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# Limiting Surface Density Method Application to Large Arrays of Highly Heterogeneous Shipping Packages

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## INTRODUCTION

This work represents the initial phase of an overall effort to reduce the analytical burdens and increase flexibility of the K-Area Complex which provides stewardship of special nuclear materials. A hand calculation method is being extended to apply to large arrays of highly heterogeneous shipping packages. This method has the potential to quickly determine the impact of perturbations on the array.

The objective of the particular work reported herein is twofold. First, the analytical results upon which the Limiting Surface Density (LSD) method was originally based were reproduced using the modern computational code KENO-VI in SCALE 6.1. The second objective was to test the classic LSD method on storage of large arrays of highly heterogeneous shipping packages. Specifically, the objective is to determine if the method is adequate for existing arrays of 9975 shipping packages with fissile metal content. The classic LSD method was derived for simple packages of bare spheres of fissile material centered in an air filled cubic unit cell and placed in a cubic-pitch water-reflected array, the properties of which could then be translated to more practical array and package geometries. The classic method did not address highly heterogeneous packages which are close to neutronically decoupled from other identical packages. However, standard guidance (Ref. 1) implies there is some allowance for heterogeneity. The material based array constant (called  $c_2$ ) as defined by the classic method is determined specifically for the packages in question.

## DESCRIPTION OF THE WORK

The LSD method is documented in Ref. 2 through 5. Storage array guidance based on this method is included in Ref. 1, which states that the method may be used for units surrounded by up to one-half inch of steel, which is substantially less heterogeneous (less interstitial material) than the packages in question. The 9975 shipping package arrays differ from the applicability of the original LSD method because 1) the units are not cubic, but have height about twice the horizontal dimension, 2) the fissile material is not vertically centered in the unit, and 3) there is a large amount of interstitial material between the fissile material and the neighboring material. The LSD method was derived based on the number of units in the array,  $N$ , and the array buckling,  $B_N^2$  where the buckling was derived from basic nuclear physics for a parallelepiped that was assumed to have equal sides of length  $2a_n$  such that:

$$NB_N^2 = \frac{n3\pi^2}{(2a_n)^2} \left( 1 - \sqrt{\frac{4\lambda_{array}^2}{N3\pi^2} NB_N^2} \right)^2 \quad (1)$$

The method asserts that  $c = \sqrt{\frac{4\lambda_{array}^2}{3\pi^2} NB_N^2}$  is a constant independent of material and dependent only on geometry. Ref. 4 gives  $c = 0.312 \pm 0.001$ , however later work (Ref. 2 and 5) gives  $c = 0.55 \pm 0.18$  allowing for variation in  $c$  values. This is compensated for by the fact that the value of  $c$  has a small effect on the results given by the LSD method. The following relationship from the original method allows the user to find a limiting mass of the individual packages which would make the array critical by equating material based and geometry based forms of the surface density:

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

Where  $m_o$  is the unreflected critical mass of an individual package,  $c_2$  is the array constant based on material properties, and  $m_c$  is the mass of an individual package that would make the array critical.

## Examination of Geometric and Material Dependence Using a Modern Code System

Ref. 4 was issued in 1971 so the first step in this work was to reexamine the dependencies of the LSD method using a modern neutronics code with modern cross sections from ENDF-VII. Appendix B of Ref. 4 presented criticality calculations for various sizes of arrays with various unit spacing for three common fissile materials – U-233, U-235, and Pu-239. The critical dimensions were then used to compute the buckling and extrapolation distance values, which were characteristic of the arrays. This work is reproduced as a reference point. The Pu-239 cases from Ref. 4 Appendix B are tested in the following steps.

Using the material densities and geometries given in Table B-1 of Ref. 4, the ratio of the critical radius of unit material to the critical radius of a bare unreflected sphere of material ( $r_c/r_o$ ) was calculated for each combination of array dimension and unit spacing using SCALE with the 238-group ENDF-VII cross sections. The critical mass of a bare sphere is defined  $m_o = \rho \frac{4}{3} \pi r_o^3$ . The critical mass of the various units are then  $m_c = \rho \frac{4}{3} \pi r_c^3$ .

