The body of information on the criticality of subcritical components of fissile materials arranged in reflected critical arrays has grown sufficiently in the past ten years to warrant examination of density techniques and understanding of concepts employed in nuclear criticality safety. Monte Carlo calculations of experimental data have permitted valid extensions of these limited data. Correlations of the data have been effected and have resulted in an analytic expression relating the total number of units, \( N \), in an array, the edge dimension, \( d \), of cubic cells, and the mass, \( m \), of the fissile material centered in a cell. The mass necessary for criticality of such systems satisfies the equations

\[
\sigma(m) = \sigma_0 (m_0 - m) = \frac{m_0}{m} (1 - c/\sqrt{N^2}) \quad (1)
\]

In these relations, \( \sigma(m) \) is a limiting surface density \((g\cdot cm^{-2})\); \( m_0 \) is the unreflected critical mass in the geometry of the unit; \( n^2 \) is \( N \); \( c \) is a constant characterizing the geometry of center spaced units and equals 0.55; \( \sigma_0 \) is a constant dependent on the type of fissile material and is influenced by the unit shape, by the array shape, and by the array reflector material.

For cuboidal arrays of cubic cells, the mass \( m \) required for criticality satisfies the equation

\[
\left( \frac{m_0}{m} - 1 \right) = \frac{11.924}{\sqrt{N}} - 1 \quad (2)
\]

where the array shape is represented by the parameter

\[
R = \frac{N^{1/3}}{3} \left( \frac{1}{\rho} \right) \quad (3)
\]

and \( \eta_1 \) is the number of units along the three directions of the reflected cuboidal array. The value of \( R \) should not exceed 5.34. The definition of criticality by these equations has been determined as conservative below \( m = 0.1 m_0 \) and as very good for greater values of \( m \) representing criticality to within 1% in \( k_{eff} \).

The density analog representation of reflected array criticality is usually expressed in the form

\[
M = const \times \rho^s \quad (4)
\]

The power \( s \) is dependent on the size of the array approaching the value 2 as a limit and has been related to the mass of the unit. The above relations also may be expressed in this form. Cubing Eqs. (1) and (2) and rearranging the terms to yield

\[
N(1 - c/\sqrt{N})^s = \frac{\sigma_0 (m_0 - m)}{m} \frac{1}{\rho^s} \quad (cubic) \quad (3)
\]

Fig. 1. Density analog representation of reflected array criticality for 20 kg U(93.2) metal spherical units.
and

\[ N(1 - \frac{c}{\sqrt{N}})^6 = \frac{1}{(11.924)^m} \binom{m_0 - m}{6R - m - 1} \left( \frac{\rho_0 - \rho}{\rho R^{(m_0 - 1)}} \right)^3 \left( \frac{1}{\rho R^{(m_0)}} \right) \]  
(cuboidal). (4)

Application of Eqs. (3) and (4) to 20 kg U(93.2) metal spheres at a density of \( \rho_0 = 18.76 \, \text{g/cm}^3 \) for which \( c_2 \) has been evaluated as \( 1.762 \times 10^{-16} \, \text{cm}^{-1} \), results in the relations shown in Fig. 1. The shape of the cubic arrays is typical of that obtained from experimental and calculated data displaying the total mass \( (M = Nm) \) as varying inversely with density to a variable power. The data are equally well displayed as the total mass \( [M'] = (M(1 - \frac{c}{\sqrt{N}})^6) \) varying inversely as the square of the density. The departure from cubic arrays is illustrated for arrays having variable shape, \( R \), maintaining 2 cells in the vertical direction, beginning with an 8-unit array. For the same numbers of units in arrays, changing from cubic shape requires higher average densities. The arrays with constant noncubic shape were chosen to correspond to the shape of a reflected critical slab of U(93.2) metal.\(^4\) The latter example forcefully illustrates that as the average density approaches and achieves the fissile material density, the defined critical configuration is determined by the array shape and not the shape of the units in the array.

It may be said that the density methods can give complementary interpretations of criticality. The two constants, one for geometry and the other for fissile material as spherical units in arrays, are sufficient to represent criticality as

1. a continuous function of \( N \)
2. more properly depicting the total mass, or \( N \), as varying inversely as the square of the density
3. not being limited to cubic arrays
4. capable of approximating reflected and unreflected single-unit criticality for cuboidal shapes.


