THE EFFECT OF U-234 CONTENT ON THE NEUTRONIC BEHAVIOR OF URANIUM SYSTEMS

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ABSTRACT

When analyzing uranium systems, the usual rule of thumb is to ignore the U-234 nuclide by assuming that it behaves neutronically like U-238. The absorption cross section of U-234 is indeed qualitatively very similar to that of U-238. However, thermal absorption cross section of U-234 is about 40 times that of U-238. At low U-235 enrichments, the amount of U-234 is quite small so the impact of assuming it is U-238 is minimal. However, at high enrichments, the relative ratio of U-234 to U-238 is quite large (maybe as much as 1 to 5). Thus, one would expect that some effect of using the rule of thumb might be seen in higher enriched systems. Analyses were performed on uranium benchmarks from the International Handbook. Although the benchmarks are adequately characterized as to the U-234 content, often, materials used in processing are not as well characterized. This issue may become more important with the advent of laser enrichment processes, which have little or no effect on the U-234 content. Previous work by Rucker and Johnson noted that the actual isotopics depend on specifics of the enrichment process so there is considerable uncertainty in the use of models to determine isotopics. Thus, it is important for criticality personnel to understand the effects of variation of U-234 content in fissile systems and the impact of different modeling assumptions in handling the U-234.

Analyses done on selected LEU, IEU and HEU metal and solution system benchmarks indicate that the effect of ignoring U-234 in HEU metal systems is non-conservative while it seems to be conservative for HEU solution systems. The magnitude of change in k-effective was as high as 0.4%, which has implications on selection of administrative margins and the determination of the upper subcriticality limit.

Key Words: u-234, enrichment, neutron spectrum, k-effective

1 INTRODUCTION

When analyzing uranium systems, the usual rule of thumb is to ignore the U-234 nuclide by assuming that it behaves neutronically like U-238. Thus for uranium systems, the uranium is evaluated as U-235 with everything else being U-238. The absorption cross section of U-234 is indeed qualitatively very similar to that of U-238. However, the thermal absorption cross section of U-234 is about 40 times that of U-238. At low U-235 enrichments, the amount of U-234 is quite small so the impact of assuming it is U-238 is minimal. However, at high enrichments, the relative ratio of U-234 to U-238 is quite large (maybe as much as 1 to 5). Thus, one would expect that some effect of using the rule of thumb might be seen in higher enriched systems.

Analyses were performed on selected uranium systems (LEU, IEU, and HEU) from the set of Benchmarks [1]. Although the benchmarks are adequately characterized as to the U-234 content, often, materials used in processing are not as well characterized. This issue may become more important with the advent of laser enrichment processes, which have little or no effect on the U-234 content. Analytical results based on the relationship of U-234 activity to that of U-235 have shown good predictive capability but with large variability in the uncertainties [2]. Rucker and Johnson noted that the actual isotopics vary with enrichment, design of the enrichment cascade, composition of the feed material, and on blending of enrichments so there is considerable uncertainty in the use of models to determine isotopics. Thus, it is important for criticality personnel to understand the effects of variation of U-234 content in fissile systems and the impact of different modeling assumptions in handling the U-234.

Table I shows typical approximate isotopics of uranium systems for some different U-235 enrichments and different enriching processes. Both gaseous diffusion and centrifuge processes produce about the same isotopic distribution while the laser enrichment processes only increase the amount of U-235 - leaving the same relative ratio of U-234 to U-238.

TABLE I – Isotopics for Different Enrichments

	Weight Percent				
Material	U-234	U-235	U-236	U-238	Ratio of U234/U238
DU (0.25%)	0.001	0.25	0.0	99.75	0.00001
Natural U	0.0054	0.711	0.0	99.284	0.00005
Non-laser LEU (3.5%)	0.03	3.5	0.0	96.47	0.00031
Laser (5%)	0.005	5	0.0	94.995	0.00053
Non-laser HEU (93.5%)	1.05	93.5	0.4	5.05	0.20800
Laser (93.5%)	.0004	93.5	0.0	6.4996	0.00006

2. CROSS SECTIONS

2.1 Absorption Cross Section Impact

As U-234 and U-238 are both threshold fissioning isotopes, the major effect between the two isotopes in a thermal system will be variation in absorption cross section. Figure 1 shows the absorption cross sections for U-234 and U-238.

