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VERIFICATION SUITE FOR THE APPLICATION OF THE LIMITING SURFACE DENSITY METHOD TO ARRAYS OF 9975 PACKAGES

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ABSTRACT

Criticality Safety Engineering at the Savannah River Site's K-Area Complex seeks to reduce the computational burden of analysis for the K-Area Complex. By applying an adapted Limiting Surface Density hand calculation method, the need for lengthy detailed Monte Carlo models of the storage arrays may be reduced or eliminated. After promising initial results using models of idealized but heterogeneous packages, application of the method to large arrays of highly heterogeneous 9975 shipping packages is attempted. A SCALE KENO-VI model of the 9975 package, with true dimensions and radial and axial heterogeneity is developed. A suite of array arrangements representing both arrays with equal number of packages per side (package-cubic) and arrays with unequal number of packages per side (non-cubic) are evaluated in SCALE. The cubic arrangements are used to derive the 9975-specific material and geometry constants for the LSD method. These constants are then used to implement the LSD method and compare the LSD- predicted critical mass per package to SCALE-predicted critical mass per package. Sensitivities to certain material and geometric nuances are evaluated for inclusion in the verification suite. Finally, the ability of the method to predict neutron multiplication changes induced by fissile mass changes is examined as a segue for evaluating credible abnormal conditions with the LSD method.

Key Words: **storage array, heterogeneous package, Limiting Surface Density method**

1. INTRODUCTION

In recent years the former K-reactor building of the K-area Complex (KAC) at the Savannah River Site has taken on a key stewardship role in the interim storage of plutonium. Excess non-pit plutonium from around the Department of Energy complex has been consolidated at this hardened, defensible structure. It has been observed that the vast majority of containers within the KAC are of a singular design, the 9975. Furthermore, these containers are stored in large, regular arrays.

Considering the computational burden of explicitly modeling and then evaluating arrays of fissile packages within storage facility, interest has arisen in the potential to use a conventional hand calculation method to perform criticality safety analyses. Such a method would help reduce the task of running many long-duration detailed Monte Carlo models provided that the hand calculation method could be shown to be acceptably equivalent.

Recent work (Ref. 1) has shown the potential for extension of the Limiting Surface Density (LSD) method to fill this need. The mathematical basis of the original LSD method (Ref. 2) is that the user can find some mass of fissile material which if loaded into each storage unit, would result in the array being exactly critical. The user would then need to correlate that mass to the surface density of fissile material as the number of units and their spacing changed. The original LSD method assumes the surface density linearly correlates to critical mass per unit based on the geometry of the array and based on the material properties of the fissile material being stored. The intersection of these two correlations is the limiting surface density and correspondingly the limiting fissile mass per package. Require fissile content per package to be below that mass and the array will be subcritical.

When the LSD method was originally developed, it was based on the idealized scenario of bare spherical fissile metal units at the center of cubic air-spaced packages, arranged in a cubic array, with the array reflected by water. Few realistic storage arrays would have such a configuration. Limited modifications were made early to the original LSD method to allow for non-cubic arrays greater than 64 units, concrete reflection, and fissile metal cylinders. Still the basis of the method remained constant – bare metal unit in an air spaced array. The method needs to be extended from its original concept of cubic arrays of cubic packages of metal spheres to more realistic scenarios. The work in Ref. 1 began this extension by showing the method could perform acceptably well, compared to Monte Carlo models, for a highly heterogeneous package. However, even in that work the heterogeneous package was geometrically idealized so that it could be placed into a cubic unit. This "cubic" 9975 removed much of the vertical space and associated interstitial material that was not occupied by fissile material. The radial geometry was retained and the results showed promise.

This work seeks to further extend the LSD method to arrays of 9975 shipping packages wherein the package modeled is as near the actual geometry as practical. Application of the LSD method to arrays of this package can then be examined. However, to verify that the method is applicable for actual KAC criticality safety evaluations, it must be shown to be acceptably equivalent to computational modeling. As such, effort was made to establish a suite of Monte Carlo cases of 9975 package arrays. The results of these Monte Carlo models are then compared to calculational results produced from applying the LSD method to the same suite of array calculations. Preliminary work has also been done to establish normal conditions for the arrays and to examine the ability of the method to handle reactivity perturbations similar to what may be experienced in credible abnormal scenarios. Adjustment to the method was made to account for non-linear response of surface density to fissile mass; however, detailed methodological discussions are reserved for a separate publication.