5. Special Criticality Considerations for Low-Enrichment Fuel Processing, Robert L Seale, Charles Verdon (U of Ariz)

The chemistry of UO\(_2\) in nitric acid is such that all the total uranium dissolved; the enrichment determination determines the criticality characteristics. For 5% enriched UO\(_2\) in a 100-cm-diam tank, the critical height is plotted in Fig. 1 as a function of solution inventory. At a typical dissolved uranium concentration of \( \sim 3 \, \text{M}, \) the \(^{238}\text{U} \) concentration is \( \sim 15 \, \text{g/liter}, \) so that a 1100-liter solution inventory, the system is

As fuel dissolves, heat is liberated in the following:

\[ \sim 75,000 \, \text{cal/MT due to UO}_2 \text{dissolving} \]
\[ \sim 17,500 \, \text{cal/MT due to fission heat release} \]

For 5% enriched fuel at 15 g \(^{238}\text{U} \)/liter concentration, the heat release is \( \sim 28 \, \text{cal/liter}. \) Thus, for dissolved at 50 to 85°C, cooling must be maintained to prevent boiling.

Mechanisms have been identified by which a reactor could be “forced” into an autocatalytic critical mode.

These mechanisms include:

1. Overbatching of the UO\(_2\) inventory by 40% to increase the \(^{238}\text{U} \) concentration to \( \sim 21 \, \text{g/liter}, \) the critical condition when 1000 liters are contained in the vessel.

2. Failure of cooling and solution boiling to reach the solution height while increasing \(^{238}\text{U} \) concentration to reach criticality.

Either mechanism will result in reaching the critical condition at a concentration-height combination such that the energy released due to fissions will boil off more solution and increase the uranium concentration even further.

Both situations are shown by the dotted lines on Fig. 1. Each results in a supercritical solution which expands in

![Graph](attachment://fig1.png)

**Fig. 1.** Critical height vs \(^{238}\text{U} \) solution concentration for a 100-cm-diam dissolver tank. Boiloff paths for constant inventory (a) and 40% fuel overload (b) incidents are shown.
An unresolved difficulty with Monte Carlo calculations which continues to cause concern is the liability to compute accurately the error estimates for the differential quantities (such as flux, fission densities, etc.) as a function of region and energy group. While there is no indication of error in computing the differential quantities themselves, the standard assumption when computing the statistical error that the "sample estimates" are independent is often not valid. To be correct, the statistical error calculation must take into account the correlation between "sample estimates." There is currently no general method to do this. While research on this problem continues, error estimates computed by standard techniques should be used with caution.

The Monte Carlo method provides the criticality safety specialist a rigorous, easy-to-use technique for evaluating many problems. A good understanding of the method and its limitations is essential if the user is to escape the pitfalls, which can lead to erroneous results. Undetected, these erroneous results could lead to erroneous safety recommendations.

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4. Remarks on Surface Density and Density Analog Representation of Array Criticality, J. T. Thomas (ORNL)

The body of information on the criticality of subcritical components of fissile materials arranged in reflected critical arrays has grown sufficiently in the past ten years to warrant examination of density techniques and understanding of concepts employed in nuclear criticality safety. Monte Carlo calculations of experimental data have permitted valid extensions of these limited data. Correlations of the data have been effected and have resulted in an analytic expression relating the total number of units, $N$, in an array, the edge dimension, $d$, of cubic cells, and the mass, $m$, of the fissile material centered in a cell. The mass necessary for criticality of such systems satisfies the equations

$$a(m) = \frac{m}{c_2} \left( \frac{m_0 - m}{m} \right) = \frac{m}{c_2} \left( 1 - \frac{m}{m_0} \right)^2.$$  \hspace{1cm} (1)

In these relations, $a(m)$ is a limiting surface density ($g - cm^{-2}$); $m_0$ is the unreflected critical mass in the geometry of the unit; $n$ is $N$; $c$ is a constant characterizing the geometry of center spaced units and equals 0.55; $c_2$ is a constant dependent on the type of fissile material and is influenced by the unit shape, by the array shape, and by the array reflector material.

