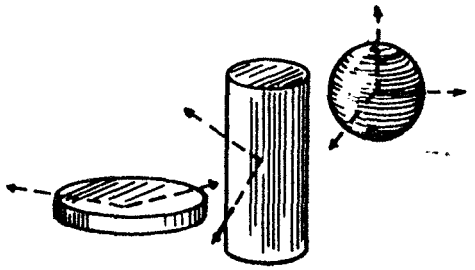

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CRITICALITY SAFETY IN THE STORAGE OF FISSILE MATERIAL

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APPLICATIONS OF THE LIMITING SURFACE DENSITY METHOD TO TRANSPORT AND STORAGE OF SPECIAL NUCLEAR MATERIALS

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ABSTRACT

The Limiting Surface Density Method has been used in criticality safety assessments for the storage and on Site movement of special nuclear materials. An extension of the existing materials database to mixed oxide, ie PuO_2, UO_2, Pu/(U+Pu) = 40%, has been made in order to increase the applicability of the method to problems of interest. Appropriate constants for mixed oxide at Hydrogen to Actinide ratios of 0.4:1, 3:1 and 10:1 were calculated using KENO4 to identify the critical state for selected air spaced, fully water reflected, cubic arrays of spherical units. Typical applications for non-cubic arrays are also described and the results are compared with direct MONK6.3 and KENO4 calculations. The utility of the existing database and its extension is demonstrated.

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APPLICATIONS OF THE LIMITING SURFACE DENSITY METHOD TO TRANSPORT AND STORAGE OF SPECIAL NUCLEAR MATERIALS

1 INTRODUCTION

The Reprocessing Complex at Sellafield, Cumbria, United Kingdom, is operated by British Nuclear Fuels plc (BNFL). Current activities include the reprocessing of natural uranium metal fuel which has been irradiated in graphite moderated, CO_2 cooled, reactors situated at Latina, Italy, Tokai-Mura, Japan, and at sites within the United Kingdom. Oxide fuels from indigenous gas cooled thermal reactors and overseas light water cooled reactors are received for storage pending operation of the THORP reprocessing plant in the early 1990s. Since the fuel for the Prototype Fast Reactor (PFR) at Dounreay, Scotland, is fabricated at Sellafield, the Site also forms part of the fast reactor fuel cycle.

Plutonium oxide and mixed oxides are manufactured, transported within the Site and stored during the activities mentioned above. This paper describes the application of the Limiting Surface Density Method to the associated questions of nuclear criticality safety and the extension of the available data in order to enable the method to be used in the assessment of mixed oxide arrays.

2 THE LIMITING SURFACE DENSITY METHOD - A BRIEF DESCRIPTION

The method[1] is designed to be used in the assessment of units of fissile material arranged in reflected cubic or cuboidal arrays. It is based on the experimental result that the fraction of neutrons which do not escape from the array boundary is a constant for arrays comprising the same units and having the same multiplication factor.

Consider a vertical stack of spherical fissile units where the space between the units is filled with air and the stack forms part of a cubic or cuboidal array reflected on all six sides. The surface density, $\sigma(a_n)$, may be defined as the total mass in the stack divided by its base area; a_n is defined as the half cell spacing i.e. for cubic unit cells it is simply half the side, and for cuboidal unit cells it is half the side of a cube of equivalent volume. Conventionally, the characteristic surface density for the array is calculated for the side with the least number of units. The limiting surface density, $\sigma(m)$, is the value of $\sigma(a_n)$ as the number of units in the array becomes indefinitely large but remains finite. The relationship between $\sigma(m)$, $\sigma(a_n)$, m, a_n, N, the total number of units in the array, and n_z, the least number of units along any side, is[2]

$$\frac{\sigma(m)}{m} = \frac{n_z}{(2a_n)^2}\,(1 - c/\sqrt{N})^2 \qquad (1)$$

c is an empirically determined constant, equal to 0.55 ± 0.18, for a range of plutonium and uranium compounds[3]. For these materials

H:Actinide is less than or equal to 20 and relatively few fissions are thermal. As the fraction of thermal neutrons carrying the chain increases so c tends towards zero.