2. DEVELOPMENT OF A SIMPLIFIED 9975 MODEL

Early work (Ref. 1) employed an idealized model of the 9975 where the radial geometry was preserved but much of the vertical space not occupied by fissile material was removed. This resulted in modeling a "cubic" 9975 with a height to diameter ratio of 1. This type of package more closely resembled the original LSD method's conceptual application basis and allowed for evaluation of the method for a heterogeneous package without considering the potential need for significant modification of the method.

In reality, the 9975 shipping package is a 35-gallon outer drum with a height to diameter ratio of about 2 and contains numerous geometric nuances making it highly heterogeneous (Fig. 1, left). The most recent documentation of the 9975 design is included in Ref. 4. The fissile material may be assumed to be sitting in the innermost container and to be radially centered but is not vertically centered. There are layers of lead, aluminum, stainless steel, and Celotex which vary radially with height. Axially there are transitions between container tops and bottoms, bolts, lids, spacers, plates, impact absorbers, etc.

In support of this work, and other KAC applications, a simplified and internally pedigreed model of the 9975 package was developed in the SCALE 6.1 code system, particularly for KENO-VI using the ENDF-B Version 7 238-group cross section set. The geometry was simplified to model containers as cylinders capped with plates. Curved surfaces on the bottom of inner vessels were recast, preserving material properties, into flats. The radial dimensions were maintained and the axial geometry, though simplified, maintained material properties and overall dimensions. The simplified model, including a spherical fissile mass, is shown on the right of Fig. 1.

At the heart of the container is the fissile material "product can" which is not explicitly shown in Fig. 1, left but is modeled in Fig. 1, right. The fissile material is assumed to always be placed inside some container independent of the 9975 design which fits into the primary containment vessel. For this analysis, a composite product can model was created to be reasonably bounding for the expected range of product cans that are received.

This simplified model, which was subjected to multiple rounds of peer review to ensure its quality, is important to KAC criticality safety analyses. Provided there is no design change to the 9975 package, the user can simply take the standardized model, load the primary container with whatever fissile material is desired to be modeled and arrange any number of units as the analyst sees fit. Therefore this model has been internally retained for use (Ref. 5) and has applications beyond verification of the LSD method.

3. ARRAY CALCULATION SUITE

In order to verify the acceptable application of the LSD method, or any empirical approach, a suite of Monte Carlo results is necessary to compare against predictions from the alternative method for the same conditions.

SCALE calculations were performed for a variety of arrays which varied the number of 9975 units per side and the spacing between the units. These included arrays with width x depth x height unit dimensions of 4x4x4, 5x5x5, 6x6x6, 7x7x7, 8x8x8, 9x9x9, 10x10x10, 2x20x1, 2x30x1, 2x20x2, 2x20x3, 4x20x3, and 5x5x3. The number of units per side was chosen for various reasons. The arrays with equal number of units each mimic the original application basis of the LSD method for cubic arrays even though the package diameters are half their height. The rectangular arrays resemble some of the potential arrangements that would be seen in KAC, and are used to test the method with varying shapes and sizes. Center to center spacing choices were 46.6 cm (edge-to-edge contact of containers), 60, 70, 80, 90, 120, and 150 cm. The latter two cases are chosen to be arbitrarily large and it is unlikely that spacings of 120 cm or more would be seen in actual storage application. Note that due to the geometry of the 9975, arrays with spacing less than 90 cm retained the vertical height of the 9975 as the only axial spacing. Arrays with spacing greater than 90 cm introduced vertical spaces between the packages. These choices were due to the nominal height of the 9975 being approximately 88.5 cm.

