Criticality Comparison Of Highly Enriched Uranium Dispersed Over Any Volume As Solid Particles, And As A Solution

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Abstract

An optimised rectilinear configuration containing a homogeneous solution of uranium metal with 1.3kg ^{235}U and water was compared to the same water-moderated and reflected configuration with uranium metal being modelled in progressively smaller lumps. The results of KENO-V.a calculations with a benchmarked ENDF/B-V library showed that, for this highly-enriched uranium (HEU) to be in a critical state, the fissile material must either be arranged in a water moderated configuration of at least 250,000 evenly spaced HEU chunks with optimum reflection, or dissolved completely in acid. A scenario involving metallic 1.3kg ^{235}U in a flooded environment would not be sufficient to cause a critical configuration. Some dissolving agent such as acid must be present to dissolve the uranium into a solution, to achieve the spacing between uranium particles that is required for a critical configuration. The macroscopic cross-section of the uranium atoms is compared to the cross section of the uranium spheres to demonstrate that a self-shielding effect remains even when the particles are smaller than 0.1 mm diameter.

1. Introduction

Highly-enriched uranium (HEU) fuel bundles are used in some non-power-generation reactors. Prior to manufacture, subcritical quantities of HEU metal are normally stored in a significantly subcritical state. Under abnormal flooding, the boxes of HEU are designed to be neutronically isolated. Two storage boxes each with a subcritical mass of 650 g ^{235}U were assumed to be on the floor in close proximity to each other, in a flooded room. Note that this configuration is considered to be beyond credible abnormal conditions, because at least two separate abnormal events must occur independently. A homogeneous solution of uranium metal with 1.3kg ^{235}U and water was simulated using KENO-V.a [1]. The k-effective of this configuration was in excess of the minimum value required for an administrative margin of subcriticality of 0.95.

A stylized calculation involving particles of ^{235}U metal in water showed an almost linear dependence of the multiplication constant on the surface area of the particles in the water moderated lattice. This relationship asymptotically approaches the multiplication constant of a homogeneous mixture, as the effect of self-shielding disappears. If this mass were to be composed of metal particles, evenly distributed throughout a water-moderated cubic lattice, then the surface area of all particles would need to be in excess of 4000 cm^2 to obtain a critical configuration.

2. KENO-V.a Computer Code

The KENO-V.a computer code was used for this study. KENO-V.a is an extension of the KENO-V.a Monte Carlo criticality program developed for use with the SCALE (Standardized Computer Analyses for Licensing Evaluation) code system [1]. The primary purpose of KENO-

V.a is to determine k-effective for an assembly of fissionable material. The main improvement over previous versions of KENO-V.a is an extended geometry capability, which allows explicit modelling of many complex, heterogeneous geometries that previously required modelling as homogeneous regions. In addition, KENO-V.a uses multigroup P_n-scattering cross-sections that are rigorously weighted for the specific problem using the cross-section processing codes of the SCALE system, rather than using a predefined subset of cross sections created using a generalized resonance correction. Other quantities calculated by KENO-V.a include mean neutron lifetime and generation time, energy-dependent leakages, energy- and region-dependent absorptions, fissions, fluxes, and fission densities.

All of the KENO-V.a models had an external albedo, representing 12 inches of concrete, as it was found to be the most conservative option. For all water-moderated models in this analysis, the fissionable material was surrounded by at least 100 cm of water, which was enough to reduce the impact of the concrete albedo to negligible amounts. Where there are non-fissionable-metal components in the system to be modelled, it is conservative to exclude them from the model, as sensitivity studies show that less iron or aluminum allows for a more critical system. This simplifies some cases that are otherwise impractical to model in KENO-V.a.

3. Cross-Section Library

The cross-section library used with KENO-V.a is the 44-group ENDF/B-V library, developed for use in the analysis of fresh and irradiated fuel and radioactive waste systems [2]. The 44-group library is a collapsed version of the ENDF/B-V 238-group library, with 22 fast and 22 thermal energy groups. Validation of the 44-group library within the SCALE system is based on a comparison of calculated values of k-effective with data from 93 experiments: 92 critical and one subcritical [3]. Some work has also been carried out within AECL on the validation of KENO-V.a for calculating k-effective values for highly-enriched uranium systems [4]. These validation studies, originally performed using the 27-group ENDF/B-IV cross-section library, were re-evaluated using the 44-group library. These experiments include a wider range of HEU systems, including uranium metal and solution experiments, and critical lattices of uranium/aluminum alloy fuel assemblies.