The description of obtaining  $NB_N^2$  is lacking in the literature. Therefore,  $NB_N^2$  is back calculated from the classic LSD method. Since all calculated arrays are critical with units of assumed material density and radius  $r_c$ , the mass of each can be calculated. The LSD method defines the surface density in terms of material as  $\sigma(m_c) = c_2(m_c - m_o)$ . The data for each material can be analyzed as surface density versus mass where  $\sigma(a_n) = nm_c/(2a_n)^2$ . The slope of the linear least squares regression fit of this data is calculated; this is  $c_2$ . Using the definition  $\sigma(m_c) \equiv \frac{m_c}{3\pi^2} NB_N^2 = c_2(m_c - m_o)$  and the  $NB_N^2$  value is calculated from this relationship.

### Cubic Test Model of the 9975 Package

The 9975 is a DOT Type B shipping package which is used for the storage and transport of special nuclear materials across the Department of Energy complex and internationally. On the surface, i.e. the outermost container, it is a stainless steel 35-gallon drum with a height to diameter ratio of roughly 2. The shipping package contains nested dunnages and canisters of Celotex, aluminum, lead, and stainless steel. At the heart of the highly heterogeneous system is a stainless steel type 3013 can inside of which is a mass of nuclear material which is offset from the vertical center of the package. For this work, the material is assumed to be 100% Pu-239 metal.

The classic LSD method is based on cubic arrays, so a cubic representation of the 9975, modified to a cylinder of height to diameter equal to 1, was made. To do so, a portion of the interstitial materials above the fissile material is removed. The radii are preserved and the geometry of the 3013 can is preserved. Fig. 1 shows the cubic 9975 wherein the height to diameter ratio has now been adjusted to 1. Any results are consequently conservative because this geometry artificially places the fissile materials closer together than they would be in reality. The dark region at the lower center of Fig. 1 is the fissile material, modeled as a cylinder of height to diameter equal to 1.

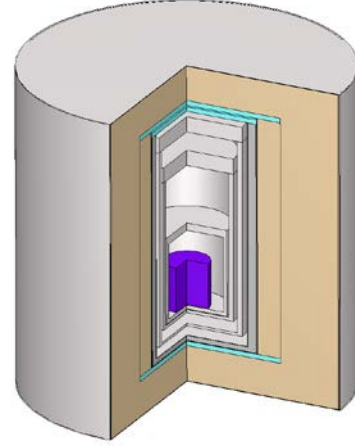


Fig. 1. Cubic representation of 9975 shipping package.

### RESULTS

The critical arrays of Pu-239 metal spheres at various spacings and array dimensions were recreated from Ref. 4. Table I highlights some of the comparisons between the reference data and the calculations carried out with SCALE 6.1 in this work. The material constant  $c_2$  for Pu-239 agreed well with the reference value as did the calculated critical radius of an unreflected metal sphere  $r_o$ . Table B1 of Ref. 4 reported the unit radius that would make the array critical,  $r_c$ , divided by the unreflected single unit critical radius,  $r_o$ . These values also agreed well with the reference, being on average less than 0.9% difference.  $NB_N^2$  were calculated and were similar to the Ref. 4 values but only within an average of 7.6%. The computed geometric constant  $c$  for the Pu-239 cases was  $0.232 \pm 0.294$ , which overlaps with the  $0.55 \pm 0.18$  range but not the  $c = 0.312 \pm 0.001$  range. The conclusion from this is that the modern code reasonably reproduced the original results and is able to be used with the LSD method, and in particular to calculate a new  $c_2$  for the 9975 shipping package.