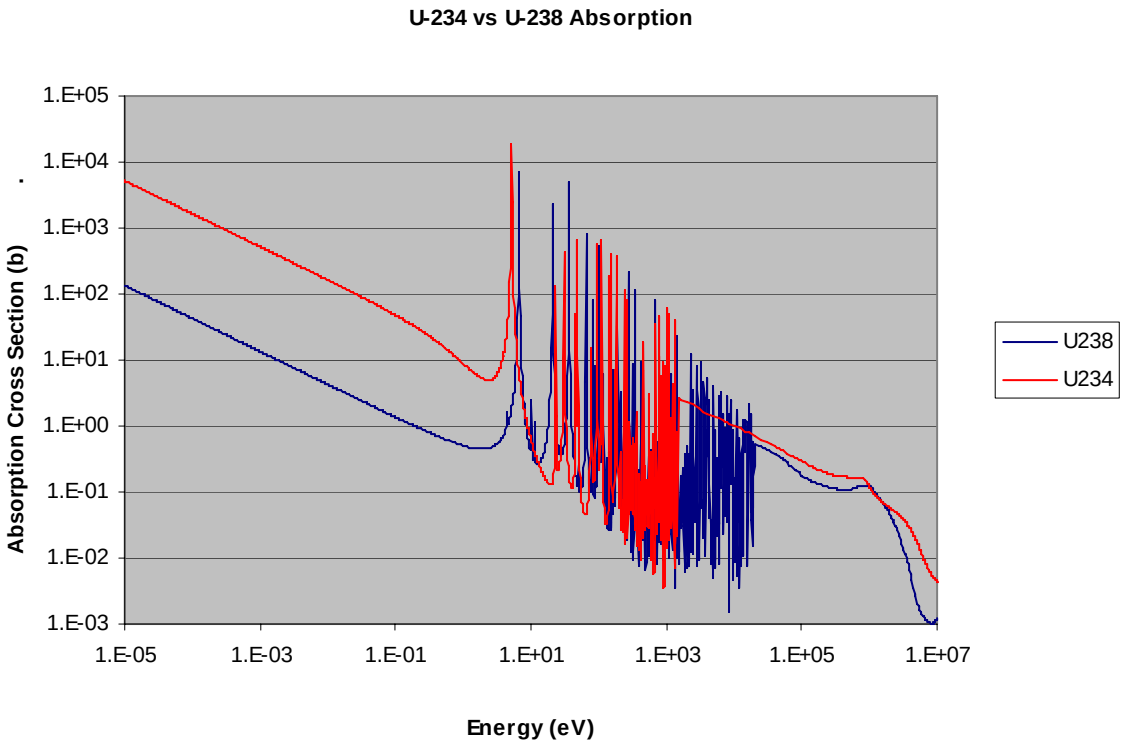

Figure 1 – Absorption Cross Sections for U-234 and U-238

U-234 has a cross section over the 1/v region that is 40 times that of U-238. In the resonance and fast regions, there seems to be little difference in absorption between the two isotopes. Thus, from a simple cross section analysis, it is expected that substituting U-238 for U-234 will have minimal effect on k-effective for LEU thermal systems, but because of the larger atom density, the presence of U-234 could have a larger, absorptive effect for HEU thermal systems. The overall impact of U-234 in high enriched systems will depend on the spectrum and the contribution from fission events as well as absorptions.

2.2 Fission Cross Section Impact

For uranium systems, the impact of the presence of U-234 on k-effective will depend on the average neutron energy and the relative ratios of fission and absorption cross sections for U-234 and U-238. In the previous section, it was noted that there was little difference between the absorption cross sections of the two isotopes in the fast region. So for fast systems, the major difference between U-234 and U-238 will be due to any differences in fission cross section. In solution systems, the impact will depend on the atom ratio of U-234 to U-238 as production from U-234 fission will exceed losses due to U-234 absorption. Figure 2 shows the ENDF-VII pointwise fission cross sections for U-234 and U-238.