It is possible, by calculation or experiment, to identify the unit mass required for array criticality in arrays of identical units. Thomas's precursory Monte Carlo calculations[4] were based on cubic arrays of identical units with full water reflection around the array as a whole. Unit sizes giving array criticality were calculated for various materials in cubic arrays containing 64 to 1000 units at a_n values from 25.4 to 60.96 cm. For each array the value of $\sigma(m)$ was calculated using equation 1 and the result plotted in the $\sigma(m)$, m plane. Each point in the $\sigma(m)$, m plane represents a family of arrays since, for cubic arrays, $\sigma(m)$ also depends on the values of a_n and N; for cuboidal arrays there is a further dependency on n_z. The locus of such points for critical arrays having not less than 64 units was established empirically as a straight line Therefore, only two numbers are needed to represent the family of cubic arrays of this material, namely, c_2, the absolute value of the slope and m_o, the intercept on the mass axis. Thus the equation of the line may be written,

$$\sigma(m) = c_2 (m_o - m) \qquad (2)$$

or

$$\frac{\sigma(m)}{m} = c_2 (\frac{m_o}{m} - 1) \qquad (3)$$

Since N, a_n and n_z are attributes of the array, it follows from equation 1 that $\sigma(m)/m$ is a function of the array geometry only Thus the line $\sigma(m)/m$ describes the geometry of a family of arrays in the $\sigma(m)$, m plane. The point of intersection with the line described by equation 2 occurs at the unit mass required for criticality in these cases

Two conclusions may be drawn. Firstly, for a given N and n_z, a_n tends to infinity as m, the unit mass required for array criticality, tends to m_o. The physical significance of m_o is that it is the unreflected critical mass for the material concerned. Secondly, neutron coupling between units must be weak in critical arrays for which m is close to m_o. Where m is much less than m_o the neutron coupling is strong and it is in this region that interstitial moderation by low density materials such as water sprays is important.

Thomas has calculated values of c_2 and m_o for 46 materials[3]. More recently, the storage of low enriched uranium oxide has been analysed[5]. The calculation of c_2 and m_o values for mixed oxides described in section 3 extends this database.

For each material i, the unit mass required for array criticality is given by the intersection of the appropriate $\sigma(m)/m$ line with the characteristic line $\sigma(m_1) = c_{2i} (m_{oi} - m_i)$. This construction is illustrated in Figure 1. The masses at the intersections are equivalent in the sense that the substitution of one unit of a given material and mass

for another of a different mass and material maintains the criticality of the array. The equivalence relation in this case is,

$$\frac{m_o'}{m'} = 1 + \frac{c_2}{c_2'}\,\frac{(m_o - 1)}{m} \qquad (4)$$

where the primed quantities indicate the second material.

The unreflected critical mass, m_o, is inversely proportional to the square of the material density[6]. For a given material composition the value of m_o can therefore be scaled to a different density. $\sigma(0)$ is unaffected.

Similarly, m_o can be adjusted to include the effects of changes in unit shape; the effect on $\sigma(0)$ is insignificant. Calculations done for cylindrical units[7] indicate that the ratio of height to diameter must lie in the range 0.3 to 3 for the method to apply.

A change in the shape of the array will not, by definition, affect the value of m_o. Such a change does, however, affect c_2 and hence $\sigma(0)$. The effect of changing the array shape, everything else remaining constant, is to increase neutron leakage. Consequently, the unit mass must be increased in order to maintain array criticality. The effect in the $\sigma(m)$, m plane is shown in Figure 2. Both lines are affected, the one described by equation 2 because of the change in c_2 and the $\sigma(m)/m$ line because of the change in n_z. Thomas studied the effect of array shape changes for arrays of 10.42 kg spherical U(93.2) metal units. With full water reflection, 512 such units are critical in cubic array at an a_n of 16.454 cm[8]. For these units the mass required for array criticality is found to vary with the shape of the array according to the relationship,

$$m' = m\,R^{0.672} \qquad (5)$$

where, $m = 10.42$ kg

m' is the mass required for array criticality in the cuboidal array with shape factor R.

and R is the array shape factor representing a surface area ratio with respect to the cubic array with the same number of unit cells.