Table I: Comparison of Calculated Values to Reference		
Parameter	Reference	Calculated
Critical Radius of Bare Sphere (cm), $r_o$	4.90	4.97
Difference in $r_c/r_o$ (%)	$-0.853 \pm 0.651$	
Material Constant, $c_2$	-0.00434	-0.00444
Geometric Constant, $c$	$0.55 \pm 0.18$	$0.232 \pm 0.293$
Difference in $NB_N^2$ (%)	$7.626 \pm 3.94$	

To test the 9975 model, at the center of each package is a cylindrical mass of Pu-239 metal at a density of 19.84

$\text{g/cm}^3$ . The mass is adjusted for various cases but the height to diameter ratio is preserved at 1. The upper limit of mass is that which would make a single unreflected package in air critical and that was calculated to be 8.64 kg.

The cubic 9975 model was placed into a cubic water reflected array and the mass adjusted to be critical for various cubic array sizes (4, 6, 8, and 10 units per side) and for various center-to-center spacing ranging from 46.5987 cm (edges in contact) to 150 cm. This data when plotted (Fig. 2) shows a very steep slope. This data is compared to the bare sphere Pu-239 array data reproduced from Ref. 4 using KENO-VI (series “Pu239”). For comparison, the data from Ref. 4 itself is also plotted (series “Pu239 – Ref”) which demonstrates that the modern code is reproducing well the original calculations. The data sets have about the same range of surface density variation, but the 9975 data has a mass range of only about  $\sim 1.1$  kg whereas the bare sphere spanned approximately 8 kg.

The bare sphere Pu-239 data is essentially linear as was the conclusion when the LSD method was derived. Focusing on the 9975 data in Fig. 3 shows that data only becomes linear at lower surface densities. Overall the data is more akin to an exponential or power decay. The data implies that the surface density of a critical array is asymptotic as the mass drops below about 7.4 kg of Pu-239.

According to the classic LSD method, the slope of the linear fit of the 9975 data is taken as the material constant,  $c_2$ . If all of the calculated 9975 data is used, this value is 0.02017. However using only the linear region data points, neglecting the highest 8 surface density cases (of 28 cases total), only reduces this value to 0.01511.

Nine “practical arrays” were modeled as test cases to use the classic LSD method and the latter value of  $c_2$ . They were  $2 \times 20 \times 2$ ,  $2 \times 30 \times 3$  and  $4 \times 30 \times 3$  arrays with center-to-center spacing of 46.5987, 55, and 70 cm. The typical geometric constant  $c$  of value of 0.55 was used, but this has little influence on the result for large arrays. The calculations were adjusted for the non-cubic shape of the array per the shape factor procedure in Reference 2. The results are in Table II. Preferred results, i.e. those which minimize the difference between the LSD method and SCALE calculations, were achieved with  $c_2$  equal to 0.01511. This produced a value on the package mass that was lower than the critical package mass calculated using SCALE for only two cases. It is fair to note though that the non-conservative LSD results are less than  $\sim 0.1$  kg different from the SCALE results. For this work, a conservative result is defined as one in which the LSD method produced

the same or lower mass limit per package than computational modeling in KENO.

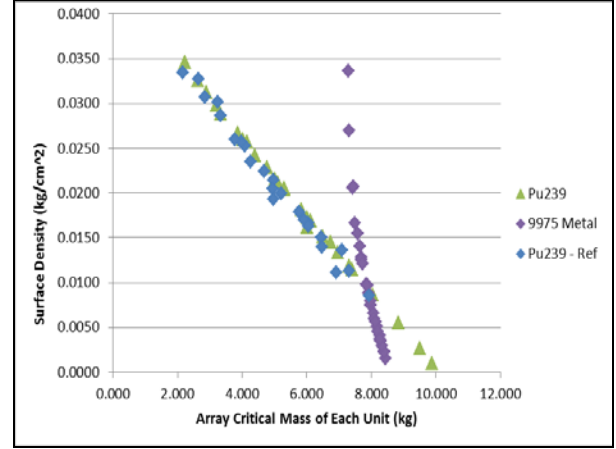


Fig. 2. Surface density versus mass for reference Pu-239 sphere and 9975 with Pu-239 metal.

