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NUCLEAR SAFETY EVALUATION OF UNITS CONTAINING ^{238}Pu AND ^{239}Pu AS OXIDES

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DISCLAIMER

NUCLEAR SAFETY EVALUATION OF UNITS CONTAINING ^{238}Pu AND ^{239}Pu AS OXIDES

This study evaluates the nuclear safety of ^{238}Pu as PuO_2 under a variety of conditions for shipping and storage and is applicable to mixtures of ^{238}Pu-^{239}Pu oxides. Evaluations were made for a three-container shipping package (Plutonium Finishing Shipping Cask) for compliance with AECM Chapter 0529 Shipping Regulations. Some calculations were also made for the two inner containers as a storage unit to extend the usefulness to cover B-Line storage[1]. In all cases the PuO_2 is contained in cans 1.025" ID x 13" inside height and with few exceptions has a bulk density of 2.6 g/cm^3.

This safety analysis was developed with ^{239}Pu and ^{238}Pu cross sections using KENO, a multigroup Monte Carlo criticality program. Conservative allowances were made so that the results are valid for all mixtures of ^{238}Pu-^{239}Pu.

SUMMARY OF RESULTS

An infinite array of undamaged three-container packages containing up to 357 g plutonium as PuO_2 with a maximum bulk density of 2.6 g/cm^3 PuO_2 is safe. KENO calculations yield Keff = 0.13 for an infinite array with water between the undamaged packages and Keff = 0.15 with air between the packages. Keff values were calculated for arrays of damaged packages for several conditions. The results support a previous letter[2] which reported that: packages may be shipped with a transport index of 0.6 as Fissile Class II; and 192 packages may be shipped as Fissile Class III. The present calculations studied the effect of array size and would support a lower Fissile Class II transport index, 0.3, and a 400-package limit as Fissile Class III. For storage conditions a planar array one unit tall of two or three container packages is shown to be safe regardless of water moderation provided the spacing is 2.375" center-to-center or greater.

NUCLEAR CRITICALITY SAFETY EVALUATION

This nuclear safety evaluation of the PuO_2 shipping container is based on calculations using KENO. KENO[3] is a multigroup Monte Carlo criticality program written for the IBM 360-65 by G. E. Whitesides and N. F. Cross of the Nuclear Division of Union Carbide Corporation. After some minor modifications at SRL, KENO was tested against a number of experimental measurements[4-6]. Table 1 shows data taken from [6] which gives results from KENO II, the present SRL version for some experimental critical situations. (Keff = 1.000 for a critical situation.)

Table 1
KENO II Calculations for Some Experimental Critical Assemblies

Unit	Lattice	KENO II
Oralloy cylinder	2 x 2 x 2 (bare)	1.003 ±0.004
	3 x 3 x 3 (bare)	0.992 ±0.005
	3 x 3 x 1 (bare)	1.002 ±0.005
5 liters of 415 grams U/liter solution	2 x 2 x 2 (bare)	0.974 ±0.006
	3 x 3 x 3 (bare)	0.955 ±0.006
	4 x 4 x 4 (bare)	0.936 ±0.006
	5 x 5 x 5 (bare)	0.955 ±0.006
	2 x 2 x 2 (reflected)	0.989 ±0.007
	3 x 3 x 3 (reflected)	0.981 ±0.006

The calculated results for bare oralloy cylinders are in good agreement with the experimental Keff measurements. For units of uranium solution the KENO results are slightly less than 1.00, on the order of 0.95 for bare arrays. However, calculations for reflected solution units yield Keff values above 0.98 for experimental critical assemblies.

In order to give maximum effect to the AECM 0529 Fissile Class II accident the Pu Finishing Cask, in the damaged condition, was assumed to be a single cylinder with a diameter of Schedule 40 304 SS pipe used to make the cask liner. For simplicity in KENO, the large bulbous end (4.5" OD) and the 9" OD flange were omitted and the entire body was assumed to be 2.375" OD. These simplifying assumptions allow more interaction than the actual case. The assumptions, all of which are conservative, used to describe the shipping package as a unit cell, are given in the order used by KENO, from the inside out.