The mean multiplication constant k_m of the 27 critical experiments involving HEU systems from this study (reflected and unreflected, moderated and unmoderated) is $k_m \pm \sigma_m = 0.9976 \pm 0.00883$, calculated using the using the 44-group library. Note that the mean k_m should be as close to 1.0 as the modelling accuracy of KENO-V.a will allow. The standard deviation σ_m from the mean is expected to be larger than the statistical σ calculated by KENO-V.a for each case, because it reflects modelling and manufacturing inaccuracies.

The results calculated by the 238-group ENDF/B-V library were within one mk of the results calculated by the 44-group ENDF/B-V library. The definition of ‘mk’ (milli-k) is a measure of reactivity: a critical system is 1000 mk (i.e., k-effective=1). The critical experiments modelled with the 44-group library had a slightly higher k-effective on average and a slightly larger uncertainty. According to [4], the large uncertainty is caused primarily by the inaccuracy of the experimental modelling documentation or implementation rather than by the KENO-V.a code. The simple models are more accurate (closer to the measured k-effective of 1) because there are less modelling or manufacturing inaccuracies.

4. Acceptance Criterion for Safe Subcriticality

The purpose of the validation studies was to establish the bias and uncertainties in KENO-V.a's prediction of the multiplication constant also referred to as k_{eff} or k-effective. The convention adopted in the code validation and uncertainty assessment of KENO-V.a requires a modelling uncertainty allowance of 3 σ_m, which, based on the results of the validation exercises, is up to 0.03. The analysis limit to ensure subcriticality for normal operation involving well defined conditions and configurations of fissionable material, and for which adequate benchmark data exists to judge the accuracy of the calculated k-effective value, is

$$(k_{eff} + 3\sigma) < 0.95 \quad (1)$$

where σ is the statistical standard deviation in the calculated k-effective determined by KENO. This limit is consistent with the definition of safe subcriticality in Section 4.1.2 of Reference 5, which is accepted international practice. Thus, if KENO-V.a predicts a $(k_{eff} + 3\ \sigma) \geq 0.95$, criticality is assumed to be feasible. For the criticality analyses presented below, the conservative assumptions for the event scenarios account for all other uncertainties and further uncertainty allowances in the acceptance criterion are not required. The upper limit on safe subcriticality of 0.95 includes allowances for uncertainties in the computer program and nuclear data (simulation offset or bias), specification and modelling of scenarios, any other uncertainty sources, and an additional arbitrary allowance, which, in total, are less than 0.05. This total allowance is consistent with the American and Canadian standards [5,6].

In all cases below, the "maximum k-effective" quoted is that of k-effective + uncertainty of 3σ, which represents the upper bound of the range of k-effective, 99.5% of the time, for the most limiting geometry, as determined by the KENO-V.a code. The definition of "safely subcritical" refers to the "maximum k-effective" of a configuration being less than 0.95, which we set as the threshold for safe subcriticality based on KENO-V.a calculations.

5. Lattice of Uranium Particles

The purpose of this paper is to show the extent to which uranium metal must be oxidized into tiny particles, to create a criticality hazard without being dissolved in some form of acid. KENO-V.a [1] was used to model 1.3 kg ^{235}U in the form of 93.5wt% ^{235}U uranium metal that was subdivided into progressively smaller spheres. Each time, a lattice was designed in which each uranium sphere was placed in the centre of a water filled box. This lattice of uranium spheres is surrounded on all sides by up to one meter of water.