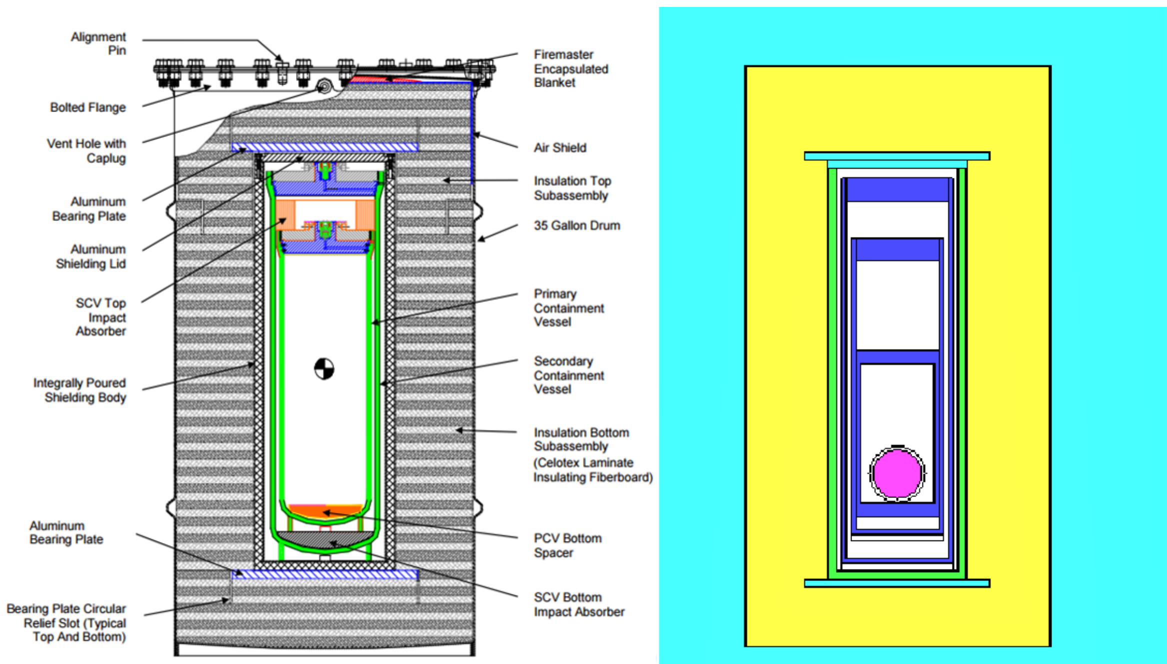

Figure 1. Elevation view of the 9975 Shipping Package Actual Geometry (l) and Simplified Geometry (r)

An unclassified look into the KAC storage area is shown in Fig. 2. The 9975 packages, loaded four to a pallet are stacked, no more than 3 high, and kept in monitored storage. The old K-reactor control rod guide can be seen in the background sitting atop the reactor vessel head which is nearly flush with the floor.

Since one of the primary functions of the LSD method is to determine the mass of the fissile material per package that results in a critical array, the calculations for the above scenarios were to determine the mass of alpha-phase ^{239}Pu metal required per package to result in a critical array. The results are presented below. The 2-sigma uncertainty associated with the values in Table 1 is ~20 g.

Figure 2. 9975 Shipping Package Storage Array

Table 1. SCALE Computed Mass (g) Per Package for a Critical Array**.**

	Center-to-Center Spacing, all in cm.						
Array dimensions	46.6	60	70	80	90	120	150
4x4x4	7798	7923	7975	8028	8058	8167	8231
5x5x5	7749	7871	7930	7975	8009	8134	8198
6x6x6	7725	7834	7880	7925	7969	8090	8171
7x7x7	7698	7804	7856	7896	7929	8061	8144
8x8x8	7691	7787	7829	7875	7913	8039	8126
9x9x9	7666	7763	7814	7853	7886	8014	8094
10x10x10	7668	7760	7802	7837	7865	7984	8081
2x20x1	7876	8028	8093	8137	8174	8245	8304
2x30x1	7869	8020	8088	8145	8178	8250	8302
2x20x2	7847	7982	8045	8086	8123	8217	8259
2x20x3	7843	7961	8027	8066	8093	8200	8255
4x20x3	7737	7862	7922	7967	8009	8128	8198
5x5x3	7761	7892	7958	7999	8041	8154	8223