1. The cavity of the B-Line Shipping Container is assumed to be uniformly cylindrical with 175.85 cc volume whereas the actual container[7] has tapered bottleneck ends and a measured volume of 156 cc.
2. The end caps of the B-Line Shipping Container were the minimum thickness in the cap.
3. Length dimensions of the EP-61 can (the secondary can)[8], and the Pu Finishing Shipping Cask (outside container)[9] were modified so that container ends are in contact from the B-Line container out. This results in optimum interaction end-to-end in an array and is more conservative than the actual case.
4. The Pu Finishing Shipping Cask was assumed to be a simple cylinder with the diameter of the main body. This simplification removes the approximately 4" OD section at the top and the accompanying 9.125" diameter flanges which would impose some separation in any random array. Neglecting the approximately 4" OD bulb at the top reduces the effective center-to-center spacing by 19.9% in the closest packed arrangement and is more conservative than the actual case. Consideration of the separation imposed by the flanges would further increase the center-to-center separation with accompanying lower Keff values.

Table 2 lists the actual and calculational dimensions used in KENO. In cases where they differ the calculational dimension allows more interaction or less neutron absorbing material than the actual unit.

Table 2
Compendium of Actual Unit and Calculational Unit

Parameter	Actual	Calculational
PuO_2 density, g/cm^3	1.0-1.1	1.0-2.6
Pu density, g/cm^3	0.88192-0.97011	0.88192-2.29299
B-Line Shipping (Primary) Container (316 SS)		
ID, inch	1.025	1.025
OD, inch	1.690	1.690
I. Ht., inch	13.00	13.00
O. Ht., inch	14.36	13.86
EP-61 (Secondary) Container (304 SS)		
ID, inch	1.760	1.760
OD, inch	2.000	2.000
I. Ht., inch	16.125	13.860
O. Ht., inch	16.438	14.360
Pu Finishing Cask (Tertiary Container) (304 SS) (damaged)		
ID, inch	2.062-2.093	2.067
OD, inch	2.352-2.354	2.375
I. Ht., inch	19.013	14.360
O. Ht., inch	23.250	14.960
(undamaged)		
ID, inch	2.352	2.352
OD, inch	16.750	12.250
I. Ht., inch	19.9	14.360
O. Ht., inch	33.125	14.960

Table 3 lists the materials and their atomic densities used in these KENO calculations.

Table 3
KENO Input for Materials in Unit Cells

Material	Nuclide	Atomic Density (atoms/barn-cm)
PuO_2 (dry)	^{239}Pu, ^{238}Pu	5.77840×10^{-3}
	O	1.14946×10^{-2}
PuO_2 (wet)	^{239}Pu, ^{238}Pu	5.77840×10^{-3}
	O	3.73528×10^{-2}
	H	5.15875×10^{-2}
316 SS	Fe	5.979×10^{-2}
	Cr	1.468×10^{-2}
	Ni	1.138×10^{-2}
304 SS	Fe	6.064×10^{-2}
	Cr	1.652×10^{-2}
	Ni	8.936×10^{-3}
Water at 25°C	O	3.3363×10^{-2}
	H	6.6726×10^{-2}

The calculations in this study were made, with few exceptions, for PuO_2 at 2.6 g/cm^3 either wet or dry. For the wet oxide studies the volume occupied by the PuO_2 was determined from its mass and theoretical density, 11.46 g/cm^3. The remaining volume was filled with water at 25°C. The PuO_2 was uniformly distributed throughout the volume of the innermost container.

The calculations, intended for ^{238}Pu, were made using both ^{238}Pu and ^{239}Pu cross sections in unmoderated systems where fast neutron behavior dominates. In slow or thermal systems only ^{239}Pu nuclear data were used since ^{238}Pu is not fissile in thermal systems. The results should be valid for all mixtures of ^{238}Pu-^{239}Pu.

For the calculational model used in these KENO arrays radii are in the X-Y plane and the cylindrical axis is in the Z direction. Center-to-center spacings are spacings in the X-Y plane for cylindrical axes parallel to the Z axis. Array descriptions give units in the order X, Y, Z; i.e., a 10 x 10 x 2 array is 10 units in the X direction by 10 units in the Y direction and stacked two units high with all cylinder axes parallel to the Z axis. These are conventional descriptions and are emphasized here to caution those not familiar with them. Proper care must be taken when applying results from this document to some other array because the geometric orientation can be important.

Arrays of Units

Each calculational unit contains 403 g ^{239}Pu as PuO_2 at 2.6 g/cm^3. At this density the actual container capacity would be 357.5 g ^{239}Pu as PuO_2. Arrays, where water was considered between units, were assumed to be surrounded by a 30 cm water reflector, based on differential albedo data in the KENO library[6]. In unmoderated arrays air was treated as void between units.