The shape factor R must be less than or equal to 5.34 for the method to apply.

Analytically, $R = (N)^{(1/3)}\,(1/n_x + 1/n_y + 1/n_z)$ (6)

Using equation 5 and equating for similar $\sigma(m)/m$ in equation 2 the following relationship is obtained.

$$c_2(R,(n_z/n)) = \frac{n_z\ 4\ c_2}{n\ (5R^{-0.672} - 1)} \qquad (7)$$

If the unit mass for criticality in a non-cubic array is required, the first step may be to calculate the corresponding unit mass of U(93.2) metal. As the same value of $\sigma(m)/m$ applies, the equivalent mass for the material of interest can be calculated using equation 2. Since the ratio of c_2s is independent of the shape factor R, the c_2 values for cubic arrays may be used in equation 4.

The effect of replacing full water reflection with concrete reflection around the array will depend on the concrete thickness and may either increase or decrease the unit mass required for array criticality. The change in k-effective is dependent on the shape of the array and the fissile material involved; the higher the neutron leakage the more susceptible the array will be to changes in reflector. Thomas[9] has calculated the effect of replacing a full water reflector with varying thicknesses of concrete for arrays of 9 kg U(93.2) spherical metal units and presented the results in the $\sigma(m)$, m plane.

3 THE CALCULATION OF c_2 AND m_0 VALUES FOR MIXED OXIDES

The manufacture of fuel for PFR involves the granulation, pelleting and sintering of a mixture of plutonium and natural uranium oxides. The mixed material thus exists at various densities and degrees of moderation. For nuclear criticality assessment purposes it is appropriate to assume that plutonium accounts for 40 wt% of the actinide content, that all Pu is Pu239 and that the uranium is unenriched. Three sets of c_2 and m_0 values have been calculated, one for each of the H/(Pu + U) values 0.4,3 and 10.

The mixed oxide material specified is neutronically similar to $U(30)O_2$ for which data has been published[3]. In calculations for these materials, the KENO code[10] with the Hansen-Roach nuclear data library[11] had been used. A similar strategy was adopted in this case; a limited number of check calculations, using KENO with a 123gp working library of GAM-THERMOS data[12] processed through NITAWL[13], were also performed.

The 18 arrays which were calculated are specified in Table 1; they were chosen for their span in the $\sigma(m)$, m plane as shown in Figure 3 where the calculated unit masses required for array criticality are plotted. Each point shown was derived by interpolating the results of several KENO calculations within 0.02 in k-effective of the condition of array criticality. Each calculation in these individual series ran for some 10,000 neutron histories giving a quoted standard deviation of 0.008 approximately. The c_2 and m_0 values obtained by a least squares fit to the points on each line are given in Table 2.

Additional Monte-Carlo calculations for a few of the arrays predicted as critical by the method described above were performed. The arrays were chosen to span the characteristic line for mixed oxide at H:(U+Pu) of 3 in the $\sigma(m)$, m plane. KENO and MONK6.3[14] were used with 123 group and point data libraries; the results are shown in Table 3. In addition a KENO 123 group calculation for the predicted bare minimum critical mass gave a k-effective value of 1.006 (0.006).

The effect of array shape changes was investigated using KENO and 16 group Hansen Roach data. With the database as it stood prior to this study, array shape could be catered for by calculating the unit mass of U(93.2) metal required for array criticality and converting to the material of interest via the equivalence equation (eqn 4). This was the method used to calculate the unit mass required for criticality in the arrays shown in Table 4. The KENO results indicate that the higher the array shape factor, the greater is the discrepancy. A similar set of results were obtained for $U(30)O_2$ at H:U of 0.4, 3 and 10 in the 128x2x2 array.