The array calculations selected above assume all material and geometric properties of the individual containers are constant, except of course the fissile mass. During the development of the verification suite, the question was raised as to whether changes in the individual package geometry or material would affect the resulting allowable fissile mass per package. Changes in the individual packages that do not affect the array spacing or unit dimensions would not directly impact the surface density. Therefore changes in the non-fissile material or geometry of the individual packages would have only the second order effect of potentially altering the neutronic coupling between the packages. Since the LSD method employs correlations based on the geometric and material properties of the array to arrive at a limiting fissile mass per package, variation in those properties could potentially alter the correlation coefficients.

Geometric and material properties of the 9975 package were examined per expected package variations. The properties qualitatively determined to be most likely to impact neutronic interaction of the packages were Celotex density (material property) and primary container wall thickness (geometric property).

3.1. CELOTEX DENSITY SENSITIVITY

Monte Carlo models were executed to determine the sensitivity to Celotex density over the range of an arbitrarily small minimum of 0.01 g/cm^3 up to a density of 0.31 g/cm^3. The effect was evaluated for bare (i.e. no reflection) surrounded by air, concrete floor (i.e. partial reflection) with air on all other sides, and concrete cuboid (i.e. thick reflection) on all sides. The results with 2-sigma uncertainty bars (Fig. 3) show the change in k-effective for a 9975 package containing the nominal limit of 4400 g Pu-239 metal is less than 0.01Δk over the whole range of expected density variations. Celotex density variations were therefore not included in the verification suite, A density of 0.31 g/cm^{3}was used for all cases.

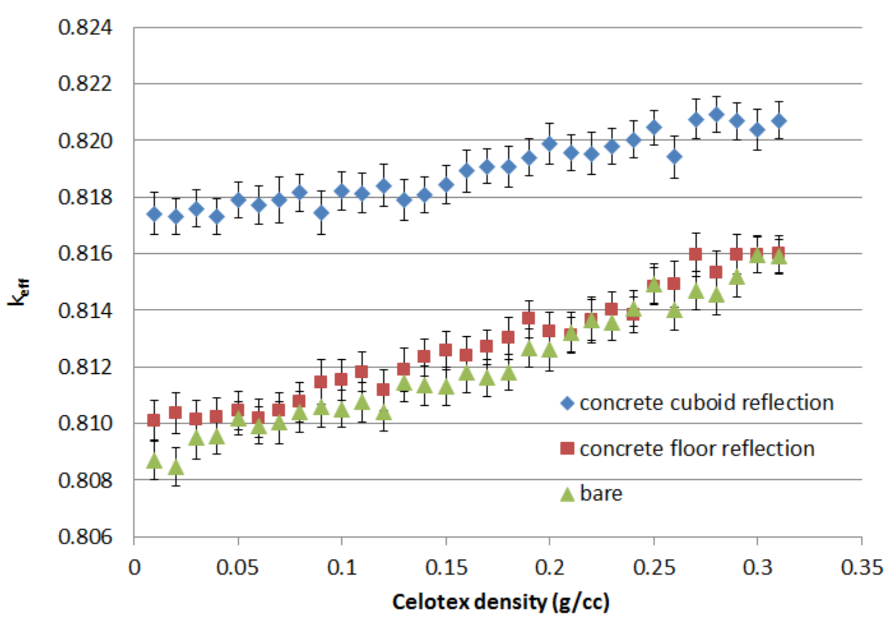

Figure 3. Effect of Celotex Density on Multiplication Factor

3.2. CONTAINER WALL THICKNESS SENSITIVITY

Monte Carlo models were executed to determine the sensitivity to fissile material container wall thickness over the range of an arbitrarily small minimum of 0.1 cm up to ~0.766 cm which would result in the container wall extending to the inner wall of the primary containment vessel. The effect was evaluated for an infinite planar array with top and bottom reflection. The results with 2-sigma uncertainty bars (Fig. 4) showed the change in k-effective for a 9975 package containing the nominal limit of 4400 g Pu-239 metal were less than 0.012Δk over the whole range of variations and less than 0.01Δk over the expected variations from ~0.4 cm to ~0.766 cm. Fissile material container wall thickness variations were therefore not included in the verification suite. A primary container wall thickness of 0.61 cm was used.