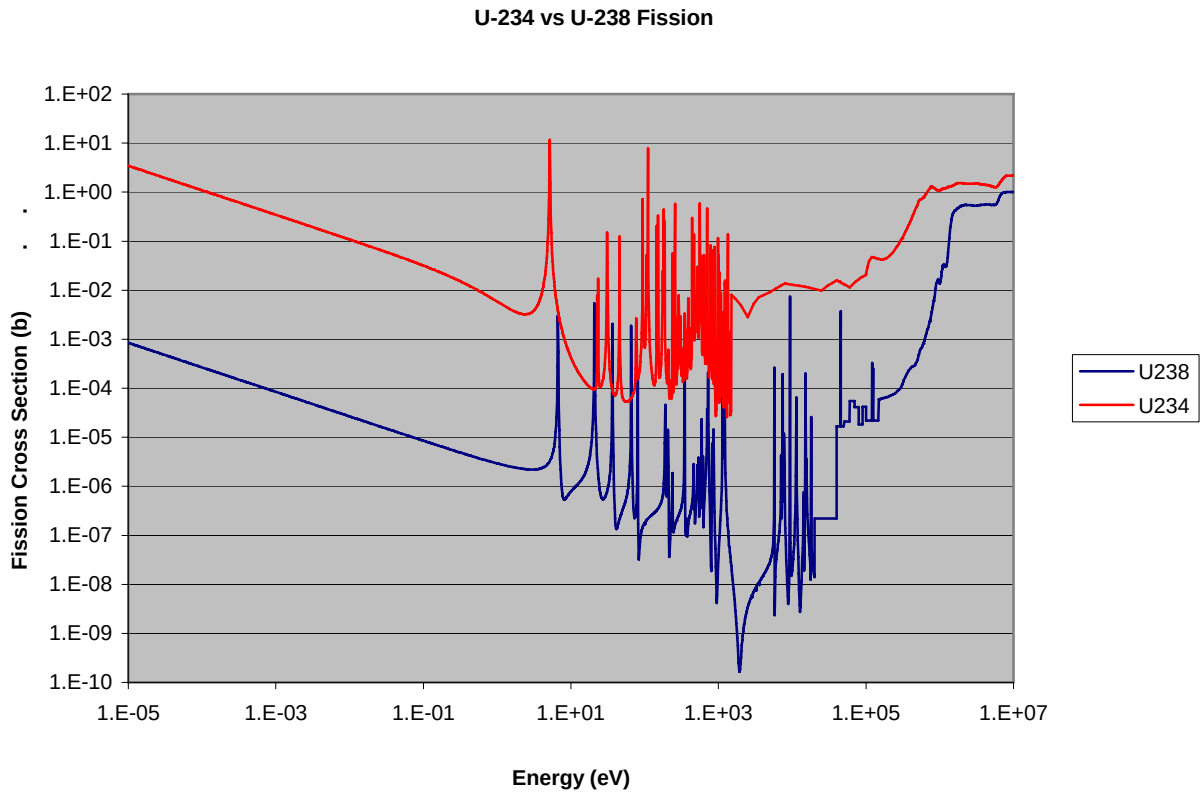

Figure 2 – Fission Cross Sections for U-234 and U-238

For fast systems, the impact is non-conservative as U-234 has a significant fission cross section below the 2 MeV fission threshold of U-238. At 1 MeV, the U-234 fission cross section of 1.09 barns is 80 times that of the 0.0136 barns U-238 fission cross section. The difference is less pronounced above 2 MeV where the U-234 fission cross section averages about 1.5 barns compared to an average of about 0.55 barns for U-238. Because the U-234 fission cross section is higher in the fast region, substitution of U-238 for U-234 will under predict the neutron production rate by fission. This will have little impact in LEU systems due to the low atom density of U-234. However, for HEU systems, this could have a significant impact with the atom density of U-234 being as high as 20% that of U-238.

Like the absorption cross section, the fission cross section for U234 is significantly higher than that of U238 in the thermal, 1/v region. However the ratio is significantly larger than for absorption with the U234 cross section being about 4000 times that of U238 at thermal energies and about 3000 times that of U238 in the epithermal region. In fact, U234 can add to neutron multiplication in the thermal region while U238 does not. For LEU systems, this significant difference will still have little impact due to the very small amount of U-234 in the system.

There is little difference in the scattering cross sections of the two isotopes with U234 being only a factor of two more than that of U238 in the thermal and epithermal regions. There is almost no difference in scattering cross section for the two isotopes above 10 eV.

