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CALCULATED CRITICAL ARRAYS
OF FISSILE MATERIALS

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October, 1970

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CALCULATED CRITICAL ARRAYS OF FISSILE MATERIALS

To safely ship and store large numbers of fissile units, the reflected array critical size must be determined for each mass, spacing and internal moderation of the subcritical unit under consideration.

This paper presents the results of Monte Carlo calculations using both the GEM4⁽¹⁾ and KENO⁽²⁾ codes for critical arrays of 2, 3 and 4 kg spherical plutonium metal pieces and critical arrays of 10 liter (L-10) and 3 liter (L-3) Dow-type⁽³⁾ shipping containers of ^{233}U nitrate and plutonium nitrate solutions.

Good correlation has been established between GEM4 and KENO calculations and small experimental critical arrays of plutonium metal; the reactivities calculated by both codes for larger, less dense arrays also show good agreement^(4,5). The codes have also been correlated with plutonium and uranium critical solution experiments⁽⁶⁾ as well as ^{233}U solution experiments. All KENO calculations were made using 16-group Hansen-Roach cross sections⁽⁷⁾.

The number of identical subcritical units of plutonium metal in a critical cubic array is given in Figure 1 as a function of average plutonium density using parameters of constant mass with variable cell size and constant cell size with variable mass. The units are plutonium metal spheres of 19.7 g/cm^3 density and 5 wt% ^{240}Pu . The arrays were assumed to be closely reflected on all sides with 12-inch thick Portland Concrete⁽⁸⁾, which is the most efficient common reflector at Hanford. The calculated arrays shown by the curves have a k-effective of 0.98, one standard deviation subcritical, and therefore should

have a k-effective of less than 1.0 at the 83 percent confidence level.

The variation in critical array size with unit mass, type of reflector, and interspersed moderation is presented in Figure 2. The curve for 3 kg spheres with a k-effective equal to 0.98 is identical to that in the first figure. The curve for k-effective of 1.0 shows that the array size is increased by 35 percent or the plutonium density of the array is increased by 19 percent as contrasted to the curve where k-effective is equal to 0.98.

The reflection effects caused by two different materials, 6-inch water and 12-inch concrete (the composition is shown in Table 1), were also compared in Figure 2. That concrete can sometimes be a more efficient reflector than water has been known for some time and the comparison here with a 6-inch water reflector demonstrates the magnitude of this efficacy. For instance, with identical cell geometry, a concrete reflected array of only 350 units is critical compared to 1000 units critical when reflected with water. Thus, a Class III shipment, which meets the regulation that requires two combined shipments to be subcritical when reflected by water^(9,10), could become critical when one such shipment is reflected with concrete. Therefore, care must be exercised when plutonium metal shipments that meet regulations are to be stored within thick concrete enclosures. Replacing the water reflector with concrete would require a mass reduction of 30 percent, or a unit cell volume increase of 59 percent to maintain equivalent array reactivity with the same number of units.

Optimum interspersed water moderation was determined by holding the unit cell volume constant and increasing the

water density in the void. For an array of 1000 3-kg units (array density of 0.067 g/cm^3), the optimum water in the void was found to be 0.05 g/cm^3 or H/Pu of 20. Optimum moderation at an H/Pu of 20 between the spheres decreased the critical array density by 24 percent or the array size by 43 percent from a non-moderated array.

The second part of this paper deals with arrays of shipping and storage containers. An example of the L-3 and L-10 Dow-type shipping containers is illustrated in Figure 3. These containers are identical except for length, the L-3 is contained in one 55-gallon drum and the L-10 in two drums welded end to end. The solution is contained in a 4.72-inch O.D., 0.1-inch thick, polyethylene bottle within a flanged 5-inch schedule 40 or 80 pipe, centered inside the drum by framework and insulated with 0.134 g/cm^3 vermiculite. The vermiculite composition used in the calculations is shown in Table 1. Since the shipping containers can be stored on a triangular pitch which is not effectively handled by the KENO code, the cross-sectional area of the containers was reduced by 17 percent. This reduction in cross-sectional area allowed calculating the array in a square arrangement with a fissile density equivalent to the triangular pitched array. The Atlantic Richfield Hanford Company (ARHCO) storage container is illustrated in Figure 4. This 10-liter storage container is essentially the same as the L-10 except that vermiculite insulation is absent. The centering framework, inner receptacle size and closure mechanism are different and channel-iron legs are present. The legs permit easy movement of the containers by plant dolly and in addition they also prevent placing the containers in a triangular pitch array. Therefore, the k-infinity calculation for this container was made using full cross-sectional area and square pitch.