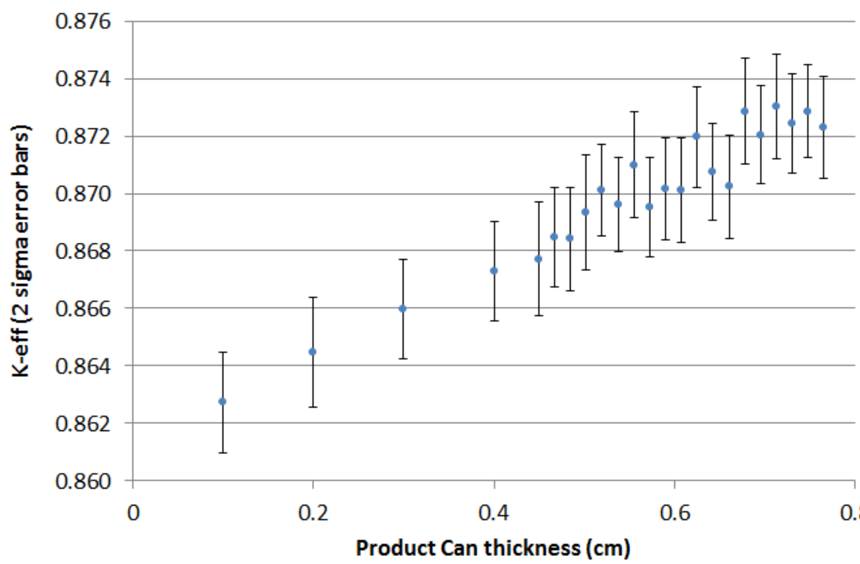

Figure 4. Effect of Fissile Material Container Wall Thickness on Multiplication Factor

4. IMPLEMENTING THE LSD METHOD

The classic LSD method assumes linear correlation between the fissile mass and the surface density given the center-to-center spacing of the packages $2a_n$ and the total number of packages N for the cubic array. The practical equation of the LSD method from which the limiting fissile mass per container is set comes from equating two correlations of the surface density:

$$\frac{m_c n}{(2a_n)^2}\left(1-\frac{c}{\sqrt{N}}\right)^2 = c_2(m_c - m_o) \qquad (1)$$

Here m_c is the limiting fissile mass per package that the user seeks and m_o is the fissile mass which would make a single unreflected package critical. There user must determine the values of c, which is called the geometric constant, and c_2, which is called the material constant. The user then sets the spacing and number of packages for the array. The m_o value may be determined from Monte Carlo modeling or from experiment.

4.1. DETERMINATION OF CONSTANTS

The constant c depends on knowledge of the array's buckling behavior. The LSD is sometimes also called the NB_N method because

$$c = \sqrt{\frac{4\lambda_{array}{}^2 NB_N^2}{3\pi^2}} \qquad (2)$$

where NB_N is the number of units times the array buckling. This value can be determined with analytical effort. Thomas also indicated a constant value of 0.55 could be used provided the user was aware of the ± 0.18 uncertainty on that value. Analytical work, not shown, for this application determined c = 0.44 for the 9975 array, which is in agreement with 0.55± 0.18.

The material constant c_2 is somewhat less analytically intensive to compute. The Monte Carlo modeling provided the fissile mass per package that would result in a critical array for a number of cubic arrays at various spacing. To determine c_2 per the classic LSD method, these masses and the number of units in the vertical direction are projected onto the horizontal plane of the array. The unit spacing and number of units in the x and y directions (which are the same for cubic arrays) determine the surface area of this plane. This yields a surface density value for each arrangement. The surface density verses fissile mass per package resulting in a critical array is plotted (Fig. 5).

