the Transactions of the American Nuclear Society Vol. 27[8] "Pulsed Reactivity Measurements of Large ^{235}U-Al Castings in H_2O is shown in Table IV.

Table IV. Isotopic Composition of Reprocessed Uranium

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

Now excluding ^{235}U, we determine ratios between ^{234}U, ^{236}U, and ^{238}U, which are shown in Table V.

Table V. Ratios of Non ^{235}U Isotopes

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

Using the information from the uranium composition of both fresh and reprocessed material, two important assertions are made.

Assertion 1: In uranium enriched up to 94% ^{235}U by weight the maximum weight fraction of the three lesser uranium isotopes (^{234}U, ^{236}U, and ^{238}U) for ^{234}U is 0.20 as shown by equation (1) below:

$$^{234}\text{U Weight Fraction} = \frac{234U}{234U + 236U + 238U} = \frac{4.00}{4.00 + 1.00 + 15.00} = 0.20 \quad (1)$$

Note that these numbers taken for fresh uranium compositions bound reprocessed uranium compositions, as the fraction of ^{234}U is even smaller for reprocessed material.

Assertion 2: In uranium enriched up to 94% ^{235}U by weight the minimum ratio of ^{238}U to ^{236}U is 1:1 as shown in Table V.

Note that these numbers taken for reprocessed uranium compositions bound fresh uranium compositions, as the ratio of ^{238}U to ^{236}U is much higher for fresh material.

Using the above assumptions a conservative bounding model was developed to show that ^{234}U can be ignored in regards to uranium mass limits. The model consists of an infinite homogenous medium composed of uranium isotopes. The uranium density used in the model is taken from the maximum density listed in the ANSI/ANS-8.1-1998[1] standard and is 18.81g/cm^3. Three different isotopic parameters were used: (1) ^{234}U mixed into ^{236}U in weight fractions ranging from 0.0 to 1.0 of ^{234}U, (2) ^{234}U mixed into ^{238}U in weight fractions ranging from 0.0 to 1.0 of ^{234}U, and (3) ^{234}U mixed into a ratio of ^{238}U:^{236}U of 1:1 in weight fractions ranging from 0.0 to 1.0 of ^{234}U. The third isotopic parameter is the conservative model incorporating information from Assertion 2. Figure 2 shows the three curves generated using these isotopic compositions.