Early in the study calculations were made for full B-Line Shipping Containers of varying oxide concentrations which demonstrated that reactivity increased with increasing PuO_2 concentration (see Table 4).

Table 4
Keff as a Function of PuO_2 Concentration

Array size[a]	10 x 10 x 5	20 x 20 x 5
Moderator	Air	Air
^{239}Pu oxide condition	Wet	Wet
PuO_2 Density, g/cm^3	Keff	Keff
1.0	0.79	1.05
1.4	0.84	1.11
1.8	0.85	1.17
2.2	0.87	1.19
2.6	0.93	1.24

[a]Each unit of the array is the 3-container assembly with Pu as PuO_2 plus water in the innermost container. The units are 2.375" center-to-center.

A study of moderator density for arrays where the unit cell may contain oxide and water in the innermost can indicated that maximum reactivity occurs when there is no moderator between units of the array (see Figure 1). Separate safe limits must be established for cases where this situation is possible, i.e., storage and handling of oxide scrap.

With PuO_2 at 2.6 g/cm^3 in the innermost container and the remaining volume filled with water at 25°C, the resulting H/Pu ratio is approximately 9. Unpublished work for a similar material, $^{233}UO_2$, in cylindrical containers on Keff as a function of oxide height showed that full containers were more reactive than partially filled containers at constant mass.

These scouting studies indicated that:

1. full cans were the most reactive configuration
2. calculated Keff increased with increasing oxide density
3. water inleakage increased Keff for ^{239}Pu and Keff was still increasing when cans were full of PuO_2 and water (H/Pu $\simeq$ 9).

Accordingly, the basic unit in all configurations is filled with PuO_2 at 2.6 g/cm^3 (an arbitrary maximum chosen to cover shipping and storage problems arising from handling PuO_2 at SRP) and when internal moderation is considered, water was allowed to fill the available space. ^{239}Pu has a higher fission cross section for neutrons below 0.5 Mev than ^{238}Pu so that atom-for-atom substitution of ^{239}Pu for ^{238}Pu in thermal neutron systems is conservative.

Calculations were made to answer pertinent sections of AECM Chapter 0529 shipping regulations and to establish storage conditions for SRP. All pertinent sections of AECM Chapter 0529 are explicitly discussed in the Appendix.

Keff values were calculated for arrays of units in contact edge-to-edge with water in all containers including the B-Line Shipping Containers with the PuO_2 at full density but uniformly dispersed in water. Calculations for this case were not necessary since the B-Line Shipping Container meets the requirements of AEC 0529 Annex 2.

Studies on The 3-Container Package

Figure 2 shows the effect of spacing on large planar arrays. These results demonstrate that Keff decreases on separation of units.

The work in Table 4, Figures 1 and 2 shows that Keff increases with:

- increasing mass Pu per unit
- decreasing water between units
- decreasing space between units.

A number of calculations were made to study the effect of array size and arrangement. The results and the conditions considered are given in Table 5. Data in the first five rows of results show that wet ^{239}Pu oxide arrays have higher Keff values than dry ^{239}Pu or ^{238}Pu oxide arrays by a substantial margin. In small arrays where the units contain wet $^{239}PuO_2$, arrays having water between the unit cells have slightly higher Keff values than the corresponding arrays with air between the units. However as the size of the array increases, the air moderated arrays increase in Keff faster than those that are water moderated. Accordingly, the effects of array size and arrangement were investigated for air moderated units containing wet $^{239}PuO_2$.

Table 5
Keff As A Function of Array Size
(unit cells are 2.375" C-C)

Case	1	2	3	4
Pu Isotope	239	239	239	238
Oxide Condition	Wet	Wet	Dry	Dry
Moderator	Air	Water	Air	Air
Array Size	Keff	Keff	Keff	Keff
6 x 6 x 6	0.64	0.69	0.37	0.21
8 x 8 x 3	0.75	0.78	0.43	0.26
8 x 8 x 5	0.79	0.78	0.43	0.26
9 x 9 x 5	0.88	0.81	0.48	0.29
10 x 10 x 5	0.88	0.84	0.52	0.31
12 x 12 x 2	-	0.83	-	-
12 x 12 x 3	-	0.84	-	-
12 x 12 x 4	0.98	0.87	-	-
13 x 13 x 1	0.76	-	-	-
13 x 13 x 2	0.95	-	-	-
13 x 13 x 3	1.01	-	-	-
15 x 15 x 1	0.82	-	-	-
15 x 15 x 2	0.99	-	-	-
20 x 20 x 5	1.24	-	-	-
50 x 50 x 1	0.92	0.87	-	-
50 x 50 x 2	1.25	-	0.77	0.51
50 x 50 x 3	1.32	-	0.96	0.60
50 x 50 x 5	1.42	-	1.12	0.65
50 x 50 x 10	1.45	-	1.20	0.72