3. NEUTRONIC IMPACT AND K-EFFECTIVE

3.1 Benchmarks

Four benchmarks [1] were selected for the evaluation of the impact of the U-234 on the system k-effective. The 4 were:

- LEU-SOL-THERM-002-001
- IEU-MET-FAST-003-001,
- HEU-SOL_THERM-001, and
- HEU-MET_FAST-018-001s

These four cover both metal and solution systems and the fast, intermediate, and thermal energy regions. All analyses were done with KENO V.a and the v7-238 library. Most cases were evaluated with 100,000 neutrons per generation for 1500 generations, skipping 500 generations. As the statistical uncertainties for runs within each experimental set are roughly equivalent, the difference in k-effective should be significant in both sign and magnitude. The fissile isotopics for the benchmarks are shown in Table II.

Table II. – Fissile contents of Benchmark experiments

	Weight Percent			
Material	U-234	U-235	U-238	Ratio of U234/U238
LEU-SOL-THERM	0.02	4.89	95.09	0.00021
IEU-MET-FAST	0.32	36.53	63.14	0.00514
HEU-SOL-THERM	1.02	93.17	5.81	0.17591
HEU-MET-FAST	1.10	89.50	9.40	0.11744

3.1.1 Analyzing LEU-SOL-THERM-002

This experiment involved bare 174-liter spheres (34.399 cm radius) of 4.9% Uranium Oxyfluoride solution. For the low-enriched, thermal solution system, including U-234, the k-effective was 1.00030 +/- 0.00007. Removing the U-234 and replacing with the same atom density of U-238, the k-effective was 1.00097 +/- 0.00007. Thus the replacement of U-234 with U-238 increases the k-effective (by about 0.07% +/- 0.1%) in a low enriched solution system likely due to the lower absorption cross section of U-238 in the thermal region.

3.1.2 Analyzing IEU-MET-FAST-003-001S

This benchmark is a bare, metal spherical system of U(36), with an effective radius of 15.324 cm. For the intermediate enriched, fast metal system including U-234, the k-effective was 1.00455 +/- 0.00008. Removing the U-234 and replacing with the same atom density of U-238, the k-effective was 1.00242 +/- 0.00008. Thus the replacement of U-234 with U-238 decreases the k-effective (by about 0.21% +/- 0.1%) in an intermediate metal system likely due to the threshold of the fission cross section of U-238 being above 1 MeV.

3.1.3 Analyzing HEU-SOL-THERM-001

This experiment involved minimally reflected cylinders of highly enriched solutions of uranyl nitrate, 93.172% U-235. For the heu, thermal solution system, two different experimental configurations with different solution concentrations were analyzed. For Case 1 (0.146 g U/cc), including U-234, the k-effective was 0.99805 +/- 0.00011. Removing the U-234 and replacing with the same atom density of U-238, the k-effective was 0.99998 +/- 0.00011. Thus the replacement of U-234 with U-238 increases the k-effective (by about 0.19% +/- 0.2%) in a thermal solution system likely due to the lower absorption cross section of U-238 in the thermal region. For Case 10 (0.064 g U/cc), including U-234, the k-effective was 0.99832 +/- 0.00011. Removing the U-234 and replacing with the same atom density of U-238, the k-effective was 1.00022 +/- 0.00011. Again, for a thermal, high enriched system, the replacement of U-234 with U-238 increases the k-effective (by about 0.19% +/- 0.2%) in a thermal solution system likely due to the lower absorption cross section of U-238 in the thermal region.

3.1.4 Analyzing HEU-MET-FAST-018-001S

This benchmark is a bare, metal spherical system of U(90), with an effective radius of 9.154 cm. For the heu, fast metal system including U-234, the k-effective was 0.99952 +/- 0.00009. Removing the U-234 and replacing with the same atom density of U-238, the k-effective was 0.99541 +/- 0.00008. Thus the replacement of U-234 with U-238 decreases the k-effective (by about 0.41% +/- 0.1%) in a fast metal system likely due to the lower fission cross section of U-238 in the fast region.