Calculations were made to determine the change in reactivity with fissile concentration. In Figure 5 the reactivity of an infinite array is presented as a function of ^{239}Pu and ^{233}U concentration in nitrate solutions. ARHCO specifications limit fissile concentration to a maximum of 450 g/l with ≥ 3 wt% ^{240}Pu ; therefore, most of the calculations were made using this concentration. The individual containers are safe for the worst foreseeable condition, i.e., fissile solution in the fully water-reflected 5-inch I.D. pipe. The plutonium containers are safe by geometry (cylinder diameter ≤ 85 percent of critical diameter) and the ^{233}U containers by reactivity (k -effective of the 18-inch long pipe ≤ 0.95). In case of fissile leakage or spillage into wet vermiculite, which is unlikely during shipping but a real possibility during loading or unloading, the L-3 containers would be subcritical because the mass is limited to 1125 grams of ^{233}U by customer specifications and DOT special permit. However, the L-10 loading-unloading specifications require a mechanical device to prevent spilling plutonium into the vermiculite.

Figure 6 is a plot of array reactivity of L-10 containers in tiers. All plutonium concentrations are 250 g/l with 3 wt% ^{240}Pu except for the infinite one-tier arrays which are 450 g/l with 0 wt% ^{240}Pu . The infinite one-tier arrays are closely reflected on top and bottom by 12-inch concrete while all other arrays have a 12-inch concrete reflector on all sides. Calculations indicate that infinite one-tier arrays of the storage and shipping containers are subcritical. The negative reactivity of the vermiculite with 1.0 wt% hydrogen is shown in one calculation for the L-10 container. The interspersed moderation has a negative

reactivity effect in this case because steel surrounds the units. This result is consistent with a previous criticality safety evaluation of similar containers which indicated the maximum array reactivity occurred when the vermiculite was at normal dryness⁽¹¹⁾.

The dashed line represents one standard deviation above the reactivity curve derived from the GEM calculations. This curve indicates that 168 containers will be subcritical with 83 percent confidence while the KENO point shows that 162 containers have a greater than 95 percent statistical confidence of being subcritical—a good agreement.

In Figure 7 the number of L-3 containers full of 450 g/l ^{239}Pu and reflected with 12-inch concrete is compared to the array reactivity. The line is a least squares fit of the points. An array of 2250 containers has a k-effective of 0.98, two standard deviations less than 1.0 and the array will thus be subcritical at the 95 percent confidence level. One calculation was made to determine the effect of reflector material upon the reactivity of the array. With a 6-inch water reflector the reactivity decreased approximately 2 percent or the array size increased 15 percent.

The reactivity of L-3 container arrays of 450 g/l ^{233}U as uranyl nitrate solution is shown in Figure 8; the least squares fit of the points indicate that 600 containers have a k-effective of 0.98 with a 95 percent confidence of being subcritical. This array is more sensitive to the reflector material than the plutonium array with a 39 percent increase in array size when the concrete reflector is replaced with a 6-inch water reflector.

The Oak Ridge Laboratory fabricated a shipping container similar to the Dow L-3 except that foam glass containing

1.5 wt% boron replaced the vermiculate insulation. The negative reactivity of foam glass insulation is illustrated in this figure; k-effective decreased 11 percent or the array size increased approximately 200 percent.

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TABLE I

COMPOSITION OF MATERIALS USED IN CALCULATIONS

<u>Element</u>	<u>Portland Common (8)</u> <u>Concrete, 2.3 g/cm³</u>		<u>Vermiculite (11)</u> <u>Pack Density 0.134 g/cm³</u>	
	<u>Wt%</u>	<u>Atoms/barn-cm</u>	<u>Wt%</u>	<u>Atoms/barn-cm</u>
H	1.0	.01386	1.0	.000807
C	0.1	.00012		
O	52.9	.04582	48.0	.002420
Na	1.6	.00100		
Mg	0.2	.00011	12.7	.000421
Al	3.4	.00174	7.8	.000233
Si	33.7	.01063	18.2	.000524
K	1.3	.00046	5.8	.000120
Ca	4.4	.00152		
Fe	1.4	.00035	6.3	.000091

FIGURE 1

FIGURE 2

TYPE L-10 CLASS II
SHIPPING CONTAINER
DOT SP 5061

FIGURE 3

PLUTONIUM NITRATE STORAGE CONTAINER
(10 LITER)

FIGURE 4

FIGURE 5

FIGURE 6

FIGURE 7

FIGURE 8

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