Five hundred units, the largest number permitted under AECM 0529 regulations for Fissile Class II with the minimum transport index of 0.1, are not always safe. The Keff calculated for a 13 x 13 x 3 array is 1.01 while that for a 15 x 15 x 2 array is 0.99. For arrays one unit high, as many as 2500 units are permissible, but stacked two units high a 15 x 15 x 2 array of 450 units has Keff $<$ 0.99. A 13 x 13 x 2 array of 338 units has Keff = 0.95, and a 9 x 9 x 3 array must have Keff $<$ 0.88, that for a 9 x 9 x 5 array. Therefore, 400 units in any arrangement are subcritical so one-half that number or 200 units may be shipped as Fissile Class II with a Transport Index of 0.3. As mentioned earlier, a 50 x 50 x 1 array is safe; the calculated Keff is 0.82. Such an array could be useful in storage situations.

Cases 3 and 4 of Table 5 contain Keff results for fast neutron cases involving ^{239}Pu and ^{238}Pu, respectively. In all cases the results in 3 and 4 are less than the corresponding wet ^{239}Pu wet oxide studied in Case 1. Thus air moderated wet $^{239}PuO_2$ is the limiting condition for this study.

The calculations thus far have been for reflected arrays where the reflector is 30 cm thickness of water accomplished via an albedo option in the KENO code. The results in Table 6 are for identical arrays as function of array size for water reflector and no reflector cases. For the cases presented the water reflector increases Keff by as much as 0.1 Keff units so that partially reflected and unreflected applications offer additional safety margin.

Table 6
Effect of Reflector on Keff of Arrays
(Units are identical with those for Case 1, Table 5)

Array Size	Water Reflector Keff	No Reflector Keff
10 x 10 x 5	0.88	0.79
12 x 12 x 4	0.98	0.91
13 x 13 x 1	0.76	-
13 x 13 x 2	0.95	0.88
13 x 13 x 3	1.01	0.92
15 x 15 x 1	0.82	0.68
15 x 15 x 2	0.99	0.89
15 x 15 x 3	-	1.03

Storage Considerations: Two-Container Package

Considerations for safe storage of the two inner containers as a unit cell prompted some additional calculations. (The two-container unit is not being considered as a shipping unit and therefore is not treated in the appendix where shipping regulations are considered.) Keff calculations were made for essentially infinite planar arrays (50 x 50 x 1) for wet and dry oxide with no moderator. The results, shown in Figure 3, have higher Keff values for the two-container unit than the corresponding three-container unit under similar conditions. A significant difference is that a large planar array of the two-container units, 2.0" diameter, can become critical at close spacing. Therefore spacing the units or limiting the number of units is necessary even in a planar arrangement of two-container units. However, a spacing 2.375" center-to-center or greater will provide safe storage for a planar array of 2500 (50 x 50 x 1) two-container units. (See Figure 3.) In ^{238}Pu operations none of the conditions for Case 1 in Table 9 should be realized, certainly not all conditions simultaneously. The product should have a high percentage ^{238}Pu, the cans were designed and tested against leakage, and the intent is to store these units in a water bath at spacings greater than 2.375" center-to-center. The likelihood of achieving Case 1 conditions in ^{238}Pu operations for a large number of array units is quite small. However it should not be neglected for situations such as low array material and scrap operations because of the many possibilities.

Some calculations were made to investigate the feasibility of a simple number limit for two-container packages that is a limit on the number of units that could be together regardless of spacing, moderator, oxide condition or isotopic distribution. The results in Table 7 for the most reactive condition (wet ^{239}Pu oxides, void between cans in contact) show that the Keff depends strongly on the arrangement rather than the number of units in the array. Therefore process control on material outside of specific storage conditions should be controlled by geometry (spacing) or mass of PuO_2 rather than number of containers. (However the mass limit may be used to limit the number of containers.)