It is possible to derive a relationship analogous to equation 5 for any material. A few additional KENO calculations, shown in Table 5, enabled this to be undertaken in a preliminary fashion for the mixed oxide material at H:(U+Pu) of 3. The resulting expression, which enables a more direct estimate of the appropriate c_2 value, is,

$$m' = 7.64\ R^{0.845} \qquad (8)$$

4 APPLICATIONS TO SNM TRANSPORT AND STORAGE

4.1 SNM movements

The movement of SNM within buildings occurs in trolleys which are designed to maintain a centre to centre spacing between units of 40cm. The minimum distance to any reflecting surface outside the trolley is 20cm. During any movement along a building corridor the ceiling and walls on at least 2 sides will be a metre or more away. People pushing the trolleys represent a limited degree of reflection and interstitial moderation. The trolleys cannot be stacked. The model adopted to encompass this rather indeterminate transport environment was a planar storage array, in particular an 8x8x1 array of trolleys with full water reflection around the array as a whole. The units themselves were treated as air spaced, the presence of moderating jackets, such as neutron shields, being evaluated at a later stage in the analysis.

The unit mass for array criticality may be calculated using the Limiting Surface Density Method for the range of materials shown in Table 6. These masses are the equivalents of 29.21 kg U(93.2) required for criticality in the model array. The reduction in unit mass required to reach the desired value of array k-effective was arrived at by applying the same reduction in radius is required for a single bare spherical unit[15]; a k-effective of 0.86 was chosen for the metal array in order to make the unit mass less than the value for a fully water reflected single unit. Separate KENO calculations, shown in Table 5, were used to establish the relationship between unit k-effective and radius. The approximation of r/r_c for k-effective was adequate for all units except those of plutonium metal and mixed oxide at a H: Actinide of 10.

The k-effective values corresponding to the unit masses shown in Table 6 were calculated directly using MONK6.3 with point data derived from the United Kingdom Nuclear Data Library and KENO4 with the 16 group Hansen Roach library; these results are also shown in the Table. The reflector was modelled explicitly in both cases. The MONK6.3 output includes a value

for the mean neutron energy causing fission which for these cases was of the order of hundreds of keV.

A direct comparison of unit masses of plutonium oxide and mixed oxide, producing the same array k-effective, may be made at H:Pu ratios of 3:1 and 10:1. It is interesting to note that less plutonium, or fissile, is present in the mixed than in the pure form. This is not a general result but will depend upon the array and unit cell parameters. For example, if the unit cell side were to be doubled the reverse would be true. There will exist in the $\sigma(m)$, m plane a family of arrays for which the plutonium contents of mixed and pure oxides match exactly.

The $\sigma(m)$, m plane representations of the different arrays indicate that the plutonium oxide, (H:Pu = 1, compound density = 3 g/cm^3), array is the most strongly coupled; conversely, the plutonium metal array is most weakly coupled, the units being close to a fully water reflected minimum critical mass. Therefore, it is the latter array which is most susceptible to the presence of hydrogeneous jackets, such as neutron shields, within the trolley itself. This is confirmed by the results shown in Figure 4 where the two limiting arrays are examined from a k-effective of 0.86.

The results shown in Figure 4, Tables 5 and 6 form the basis of the full criticality safety assessment, a description of which is beyond the scope of this paper.

4.2 SNM storage cell

In this hypothetical storage cell cans of plutonium oxide are placed end to end in an essentially open channel. The storage cell contains a 9 x 4 array of parallel, horizontal channels. A study of cell k-effective as a function of package diameter, Pu240 content, moisture content and oxide density was performed with full water reflection around the air spaced array.

Initially, the oxide was assumed to be $Pu(100)O_2$ with admixed water providing a hydrogen to plutonium ratio of 3. The packages were assumed to have a diameter of 5.375" (13.65 cm) and a channel of packages was modelled as a continuous cylinder of powder. The results of a set of calculations at a H:Pu of unity are shown together with the corresponding KENO4 and MONK 6.3 results in Table 7; the differential albedo option in KENO was used.