3.2 Effect of U-234 content on k-effective

There are two impacts from U-234 in uranium systems: impact of atom density and impact of cross sections based on the system spectrum. The larger the atom density of U-234 in the system, the larger the impact on k-effective. So in typical systems where the U-234 content follows (is proportional to) the U-235 content, the higher enriched systems are more impacted by the presence of U-234. However, the sign of the impact depends on the spectral characteristics of the system. Metal systems with a fast neutron spectrum see a decrease in k-effective with the replacement of U-234 while solution systems with thermal spectra have an increase in k-effective when U-238 is used to simulate the U-234 neutronics. Review of about 40% of the high enriched, fast metal benchmarks [1] indicated that the U234/U238 ratios were typically between 0.12 to 0.20. However, for those systems with 96 or 97% U-235, the ratios increased to between 0.8 and 1.0. It is expected that these systems will show a similar non-conservative reduction in k-effective when U-234 is replaced by U-238. The above evaluation was for bare systems where scattering effects are reduced. Fully reflected systems seem to be more sensitive to U-238 scattering; so the effect of U-238 replacing U-234 will probably reduce the k-effective variations. Further study is needed to check this.

4. SENSITIVITY ANALYSES OF 234 WITH TSUANMI

To examine the sensitivities of the systems and compare the results with the k-effective analyses, each of the benchmark systems were evaluated using Tsunami. [3] For each benchmark, sensitivities of capture, fission, and scattering were calculated for U-234 and U-238. An example of the results for fission in the heu-met-fast system is shown in Figure 3. Figure 4 shows similar values for fission in the heu-sol-therm system.