For cuboidal arrays of cubic cells, the mass $m$ required for criticality satisfies the equation

$$m = \frac{m_0 - 1}{1 - \frac{m_0}{m}} = \frac{11.924}{nd} (5R - 0.672 - 1).$$  \hspace{1cm} (2)

where the array shape is represented by the parameter

$$R = \frac{n^2}{3},$$

and $n_i$ is the number of units along the three directions of the reflected cuboidal array. The value of $R$ should not exceed 5.34. The definition of criticality by these equations has been determined as conservative below $m = 0.1m_0$ and as very good for greater values of $m$ representing criticality to within 1% in $k_{\text{eff}}$.

The density analog representation of reflected array criticality is usually expressed in the form

$$M = \text{const} \times \rho^p.$$  \hspace{1cm} (3)

The power $p$ is dependent on the size of the array approaching the value 2 as a limit and has been related to the mass of the unit. The above relations also may be expressed in this form. Cubing Eqs. (1) and (2) and rearranging the terms to yield

$$N(1 - c/m)^2 = \frac{(m_0 - m)}{m} \frac{1}{m} (\text{cubic})$$  \hspace{1cm} (3)

Fig. 1. Density analog representation of reflected array criticality for 20 kg U(93.2) metal spherical units.
and

\[ N(1 - c/N)^6 = \frac{1}{(11.924)^2} \left( \frac{m_0 - m}{5N^{0.999} - 1} \right)^3 \frac{1}{\rho^3} \text{ (cuboidal)}. \]  

Application of Eqs. (3) and (4) to 20 kg U(93.2) metal spheres at a density of \( \rho = 18.76 \text{ g-cm}^{-3} \) for which \( c_\text{z} \) has been evaluated as \( 1.762 \times 10^{-2} \text{ cm}^{-1} \), results in the relations shown in Fig. 1. The shape of the cubic arrays is typical of that obtained from experimental and calculated data displaying the total mass (\( M = Nm \)) as varying inversely with density in a variable power. The data are equally well displayed as the total mass \( [M'] = M(1 - c/N)^6 \) varying inversely as the square of the density. The departure from cubic arrays is illustrated for arrays having variable shape, \( R \), maintaining 2 cells in the vertical direction, beginning with an 8-unit array. For the same numbers of units in arrays, changing from cubic shape requires higher average densities. The arrays with constant noncubic shape were chosen to correspond to the shape of a reflected critical slab of U(93.2) metal. The latter example forcefully illustrates that as the average density approaches and achieves the fissile material density, the defined critical configuration is determined by the array shape and not the shape of the units in the array.

It may be said that the density methods can give complementary interpretations of criticality. The two constants, one for geometry and the other for fissile material as spherical units in arrays, are sufficient to represent criticality as:

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4. capable of approximating reflected and unreflected single-unit criticality for cuboidal shapes.


5. Special Criticality Considerations for Low-Enrichment Fuel Processing, Robert L Scale, Charles Verdon (U of Ariz.)

The criticality limits for a large dissolver tank containing nitric acid as used in the reprocessing of LWR fuel have been examined over a wide range of dissolved uranium concentrations. Some potentially autacatalytic criticality situations have been identified in processing low-enrichment fuel.

The chemistry of UO\(_2\) in nitric acid total uranium dissolved, the enrichment, determines the criticality characteristics. For 5% enriched UO\(_2\) in a 100-cm-diam tank, critical height is plotted in Fig. 1 as a function of concentration. At a typical dissolved uranium concentration of 3 M, the \( ^{235}\text{U} \) concentration is \( \sim 0.06 \text{ M} \). For 5% enriched fuel at 15 g \( ^{235}\text{U} \)/liter concentration, heat release is \( \sim 28 \text{ cal/liter} \). Thus, for a system operated at 80 to 85°C, cooling must be used to prevent boiling.

Mechanisms have been identified by which it could be “forced” into an autacatalytic critical state. These mechanisms include:

1. Overbatching of the UO\(_2\) inventory by \( \rho \) increase the \( ^{235}\text{U} \) concentration to \( \sim 0.06 \text{ M} \) critical condition when 1000 liters are contained in the vessel.
2. Failure of cooling and solution boiling to solution height while increasing \( ^{235}\text{U} \) concentration to reach criticality.

Either mechanism will result in reaching the condition at a concentration-height combination where the energy released due to fissions will boil the solution and increase the uranium concentration further.

Both situations are shown by the dotted lines on each graph. Each results in a supercritical solution which expands.