The results of parametric surveys are shown in Figure 5, where the reactivity loss caused by the various changes may be seen. The extension of the study, using Monte-Carlo methods to include reflection by concrete, is beyond the scope of this paper. It is sufficient to note that this may be done in such a way as to extend the Limiting Surface Density database.

5 CONCLUSIONS

The Limiting Surface Density method is applicable to a range of problems associated with the transport and storage of Special Nuclear

Materials. Its utility has been proven in a number of criticality assessments for Sellafield plant.

In this paper the scope of the method has been extended via the production of data for mixed oxide materials. The results obtained using the method have compared well with those given directly by two independent Monte Carlo codes, KENO 4 and MONK 6.3, with a range of nuclear data libraries.

Unfortunately, it is not possible to compare these results with experiment because appropriate data is not available. The results obtained for plutonium and mixed oxides should therefore be regarded as theoretical predictions whose validity has yet to be established experimentally.

6 ACKNOWLEDGEMENT

We are indebted to Dr W H Williams whose results we quote in Table 3, and to other BNFL colleagues for their encouragement.

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[15] The effect of reflector location on array criticality. J T Thomas ORNL/NUREG/CSD TM-16 (1980). Appendix.

TABLE 1: Arrays selected for mixed oxide calculations

Number of Units	$2a_n$ (cm)	$\sigma(m)/m$ cm^{-2}	Number of Units	$2a_n$ (cm)	$\sigma(m)/m$ cm^{-2}
1000	25	$1.545\ 10^{-2}$	729	30	$9.597\ 10^{-3}$
216	30	$6.177\ 10^{-3}$	512	40	$4.76\ 10^{-3}$
125	35	$3.69\ 10^{-3}$	343	45	$3.255\ 10^{-3}$
216	45	$2.745\ 10^{-3}$	216	50	$2.224\ 10^{-3}$
216	55	$1.838\ 10^{-3}$	125	55	$1.494\ 10^{-3}$
125	60	$1.256\ 10^{-3}$	343	75	$1.172\ 10^{-3}$
64	60	$9.636\ 10^{-4}$	512	95	$8.439\ 10^{-4}$
1000	120	$6.705\ 10^{-4}$	729	130	$5.111\ 10^{-4}$
343	130	$3.9\ 10^{-4}$	64	120	$2.409\ 10^{-4}$

TABLE 2: c_2 and m_o values used in the analysis

Material	H:Actinide	c_2 cm $^{-2}$	m_o kg
Mixed Oxide[a]	0.4	$7.21\ 10^{-4}$	131.4
	2.42	$7.0\ 10^{-4}$	93.0[b]
	3	$6.91\ 10^{-4}$	85.4
	10	$6.51\ 10^{-4}$	46.5
Plutonium metal	0	$4.346\ 10^{-3}$	9.95[c]
Plutonium Oxide[d]	1	$1.34\ 10^{-3}$	29.7[e]
	3	$1.113\ 10^{-3}$	28.65[c]
	10	$0.965\ 10^{-3}$	20.21[c]
Plutonium Oxide[f]	1	$1.32\ 10^{-3}$	32.6[e]

a PuO_2/UO_2, 40% Pu/(U+Pu) by weight

b interpolated

c reference 3

d $Pu(100)O_2$ ie 100% ^{239}Pu by weight

e interpolated using data from reference 3

f $Pu(94.8)O_2$ ie 94.8% ^{239}Pu, 5.2% ^{240}Pu by weight

TABLE 3: MONK6.3 and KENO 123 group calculations for cubic arrays of mixed oxide[a] predicted as critical