An array of 2 x 2 x 2 spheres of uranium metal suspended in a water-filled lattice was created. The case was repeated with each lattice being progressively subdivided into 2x2x2 smaller boxes each filled with 1/8 of the material originally modelled for the box that was eight times bigger. Overall, the uranium mass and water moderation was conserved, but the lattice unit cell was progressively reduced in size to simulate the stages of uranium dissolution. Eventually, the array became an array of 2^3 arrays of 2^3 arrays of 2^3 arrays of 2^3 arrays of 2^3 arrays of 2^3 arrays of eight elements each of which was 8^8 = 16,777,216 times smaller than the original sphere surrounded by (the same proportion of) water of various quantities. In this way, the optimum volume garnering the maximum reactivity for each array could be ascertained by varying the volume over which the lattice was arrayed. This was represented within KENO-V.a as an array of 64 cells, each with eight sub-cells, each subdivided eight times further at each sub-level. An

array of 64 cells would be represented as $64 = 4 \times 4 \times 4 = 2^2 \times 2^2 \times 2^2$ particles in each of three directions.

The results of KENO-V.a calculations with the benchmarked 44-group ENDF/B-V library, shown in Figure 1, demonstrate that 1.3kg ^{235}U in the form of HEU can exceed the state of safe sub-criticality. For this to happen, the fissile material must be arranged in a water-moderated configuration of at least 250,000 evenly spaced HEU chunks with optimum reflection, or dissolved completely in acid.

6. Comparison with a Solution of Highly Enriched Uranium in Water

During a fire, some of the uranium metal in the boxes could be oxidized and converted into UO_2 powder. The minimum critical mass of Uranium oxide is greater than that of uranium metal, but powder or small pieces could be dispersed in water more easily than chunks of solid metal, which makes criticality more credible. It would be very conservative to assume full oxidation of all the uranium, since it starts out as solid metallic chunks, in closed metal boxes. One litre of air would only oxidize two grams of uranium to UO_2, and the air inside each box would oxidize less than ten grams of uranium. However, we postulate that the oxidation is sufficient to break the uranium into smaller pieces.

Figure 1 and Table 1 show an analysis of 1.3 kg of uranium enriched to 93.5 wt% ^{235}U in an optimized rectilinear configuration (i.e. a cube). As the uranium particles become smaller, the multiplication constant of the system increases. This configuration could be part of a critical system only if the uranium were broken into many thousands of particles distributed throughout a water reflected volume. This would require the full oxidation of uranium in the boxes to powder form. The average particle size of uranium metal would need to be about 0.005 g to exceed the upper limit of safe subcriticality. These particles would be smaller than 1 mm^3, or have an average surface area of less than 0.02 cm^2.

For a constant fissionable-mass-to-moderation ratio, Figure 1 demonstrates that the k-effective increases as particle size decreases. The quantity of fissionable material described above could only form a critical system if it were to fall into a vat of nitric acid or other acidic solution that dissolved the uranium and diluted it over a volume not less than 6 L. The limiting value of k-effective for 1.3 kg ^{235}U in solution distributed over any volume is 1.1, including the 3 σ uncertainty.

7. Suspension of Uranium as a Slurry

Let us assume that the uranium is fully oxidized within the storage boxes. If the uranium is exposed to enough heat and oxygen for full oxidation, then it would also be likely to reach a higher oxide state, such as U_3O_8. The minimum critical mass for U_3O_8 powder in water is significantly higher than for a homogeneous mixture of uranium and water.

The density of U_3O_8 (8.20 g/cm^3) is still much greater than that of water, and it would tend to settle to the bottom of the box. The slurry of uranium would tend to settle into a slab at the bottom of the storage box. The maximum mass of U_3O_8 in each box would be 650 g of ^{235}U + 45 g ^{238}U + 125 g oxygen = 820 g, with a volume of 100 cm^3. For a slurry with a solid volume fraction of 10%, the volume of the mixture would be 1 L. More than six litres are required for a double batch to be critical, according to Table 2.

To obtain a critical system, the uranium oxide powder would have to be forcibly distributed throughout the boxes. This would require extended agitation of the water to stir up the uranium oxide powder, to prevent it from settling in the corners of the box. No mechanism for extended agitation has been postulated as part of a single accident sequence that also includes double batching and flooding. Therefore, this critical configuration could only be possible with at least one more independent, unlikely event.