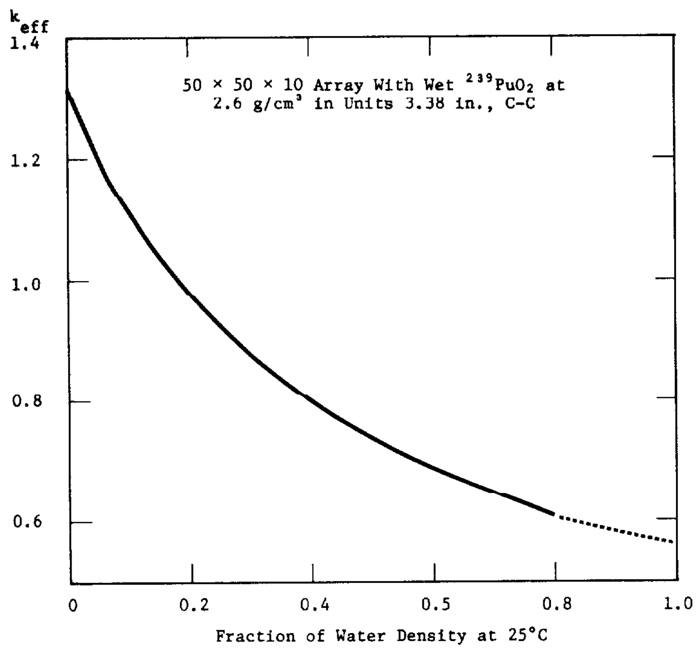

FIGURE 1. EFFECT OF MODERATOR DENSITY ON k_{eff}

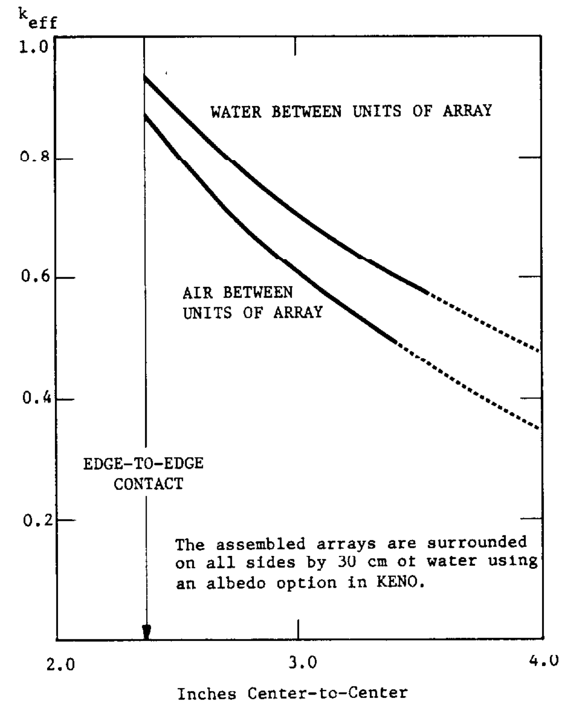

FIGURE 2. k_{eff} AS FUNCTION OF CENTER-TO-CENTER SPACING

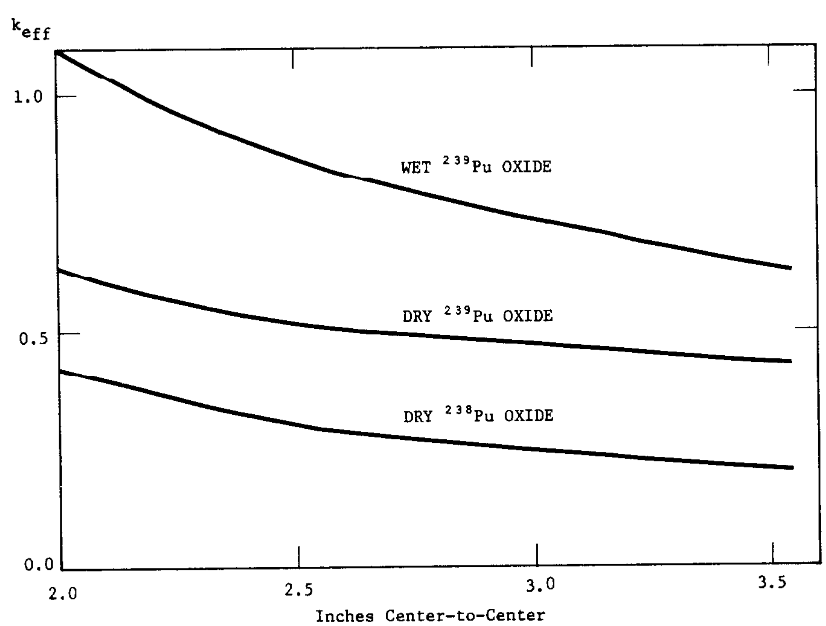

FIGURE 3. EFFECT OF SPACING ON k_{eff} FOR A 2-CAN B-LINE UNIT

Table 7

Keff as Function of Array Size

The unit cell is the two-can B-Line package, 2.00" center-to-center (contact) containing wet ^{239}Pu oxide. There is air (void) between units of the array.