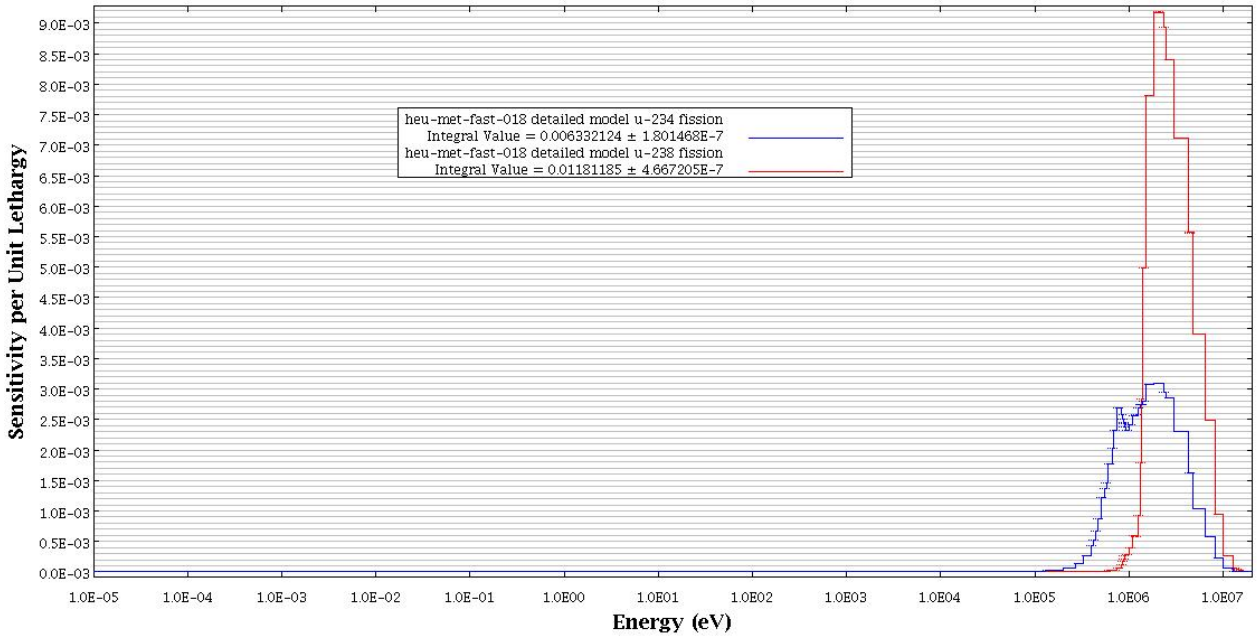

Figure 3 – Sensitivity of Fission in U-234 and U-238 for HEU-MET-FAST-018

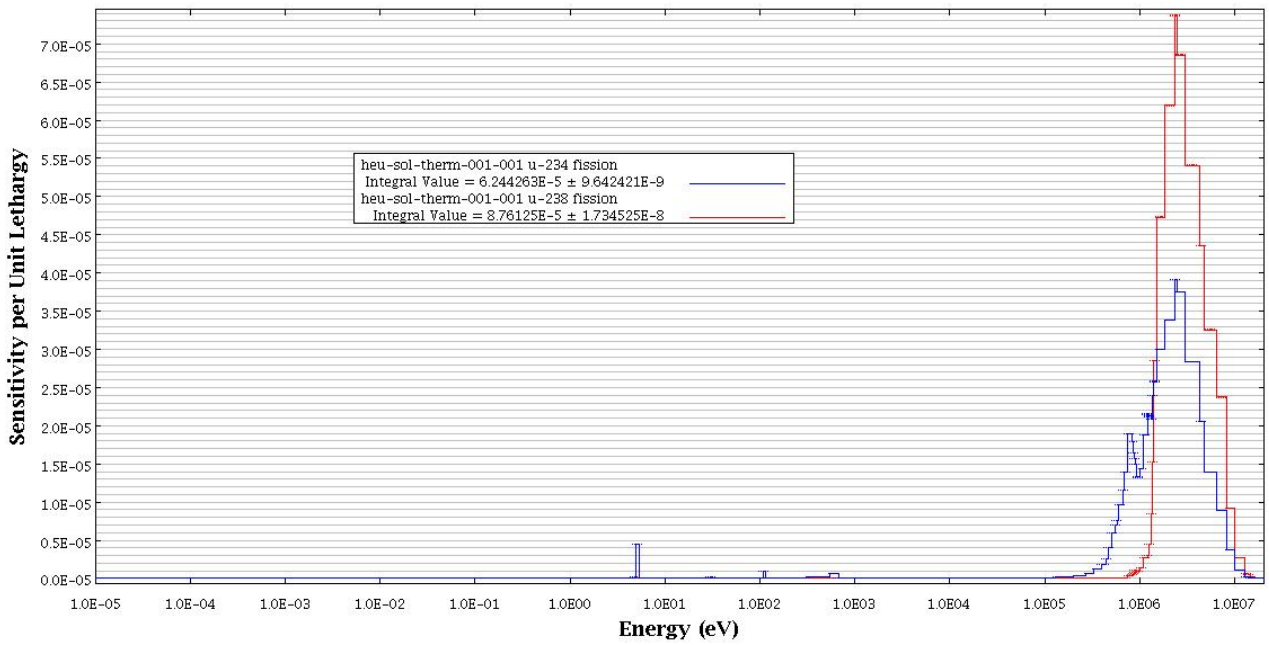

Figure 4 – Sensitivity of Fission in U-234 and U-238 for HEU-SOL-THERM-001

These two plots show that U-234 has a lower threshold for fission than does U-238, but that the high enriched systems are more sensitive to U-238 fission because its atom density is about 8 times that of U-234 in these systems. Replacement of U-234 with U-238 does not cover the same energy ranges, but the lower atom density of U-234 means the replacement has a smaller impact on a fast system.

The next two plots, Figures 5 and 6 show the sensitivity of capture/absorption to amounts of U-234 and U-238 for a high enriched thermal solution system and a low enriched thermal solution system. The U-234 sensitivity is covered by that of U-238 in both systems. However, it is interesting to note that high enriched system is more sensitive to U-234 capture even though there are 5 times as many U-238 atoms in the system. For the low enriched system, there is little sensitivity to U-234 as there is only one atom of U-234 for every 4600 atoms of U-238.