8. Sensitivity Studies

The minimum critical mass of uranium is 780g ^{235}U in HEU of about 93 wt% ^{235}U, according to Reference [3]. This mass was modelled using KENO-V.a model with the 44-group ENDF/B-V library. As expected, the k-effective for 780g ^{235}U evenly distributed throughout a sphere with a volume of 13 to 17 L was 1.000 ±0.002. The studies in previous sections were all done for a cubic geometry with a 100cm water reflector on all sides. To properly compare these cases, 780g ^{235}U was also modelled as a solution in a cubic container. The difference between a cubic geometry and a spherical geometry was an additional 20 mk for the spherical container of fissionable material, which is 2% higher than that of the cubic container for the optimum configuration. A comparison of the above results to that of the minimum critical mass of ^{235}U is shown in Table 2 and Figure 2.

The 1 σ uncertainty for most of the cases presented in this paper is about 2 mk. All numbers presented include 3 σ uncertainty, which adds a conservative bias of up to 7 mk. The uncertainty in the above analysis was minimized as much as possible by increasing the iterations to the allowable KENO-V.a memory limit. The total number of generations multiplied by particles per generation allowed for this analysis was about 400*1000 = 400000 particle histories.

The optimized configuration for reactivity of a solution or slurry is the spherical form, although the cube is within 2%. An additional 1% reactivity occurs if the water and the uranium is 90˚C instead 20˚C as for the majority of results reported in this paper. All calculations documented in this paper should be repeated independently with a different code to better estimate the accuracy of this analysis.

9. Mathematical Basis for Self-Attenuation of ^{235}U

According to Reference 7, the mean free path of a neutron in a lump of uranium is 4V/S, where V and S are the volume and surface area respectively, of the lump. The absorption mean free path Λ_a is defined by the following equation:

$$\Lambda_a = \frac{1}{N\ \Sigma\zeta(E)}\ , \tag{2}$$

where $\Sigma = \frac{S}{4} = \frac{V}{\ell}$ is the collision cross-section of the lump, (3)

ℓ is the mean distance travelled in it by neutrons

N is the number of uranium spheres per unit volume,

ζ is the sticking coefficient (whether the neutron will stay in the lump/sphere), and

E is the energy group.

Reference 7 concludes that the multiplication constant is proportional to the surface area of the uranium lumps. The lumps can be any shape, because the key ratio is that of cross-sectional area of all uranium spheres as they appear to an on-coming neutron, relative to the volume of the moderator and fuel mixture.

Figure 3 shows the peak multiplication constant of the system relative to the total uranium particle cross-section. It also shows the multiplication constant of ^{235}U particles in a fixed volume of 13.8 Litres. The macroscopic particle cross-section is S / 4, as given in Equation 3, plotted against the volume over which it has the greatest k-effective. A second curve is added to show the effect of particle size on a mixture of 93.5 wt% ^{235}U and water over a fixed volume. As the surface area of the particles increases, the reactivity of the system increases monotonically. This implies that Uranium self-shielding or self-attenuation has a large impact on the criticality of a system, when it is not in solution. As the particle size decreases, the self-shielding effect decreases, approaching the k-effective of the solution asymptotically, as the particle size is reduced to less than 1 mm diameter.

The k-effectives of Figure 3 were generated by KENO-V.a [1] for the optimum amount of interstitial water moderation and a 30 cm water reflector. The predicted k-effective was about 0.95 for a lattice of 262,000 evenly spaced spheres of HEU (93.5 wt% ^{235}U). If the box containing the 1.3kg ^{235}U is not a cube, the k-effective for a hypothetical solution in a less optimized rectilinear container is smaller.

The relationship of the multiplication constant to the surface area of Uranium in Figure 3 is not completely linear. At the low end of the curve, the lattice is not established until there are at least four particles in each direction. At the high end of the curve, the particles are so small that the self-shielding effects are reduced nearly to zero. The ^{235}U in solution has a higher k-effective than the particles of ^{235}U, as a result of the better distribution of ^{235}U in the solution. Once the particle size is reduced to below 1 mm^3, very little ^{235}U self-shielding effect is left. Even though the fissionable cross-section of ^{235}U is smaller than the area occupied by the atom, there is still a benefit to maximizing the distribution of ^{235}U over the available (optimized) volume. The thermal fission cross-section of ^{235}U is 585 barns for a pressurized water reactor lattice, according to Reference 8. The macroscopic thermal fission cross-section of 1.3kg ^{235}U in solution multiplied by the volume of uranium atoms is calculated below:

$$V_{U235} \cdot \Sigma_f = \frac{1300\text{g}^{235}\text{U}}{\rho_{^{235}\text{U}}} \cdot \frac{585 \frac{\text{barns}}{\text{atoms}} \cdot 10^{-24} \frac{\text{cm}^2}{\text{barn}} \cdot 0.6022 \cdot 10^{24} \frac{\text{Atoms}}{\text{mole}} \rho_{^{235}\text{U}}}{235 \frac{\text{g}^{235}\text{U}}{\text{mole}}} = 1949 \text{ cm}^2 \qquad (4)$$

where $V_{U235} = 1300\text{g} / \rho_{^{235}\text{U}}$ refers to the volume occupied by ^{235}U atoms.

Note that the mass of uranium in the solution incorporates the density of uranium and the volume of the solution. This implies that the majority of ^{235}U will be self-shielded until the total cross-sectional area of the uranium particles increases beyond 1000 cm^2. Since the area of the sphere that is visible to an approaching neutron is one quarter of the surface area of the sphere (i.e., the area of a circle compared to the surface area of a sphere), the cross-sectional area of all the spheres is one quarter of the surface area as defined in Equation 3. At the point in Figure 3 where the surface area is equal to the thermal fission pseudo-cross-section above, 262,000 spheres of macroscopic cross-section 1300 cm^2 and two million spheres of macroscopic cross-section 2600 cm^2 indicate an interpolated k-effective of 0.99. This implies that there is still a

(1.10-0.99)/1.10 = 10% reduction in k-effective as a result of self-shielding at this point, compared to a solution distributed over the same volume.

The volume of a uranium atom can be estimated by dividing the density of uranium by both the atomic weight of uranium and Avogadro's number. From this, the radius and surface area of a single atom can be estimated using the following formulae.

The density of natural uranium metal is 19.05 g/cm^3. The actual density of HEU is slightly lower, but the difference will be cancelled out in the volume calculation below.

$$\text{Volume of one mole uranium} = \frac{238.0\ \text{g/mole}}{19.05\ \text{g/cm}^3} = 12.49\ \frac{\text{cm}^3}{\text{mole}} \quad (5)$$

$$\text{Volume of one uranium atom: } V = \frac{12.49\ \text{cm}^3\ /\ \text{mole}}{6.022\text{E}23\ \text{atoms/mole}} = 2.075\text{E-}23\ \frac{\text{cm}^3}{\text{atom}} \quad (6)$$

$$\text{Radius of one uranium atom: } r = \sqrt[3]{\frac{3V}{4\pi}} = 1.705\text{E-}8\ \text{cm.} \quad (7)$$

$$\text{Cross-section of 1 uranium atom} = \pi r^2 = 9.13\text{E-}16\ \text{cm}^2. \quad (8)$$

The cross-section of one uranium atom is almost 10^9 barns. If the uranium spheres were the size of single atoms, then their surface area according to the above formulae would be six orders of magnitude larger than the cross-section recorded in Reference 8.

10. Discussion

The results of this analysis demonstrate the effect of ^{235}U self-shielding in water. Even when there are 1,000,000 particles smaller than 0.1 mm in diameter, the self-shielding effect of ^{235}U in water reduces the multiplication constant by 5%, relative to the value generated by a solution in the same volume. The macroscopic cross-section for uranium over the entire volume is about 1950 cm^2. The surface area of the one million particles can be nearly ten times this value, and yet self-shielding of the uranium still exists. The peak volume for a solution containing 1.3 kg ^{235}U is 17.6 L, but the total surface area of uranium particles increases monotonically as the number of particles increases, while approaching the value defined for a solution of HEU in water.

Figures 1 and 2 also demonstrate that the multiplication constant for configurations of uranium and water are dependent on the amount of moderation. Up to a certain point, extra moderation increases the k-effective. Beyond this point, additional neutron absorption in the water decreases the k-effective by overriding the benefit of extra moderation. This point is dependent on the surface area of the particles in solution.

11. References

1) "SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation," NUREG/CR-200, Revision 4, Vols. I, II and III, 1995.