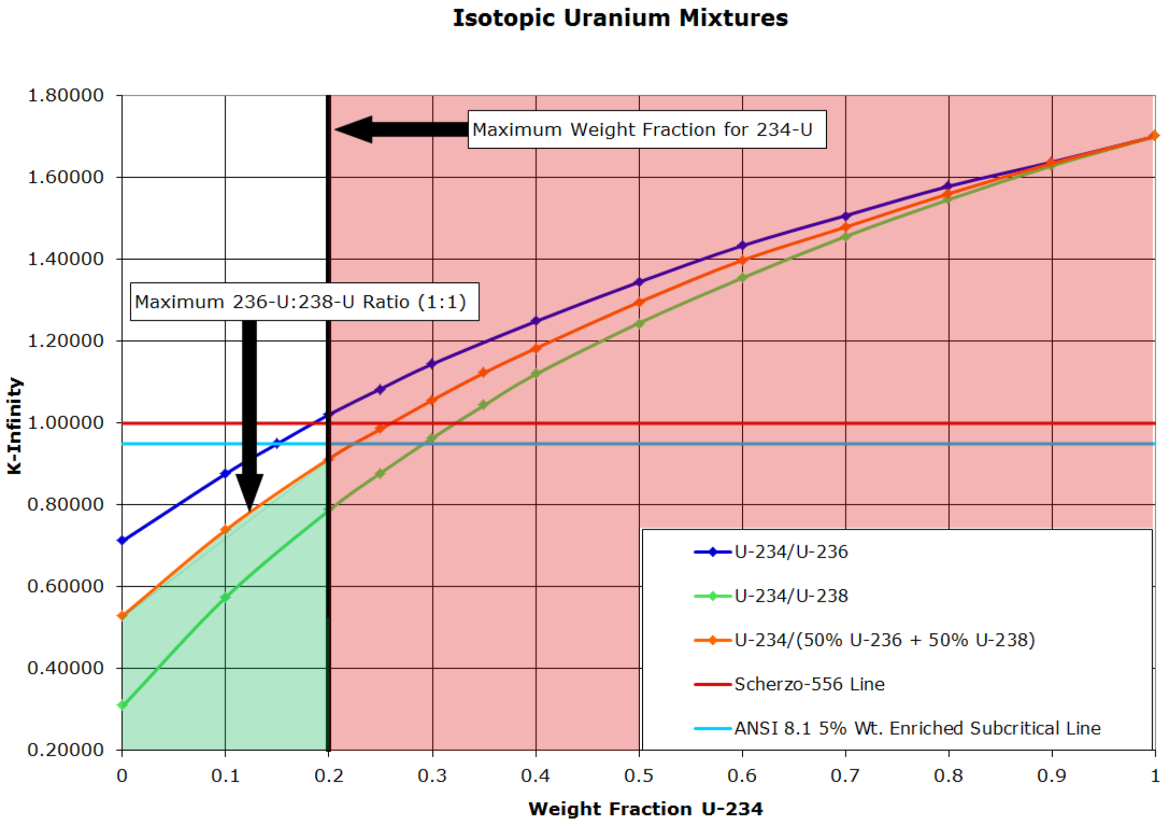

Figure 2. Isotopic Mixture k_{eff} Curves

At the far left of the plot, the points that lie on the x-axis represent points where ^{234}U is not present (weight fraction = 0). Thus, these points are the calculated k_∞'s for pure ^{236}U, pure ^{238}U, and the mixture of ^{238}U:^{236}U in a 1:1 ratio. As the weight fraction of ^{234}U increases (moving left to right on the plot), the k_∞ values also increase. At x = 0.20 the vertical line representing information from Assertion 1 is shown. This line marks where the weight fraction of ^{234}U is 0.20. As discussed in Assertion 1, 0.20 is the maximum weight fraction for ^{234}U in the lesser uranium isotopes present in uranium.

To establish a subcritical limit the ANSI/ANS-8.1-1998[1] 5% enriched ^{235}U limit is used in an infinite geometry and the resulting $k_{eff} = 0.95006 \pm 0.00110$ value is shown in Figure 2. Since the $k_\infty = 0.91367 \pm 0.00208$ of the 0.20 weight fraction ^{234}U mixed with ^{236}U and ^{238}U is less than the ANSI/ANS-8.1-1998[1] 5% weight enriched ^{235}U line it can be concluded that for any uranium enrichment level up to 94% ^{235}U by weight, ^{234}U **does not** need to be considered.

For completeness, weight fractions greater than 0.20 ^{234}U are plotted up to the point where pure ^{234}U is modeled at the far right of the plot. Here the 3 curves converge onto one point, which represents the $k_\infty = 1.70139$ value for pure ^{234}U.

Uranium enriched above 94% ^{235}U by weight has been produced. To allow for possible future operations involving this "supergrade" uranium, it is analyzed as well. Once uranium is above 94% ^{235}U by weight, the isotopic ratios change and the logic applied in Assertions 1 and 2 does not hold true. However, k_∞ values greatly exaggerate the effects of isotopic composition. Thus, for higher enrichments than 94% by weight a finite and practical application will be examined.

The benchmark HEU-MET-FAST-004[9] taken from the International Handbook of Evaluated Criticality Safety Benchmark Experiments describes an experimental benchmark of a 97.67% weight enriched ^{235}U reflected by effectively infinite water. An input was written for MCNP5 using the benchmark description. The isotopic composition for this benchmark as well as data for other "supergrade" material taken from Y-DR-128[10] is shown in Table VI.

Table VI. Isotopic Composition of "Supergrade" Uranium

Reference	Isotopic Weight Percentage			
	^{234}U	^{235}U	^{236}U	^{238}U
ICSBEP, HEU-MET-FAST-004	1.17	97.67	0.20	0.95
Y-DR-128	0.77	97.66	0.23	1.34

The benchmark case along with a few calculations encompassing changes involving the lesser uranium isotopes were executed. The results can be seen in Table VII.

Table VII. Results for Comparison to HEU-MET-FAST-004[4]

Case ID	Description	k_{eff}	$k_{eff}+3\sigma$	Difference (%)
NONE	Benchmark k_{eff}	1.00200 ± 0.00000		
HEU_01	MCNP5 Benchmark Calculation	1.00194 ± 0.00110	1.00524	+0.0000
HEU_02	^{234}U Modeled as ^{235}U	1.00243 ± 0.00108	1.00567	+0.0428
HEU_03	^{234}U Modeled as ^{238}U	1.00069 ± 0.00125	1.00444	-0.0796
HEU_04	Pure ^{235}U Sphere (Removed Other Isotopes)	1.00499 ± 0.00126	1.00877	+0.3512

As can be seen there is only a very slight difference between the benchmark and the MCNP5 calculation that is statistically insignificant. The important case to take note of is HEU_02. This case shows only a $\Delta k = 0.00043$ increase in reactivity when all the ^{234}U is treated as ^{235}U. The average total uncertainty in all 709 highly enriched uranium benchmarks in the International Handbook of Evaluated Criticality Safety Benchmark Experiments[4] is $\sigma_{total}=0.011$. An increase of $\Delta k = 0.00043$ when all ^{234}U is treated as ^{235}U is eclipsed by the uncertainty inherent in the experiments and thus is negligible in a criticality safety context.

4 CONCLUSION

The analysis in this evaluation proves that the relationship between ^{234}U, ^{236}U, and ^{238}U isotopes, described in Assertions 1 and 2 in Section 3.0 for enrichment up to 94% ^{235}U by weight, effectively bounds the ^{234}U reactivity and thus mass limits only need to be placed on ^{235}U.

5 ACKNOWLEDGMENTS

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