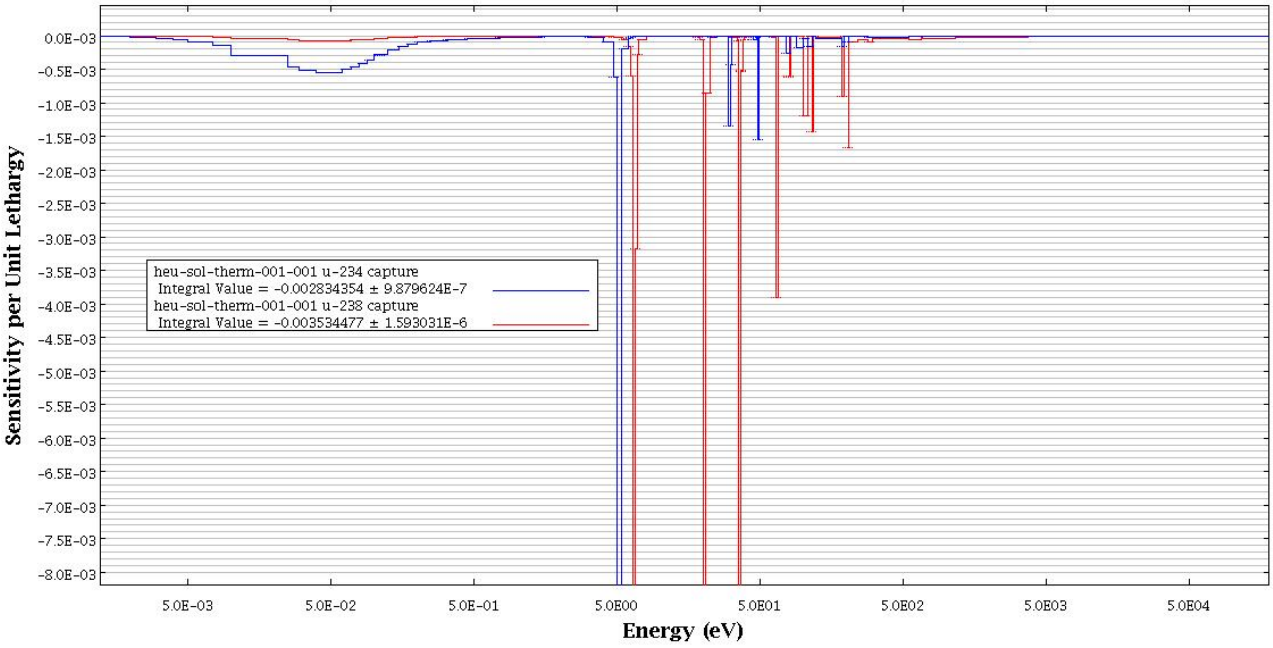

Figure 5 – Sensitivity of Capture in U-234 and U-238 for HEU-SOL-THERM-001

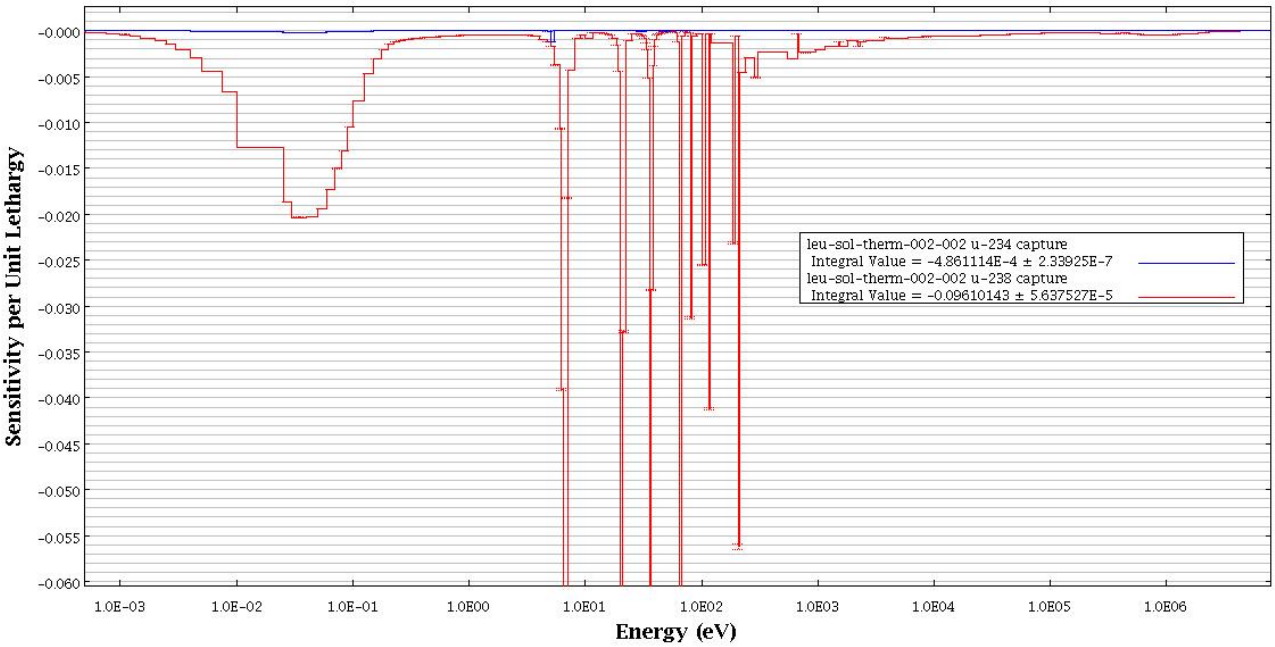

Figure 6 – Sensitivity of Capture in U-234 and U-238 for LEU-SOL-THERM-002

4.1 Calculation of Overall δk/k Changes

Calculation of the change in k-effective requires the overall sensitivity coefficients for each isotope.