2) W.C. Jordan and S.M. Bowman, "SCALE Cross-Section Libraries", NUREG/CR-0200 (ORNL/NUREG/CSD-2/V3/R5), Vol. 3 Sec. M4, Revision 5, 1997.

3) M. DeHart and S.M. Bowman, "Validation of the SCALE Broad Structure 44-Group ENDF/B-V Cross-Section Library for use in Criticality Safety Analyses", NUREG/CR6102, Oak Ridge National Laboratory, 1994 September.

4) R.H. McCamis and G.R. Penner, "Validation of KENO-V.a for Criticality Safety Calculations of Highly Enriched Uranium Systems", AECL-10146-3, 1999 January.

5) American Nuclear Society, "Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors", ANSI/ANS-8.1-1998, 1998 September.

6) Canadian Standards Association, "Dry Storage of Irradiated Fuel", CSA-N292.2-96, 1996 April.

7) A. Galanin, "Thermal Reactor Theory", Pergamon Press, 1960.

8) E.M. Baum et al, "Nuclides and Isotopes, 16th Edition of the Chart of the Nuclides", KAPL Inc. Lockheed Martin, 2002.

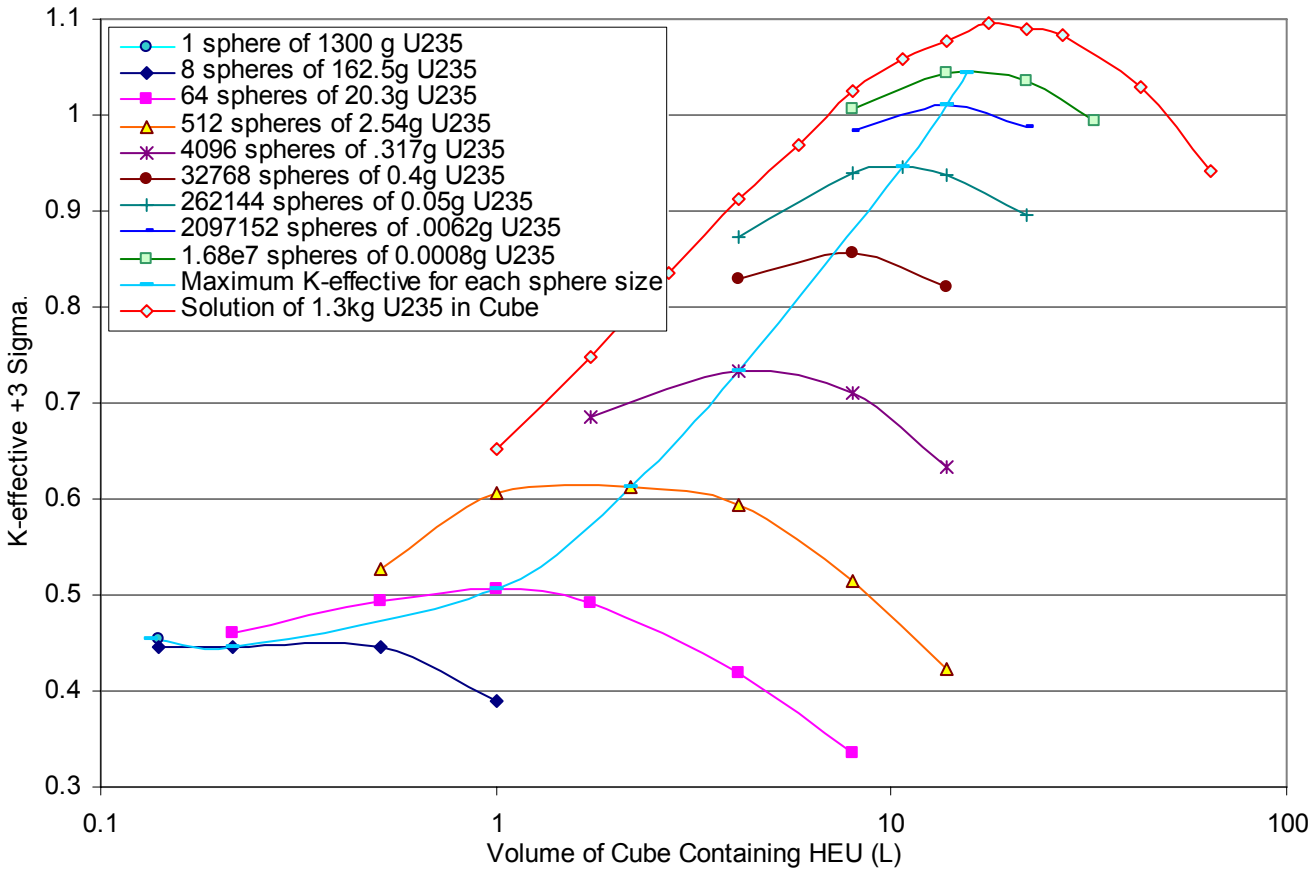

Figure 1 : K-effective +3σ for 1.3 kg ^{235}U in various forms

Table 1