Array Size	No. of Units	Keff
6 x 6 x 6	216	0.731
8 x 8 x 3	192	0.876
8 x 8 x 5	320	0.877
9 x 9 x 5	405	0.960
10 x 10 x 5	500	1.043
12 x 12 x 2	288	1.031
12 x 12 x 3	432	1.094
12 x 12 x 4	576	1.127

APPENDIX

PERTINENT SECTIONS OF AECM CHAPTER 0529

I, E. Standards for normal conditions of transport for a single package

2.(a) An individual package is subcritical on mass limits as well as by diameter of the cylinder.

(b) The package contents were assumed to completely fill the containment vessel; therefore no alteration of geometric form of contents was considered.

(c) 2) See II, C.3.

(d) The effective spacing for containers was reduced from 10" center-to-center, that of undamaged Pu Finishing Shipping Casks, to 2.375" center-to-center, the spacing for the smallest diameter (of the Pu Finishing Shipping Cask) on square pitch. This inherently assumes a reduction over the actual spacing. The Pu Finishing Shipping Cask central cavity has a bulbous end that is approximately 4" OD and a flanged closure that is 9.125" OD. This bulbous end forces spacing of container bodies at the closest point containing PuO_2 to 3.746" center-to-center. Also, to simplify calculations the cask length was reduced to the minimum length necessary to house the B-Line Shipping Container and a similarly modified EP-61 can. The simplified diameter represents a reduction in spacing of 19.9% over the actual minimum and the reduced length represents an even larger reduction.

II, C. Criticality standards for fissile materials

1. The contents of a single package are limited to 350 g ^{238}Pu (rounded off from 357.5) as PuO_2 where the PuO_2 density does not exceed 2.6 g/cc.
2. This section concerns leakage of contents that are liquid during normal transport and does not apply to PuO_2 shipments.
3. The manager (SROO) may approve exception to liquid tightness requirements when special design precludes leakage and appropriate measures are taken before each shipment to verify leak tightness of the containment vessel.

II, C. 3. (Cont'd)

While the mechanical evaluation test demonstrated adequate bases for the manager to approve such an exception it is not necessary for nuclear safety. As shown in the most active case, a dry but reflected array of damaged three-can units in contact with water in all containers, shows Keff values less than 1.0 for 400 or fewer packages. Unmoderated arrays of the same size containing dry PuO_2 have Keff values reduced by about one half (see Table 5).

II, F. Standards for hypothetical accident conditions for a single package

2. A single package is subcritical on the basis of mass alone. The calculations for damaged containers assume:
   1) most reactive credible configuration consistent with chemical and physical properties of contents
   2) most reactive water moderation
   3) reflection by water.

II, G. Evaluation of an array of packages of fissile material

See Tables 5 and 6, "Nuclear Criticality Safety Evaluation".

II, I. Specific standards for a Fissile Class II package

1.(a) An infinite array of undamaged packages would be subcritical in any arrangement even if fully moderated and reflected by water. KENO calculations yield Keff = 0.13 for an infinite array with water between the units and Keff = 0.15 with air between the units.

(b) Calculations demonstrated that 400 damaged containers will be subcritical so that 200 could be safely chosen for number of Fissile Class II packages in a single shipment. (The 8 x 8 x 3 array considered in the prior study[2] resulted in a limit of 96 Fissile Class II packages in a single shipment because no effort was made to investigate the effect of array size or to seek the maximum number to be permitted in a single shipment.)

2. The transport index for each Fissile Class II package rounded up to the next highest tenth is $50 \div 200 = 0.3$. (A transport index of 0.6 was determined for each Fissile Class II package in the study on the 8 x 8 x 3 array[2] which did not seek a minimum transport index.)

II, J. Specific Standards for A Fissile Class III Shipment

1. The undamaged shipment would be subcritical with an identical shipment in contact with it and with the two shipments closely reflected by water . . . . See II, I. 1(a).
2. The shipment would be subcritical if each package were subjected to the hypothetical accident conditions . . . with packages in most reactive arrangement and with the most reactive degree of interspersed hydrogenous moderation . . . See II, I. 1(b); 400 damaged packages are safe.

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