$$\frac{\delta k}{k} = S_{234} * \left(\frac{\Delta N_{234}}{N_{234}} \right) + S_{238} * \left(\frac{\Delta N_{238}}{N_{238}} \right) \tag{1}$$

As we are completely removing U-234, then (ΔN234/N234) will be -1.0, and we are replacing U-234 with U-238, so (ΔN238/N238) will be N234/N238. Therefore,

$$\frac{\delta k}{k} = S_{234} * (-1) + S_{238} * \left(\frac{N_{234}}{N_{238}} \right) \tag{2}$$

where S_{234} and S_{238} are are total (energy-integrated) sensitivity coefficients calculated by Tsunami. For the high enriched solution (heu-sol-therm-001) system we get total sensitivity coefficients of:

S_u234= -2.72388E-03
S_u238= -3.10289E-03

Thus δk/k = -1*-2.72388E-03 – 3.10289E-03 * (3.831e-06/2.1411e-05) = 0.217%, which is approximately what was determined from direct calculations with KENO V.a.

Table III shows the results of δk/k calculations from Tsunami and compared with the direct calculations using KENO V.a. For most cases, the direct calculations seem to match well with those from Tsunami. The solution thermal systems seem to be a little less amenable to the direct calculation method.

Table III – Comparison of KENO V.a and Tsunami δk/k Calculations

	δk/k replacing U-234 with U-238, same atom density		
SYSTEM	from KENO	tsunami	keno/tsunami
leu-sol-therm-002-001	0.00067 +/- 0.00010	0.00044	1.51
ieu-met-fast	-0.00214 +/- 0.00011	-0.00208	1.04
heu-sol-therm-001	0.00193 +/- 0.00016	0.00217	0.89
heu-sol-therm-010	0.00190 +/- 0.00016	0.00157	1.21
heu-met-fast-018	-0.00411 +/- 0.00012	-0.00423	0.97

Another advantage of using Tsunami is the ability to separate regions of coverage and impacts for specific processes (i.e., absorption, fission, and scattering). For example in the fast, metal system, the individual sensitivity coefficients show that the system is about 14 times more sensitive to U-234 fission than to U-234 capture, while it is only 4.5 times as sensitive to U-238 fission than to U-238 capture.

5. CONCLUSION

Although the absorption cross section of U-234 is much greater than that of U-238 in the thermal region and U-234 has a small thermal fission cross section (0.066 b), the replacement of U-234 with U-238 in most systems results in an absolute value δk/k of less than 0.4%. However, for metal systems, the direction of change is non-conservative where the k-effective calculated without U-234 is lower than that with U-234 by 0.4%. This amount may be as much as 20% of the administrative margin, so the assumption that U-234 "behaves neutronically" like U-238 may have a significant impact in criticality safety analyses for high enriched metal systems.

With new enrichment techniques, the amount of U-234 in enriched materials could vary by a factor of 1000. In many cases, the actual U-234 atom density is not well known, but these calculations have shown that there is less than a 0.4% effect on k-effective when all U-234 material is replaced by U-238.

6. ACKNOWLEDGMENTS

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7. REFERENCES

1. NEA Nuclear Science Committee, *International Handbook of Evaluated Criticality Safety Benchmark Experiments*, September 2010.
2. Rucker, T.L., and C.M. Johnson, Jr., "Relationship between isotopic uranium activities and total uranium at various enrichments," *Journal of Radioanalytical and Nuclear Chemistry*, **Vol. 235**, Nos. 1-2, pgs. 47-52, (1998).
3. Rearden, B.T., D.E. Mueller, S.M. Bowman, R.D. Busch, and S.J. Emerson, "TSUNAMI Primer: A Primer for Sensitivity/Uncertainty Calculations with SCALE," *ORNL/TM-2009/27*, Oak Ridge National Laboratory, Oak Ridge, Tenn. January 2009.