Maximum multiplication constant with uncertainty for 1300 g ^{235}U in lattice of particles

Volume (mL)	Number of Particles	Diameter of Particles (cm)	$K_{eff} + 3\sigma$
1000	1	2.561	0.4550
1728	8	1.280	0.4463
2744	64	0.640	0.5060
4096	512	0.320	0.6134
5832	4096	0.160	0.7343
8000	32768	0.080	0.8565
10648	262144	0.040	0.9450
13824	2097152	0.020	1.0109
17576	16777216	0.010	1.0447

Table 2

Maximum multiplication constant with uncertainty for solution of ^{235}U

Mass ^{235}U	780 g ^{235}U	780 g ^{235}U	1.3kg ^{235}U	1.3kg ^{235}U
State	Solution	Solution	Solution	Particles
Geometry	Spherical	Cubic	Cubic	Cubic
Water Volume (mL)	$K_{eff} + 3\sigma$	$K_{eff} + 3\sigma$	$K_{eff} + 3\sigma$	$K_{eff} + 3\sigma$
1000	0.6220	0.6323	0.6523	0.4550
1728	0.7257	0.7322	0.7470	0.4463
2744	0.8097	0.8123	0.8364	0.5060
4096	0.8819	0.8837	0.9125	0.6134
5832	0.9372	0.9280	0.9694	0.7343
8000	0.9728	0.9645	1.0245	0.8565
10648	0.9973	0.9790	1.0589	0.9450
13824	1.0036	0.9829	1.0776	1.0109
17576	0.9993	0.9990	1.0956	1.0447
21952	0.9771	0.9596	1.0897	
27000	0.9528	0.9534	1.0834	
42875	0.8610	0.8461	1.0284	
64000	0.7556	0.7419	0.9422	