4.2. TREATING THE NON-LINEAR RESPONSE

When Thomas derived the LSD method, cubic arrays of bare cubic sphere were shown experimentally to indeed have a linear response between surface density and the mass per package that made the array critical. Even the idealized cubic 9975 (Ref. 1) had an acceptably linear response for the range of conditions evaluated at that time. The non-cubic, highly heterogeneous 9975 package did not have a linear response as seen in Figure 5.

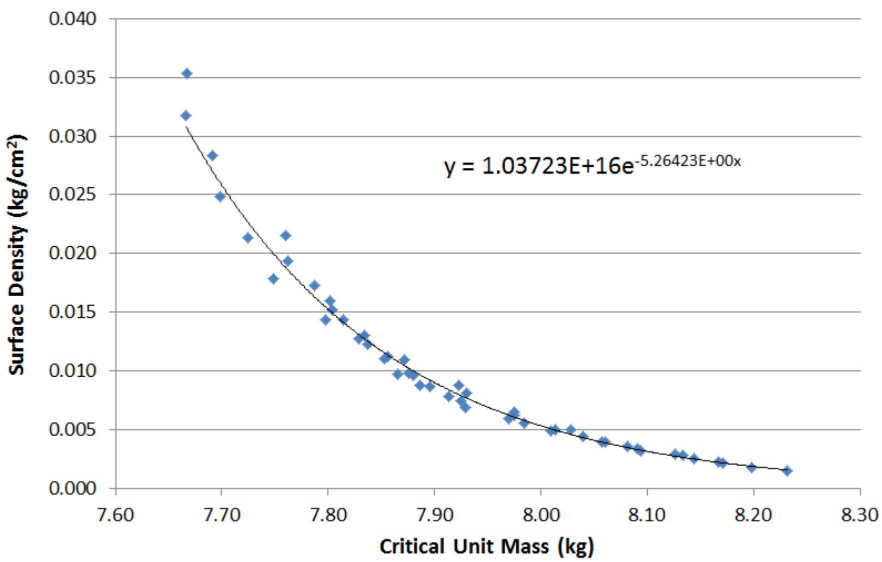

Figure 5. Surface Density Versus Critical Mass Per Unit for Cubic Arrays

However, the surface density response was very well characterized by an exponential decay function

$$\sigma(m_c) = 1.03723x10^{16}e^{-5.26423m_c} \tag{3}$$

The precision of 6 significant figures had to be retained because of the sensitivity of the exponential function to the values. It was chosen to use this form of surface density equated to the geometric (left hand side) of Equation 1 in lieu of the linear correlation. This has the benefit of decoupling the method from computationally or experimentally determining the fissile mass for a single bare package. Expanded methodological discussion is reserved for a separate publication.

4.3 ARRAY CALCULATIONS AND COMPARISON

The cubic arrays modeled in SCALE also had the critical fissile mass per package computed using the LSD method with the constants and correlations determined above. These results, shown in Table 2, compared remarkably well with the SCALE results. The LSD-computed masses for the cubic arrangements were shown to be within -0.28% to +0.46% of the SCALE computed masses.

Table 2. LSD Computed Mass (g) Per Package for a Critical Array, Cubic Arrangements

	Center-to-Center Spacing, all in cm.						
Array dimensions	46.6	60	70	80	90	120	150
4x4x4	7832	7926	7983	8033	8076	8183	8266
5x5x5	7785	7878	7935	7985	8029	8136	8218
6x6x6	7747	7841	7898	7948	7991	8098	8181
7x7x7	7716	7810	7867	7917	7960	8067	8150
8x8x8	7690	7784	7841	7890	7934	8041	8124
9x9x9	7667	7761	7818	7867	7911	8018	8100
10x10x10	7646	7740	7797	7847	7890	7997	8080

Ref. 3 provides instructions on how to adjust the LSD method for non-cubic air-spaced arrays. Essentially, an adjustment factor is derived based upon known results from a non-cubic reference array. This adjustment factor accounts for the additional leakage introduced by the non-cubic arrangement of packages. This adjustment is based on the shape factor, R, which is defined as:

$$R = \frac{\sqrt[3]{N}}{3}\left(\frac{1}{n_x} + \frac{1}{n_y} + \frac{1}{n_z}\right) \tag{4}$$

There are two complications to this. First, the common reference non-cubic system is also an array of bare metal fissile masses and the adjustment derived is empirically correlated. Second, the adjustment is applied to the material constant derived for the linear correlation of surface density, which was determined not applicable for this work.