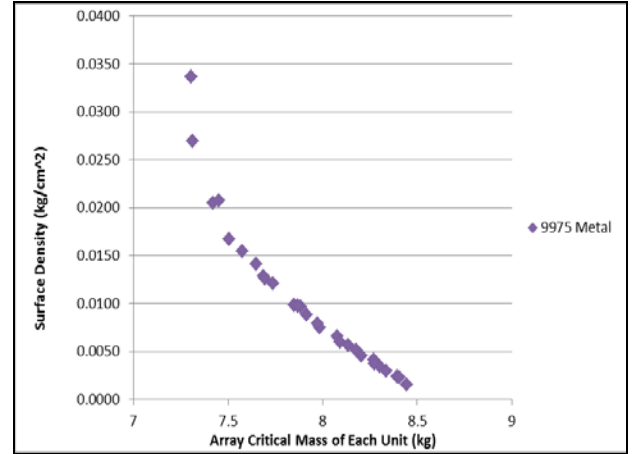


Fig. 3. Surface density versus mass for 9975 with Pu-239 metal data only (detail).

Array Dimensions (units in x,x,z)	Center-to-Center Spacing (cm)	SCALE Calculated Unit Mass for Critical Array (kg)	LSD Calculated Unit Mass for Critical Array (kg)
2x20x2	46.5987	7.90	7.99
2x20x2	55	8.08	8.17
2x20x2	70	8.40	8.34
2x30x3	46.5987	7.82	7.83
2x30x3	55	8.00	8.05
2x30x3	70	8.18	8.26
4x30x3	46.5987	7.75	7.53
4x30x3	55	7.78	7.82

4x30x3	70	8.01	8.11
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The LSD method did not produce dramatically conservative results over the SCALE calculations but in fact were within ~0.1 kg unit mass implying the methods may be applied to this an array of highly heterogeneous packages with some engineering conservatism applied. What was discovered is that due to the interstitial material and spacing, at least 7.4 kg fissile material per package is required to achieve a critical arrangement. Between about 7.4 kg and 8.693 kg, the mass can compensate for the variations in spacing. Below about 7.4 kg, the array would have to asymptotically increase in size to approach criticality. The result is not wholly unexpected as the 9975 has a substantial amount of shielding and interstitial materials between the fissile material and the outer surface.

This work will proceed for the facility in question. The next phase will work to include a more realistic 9975 model and evaluate scenarios replacing Pu-239 with U-235 and the metals with Pu and U oxides. This will be followed by investigating the effect of upsets such as fire damage or flooding. Finding a minimum critical fissile mass per package, similar to what was done in this work, would be expected under these future variations.

## NOMENCLATURE

$N$  = number of units in array

$n$  = number of units per side of the array

$B_N$  = array buckling

$a_n$  = half length of cubic array side

$c$  = geometric array constant

$c_2$  = material array constant

$\lambda$  = extrapolation distance

$\rho$  = density

$m$  = critical mass

$r$  = critical radius

*Subscripts and Superscripts*

$o$  = unreflected single critical unit

$c$  = single unit of critical array

## REFERENCES

1. A. GARCIA, et. al., "Nuclear Criticality Safety in Operations with Fissionable Material Outside Reactors," ANSI/ANS-8.1-1998, American Nuclear Society (1998).
2. D. BOWEN and R. BUSCH, "Hand Calculation Methods for Criticality Safety – A Primer", LA-14244-M, Los Alamos National Laboratory, (2006)
3. J. THOMAS, "Uranium Metal Criticality, Monte Carlo Calculations, and Nuclear Criticality Safety," Y-CDC-7, Oak Ridge National Laboratory (1970)

4. J. THOMAS, "The Criticality of Cubic Arrays of Fissile Material," Y-CDC-10, Oak Ridge National Laboratory (1971)

5. J. THOMAS, "Generic Array Criticality: An Analytic Representation of Reflected Arrays of Fissile Units," Y-CDC-13, Oak Ridge National Laboratory (1973)