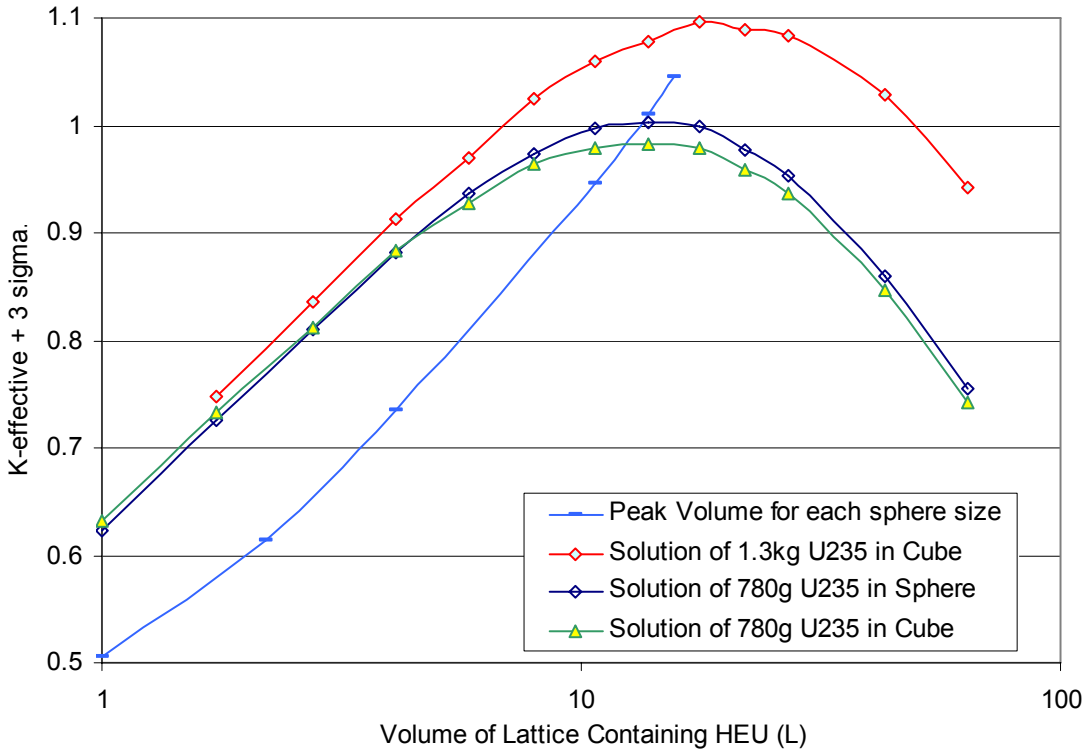

Figure 2 : K-effective +3σ for up to 1.3kg ^{235}U as a function of volume

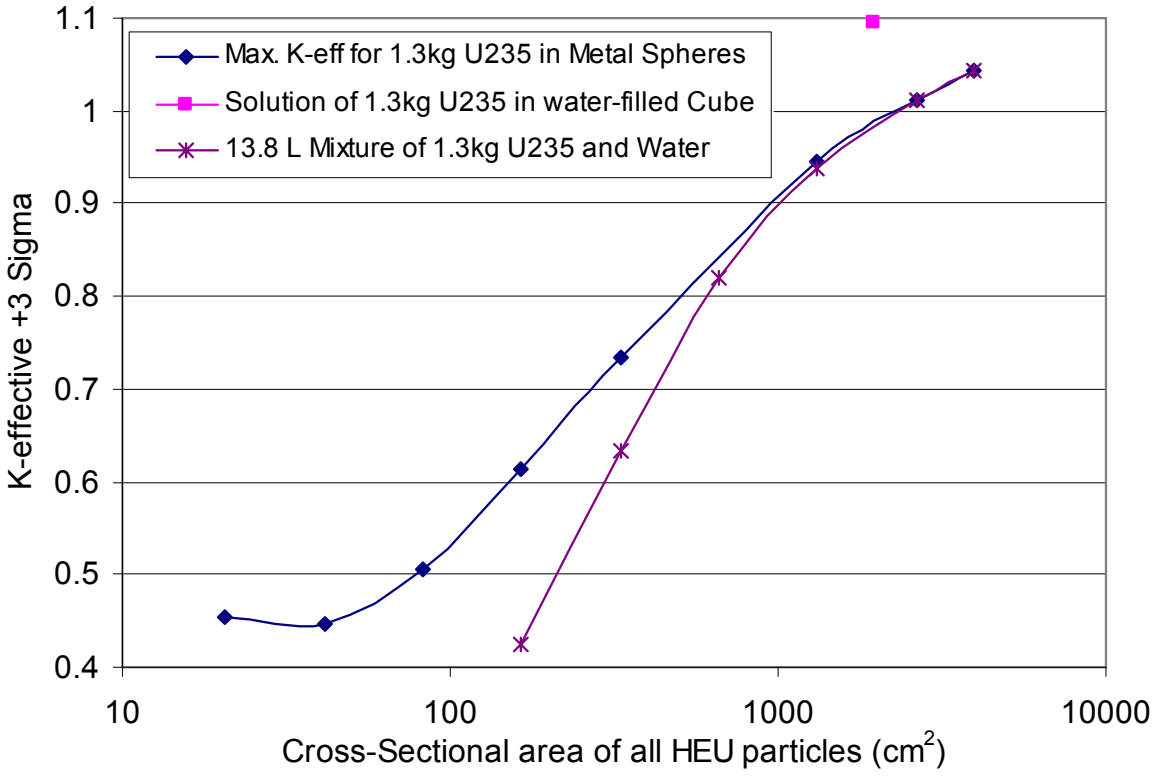

Figure 3 : K-effective +3σ for ^{235}U as a function of macroscopic cross-sectional area of HEU particles in water-filled cube