At the time of this authorship, no useful correlation has been determined for adjusting the non-linear response surface density correlation to account for non-cubic array neutron leakage. However, because the 9975 is by design a very low leakage package, LSD calculation results for non-cubic arrays agree very well with SCALE results without adjustment for array shape. Based on this verification suite, the LSD method with these specific constants should only be applied to arrays of 9975 packages where R is $\leq$ 2, and N $\geq$ 40. As a practical matter, these are not significant restrictions. The non-cubic array results, shown in Table 3, have a -1.33% to +0.77% difference with the Monte Carlo models. This is higher than the cubic array differences; however it is very good agreement for an empirical method. The user may choose to compensate for this with an engineering margin allowance.

Table 3. LSD Computed Mass (g) Per Package for a Critical Array, Non-Cubic Arrangements

	Center-to-Center Spacing, all in cm.						
Array dimensions	46.6	60	70	80	90	120	150
2x20x1	7867	7961	8018	8067	8111	8218	8301
2x30x1	7837	7930	7988	8037	8081	8188	8271
2x20x2	7816	7910	7967	8016	8060	8167	8250
2x20x3	7787	7881	7938	7988	8032	8138	8221
4x20x3	7740	7834	7891	7940	7984	8091	8174
5x5x3	7821	7914	7972	8021	8065	8172	8254

4.4. MULTIPLICATION PERTURBATION STUDIES

This verification suite demonstrates that the LSD method, given 9975 specific array constants, reliably predicts a limiting fissile mass per package for many practical array arrangements. This would alleviate the computational burden placed on the analyst to develop, potentially many, detailed models of specific or bounding arrays to establish subcriticality limits. However, the masses derived up to this point are critical values for an otherwise normal condition of storage. To be a fully usable alternative method to Monte Carlo analysis, there must also be a way to apply the LSD method to estimate system parameters for normal and credible abnormal conditions. Ideally, the user would then apply this knowledge of the effects of abnormal conditions to determine an appropriate mass limit per package for the storage array.

At the time of this authorship, some preliminary work has been done on applying the method to system changes that might encompass credible abnormal conditions. An example is presented here where 0.5 and 1.0 kg changes in the mass are introduced from a 4.4 kg base case. The fissile material is assumed to be alpha phase ^{239}Pu metal. The base case array was chosen to be 10x14x3, which has an m_c = 7.703 kg determined from LSD method calculations. The common hand calculation formula $k_{eff} = (\frac{m}{m_c})^{1/3}$ is applied to cases of m = 3.4, 3.9, 4.4 (nominal), 4.9, and 5.4 kg Pu-239. The resulting k_{eff} values are presented in Table 4 and are compared to their SCALE values. There is good agreement between the methods, though the LSD results consistently under-predicted the SCALE model results.

Table 4. Multiplication Difference for Varying Unit Mass in a 10x14x3 Array of 9975 Packages

Unit Mass (kg)	LSD-Based k_{eff}	SCALE k_{eff}	Δ_{keff} Difference
3.4	0.761	0.768	-0.007
3.9	0.797	0.804	-0.007
4.4 (nominal)	0.830	0.836	-0.006
4.9	0.860	0.866	-0.006
5.4	0.888	0.894	-0.005
7.703	1.000	1.002	-0.002

5. CONCLUSIONS AND FUTURE WORK

This extension of the LSD Method may replace complicated and time-consuming Monte Carlo modeling with simple, short, hand calculation method analyses. It provides quick and useful parameter study results for 9975 shipping package arrays. The adequacy of the method to analyze credible abnormal conditions will be further evaluated. There is also interest in extending the method to other fissile species, and to plutonium oxide.

The authors look to publish the method with its non-linear formulation in a peer-reviewed journal so that the criticality safety community can have common access to the tool.

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