Number of Units	$2a_n$ (cm)	Unit mass (kg)	k-effective		
			KENO (123)	MONK6.3	
				(123)	(pt)
125	35	13.47	0.985 (0.006)	0.985 (0.005)	0.982 (0.006)
125	55	27.0	0 997 (0.006)	0.998 (0.005)	0.988 (0.006)
64	120	63.32	1.006 (0.006)	1.016 (0.005)	1.001 (0.006)

a PuO_2/UO_2, 40% Pu/(U+Pu) by weight

TABLE 4: The effect of changing the array shape[a] for mixed oxide[b] units

H:Actinide	Array	Shape factor	Unit mass[c] (kg)	k-effective[d]	Unit mass[e] at critical (kg)
0.4	8x 8x8	1.0	12.22	-	-
	32x 4x4	1.42	16.01	0.985	-
	16x16x2	1.67	18.23	1.001	-
	32x 8x2	1.75	18.96	0.993	-
	128x 2x2	2.69	27.18	0.963	-
3	8x 8x8	1.0	7.64	-	-
	32x 4x4	1.42	10.01	0.981	-
	16x16x2	1.67	11.4	0.985	-
	32x 8x2	1.75	11.87	0.977	-
	128x 2x2	2.69	17.06	0.953	-
	64x 8x1	3.04	19.05	-	21.0
	128x 4x1	3.35	20.84	-	23.7
	256x 2x1	4.01	24.68	-	28.5
10	8x 8x8	1.0	3.94	-	-
	32x 4x4	1.42	5.17	0.991	-
	16x16x2	1.67	5.9	1.002	-
	32x 8x2	1.75	6.14	0.995	-
	128x 2x2	2.69	8.86	0.969	-

a a_n = 16.454 cm
b PuO_2/UO_2, 40% Pu/(U+Pu) by weight
c Limiting Surface Density calculation
d Calculated using KENO and 16 group Hansen Roach Library
e Interpolated from several KENO calculations

TABLE 5: k-effective as a function of radius for bare units

r/r_c	Plutonium Metal (sph)	Plutonium Oxide (spherical units)		
		H:Pu 1	H:Pu 3	H:Pu 10
1.0	1.002 (0.006)	0.994 (0.005)	0.985 (0.006)	0.990 (0.005)
0.9	0.918 (0.006)	0.911 (0.005)	0.904 (0.005)	0.888 (0.005)
0.8	0.824 (0.005)	0.800 (0.005)	0.792 (0.006)	0.797 (0.005)
0.7	0.725 (0.005)	0.694 (0.005)	0.675 (0.006)	0.658 (0.005)
0.6	0.634 (0.005)	0.584 (0.005)	0.560 (0.005)	0.535 (0.005)
	Plutonium Oxide (cyl) H:Pu 1 h:d 2.5	Mixed Oxide (spherical units)[a]		
		H:Act 0.4	H:Act 3	H:Act 10
1.0	1.004 (0.006)	1.004 (0.006)	0.983 (0.007)	1.002 (0.008)
0.9	0.903 (0.005)	0.893 (0.006)	0.912 (0.008)	0.927 (0.006)
0.8	0.803 (0.005)	0.803 (0.006)	0.796 (0.007)	0.828 (0.008)
0.7	0.703 (0.004)	0.688 (0.006)	0.678 (0.005)	0.718 (0.006)
0.6	0.586 (0.004)	0.573 (0.005)	0.567 (0.006)	0.582 (0.007)

[a] PuO_2, 40% Pu/(U+Pu) by weight

TABLE 6: Results for the 8x8x1 array

Material	H:(U+Pu)	Compound Density (g/cm^3)	Unit[a] Mass (kg)	Array k-effective	
				KENO (16)	MONK6.3 (pt)
Plutonium metal[b]	0	19.8	4.48	0.864(0 007)	0.855(0.005)
Plutonium Oxide[c]	1	3	18.66	0.88 (0.008)	0.858(0.005)
	1	5	15.54	0.901(0.008)	0.884(0.005)
	1	8.3	10.65	0.899(0.008)	0.889(0.006)
	3	5.35	9.33	0.890(0.007)	0.881(0.005)
	10	2.385	6.22	0.893(0.007)	0.882(0.006)
Mixed Oxide[d]	0.4	9.71	32.8	0.892(0.007)	0.898(0.006)
	2.42	5.873	22.8	0.891(0.008)	0.905(0.006)
	3	5.275	20.73	0.888(0.009)	0.893(0.006)
	10	2.366	10.0	0.888(0.007)	0.885(0.006)

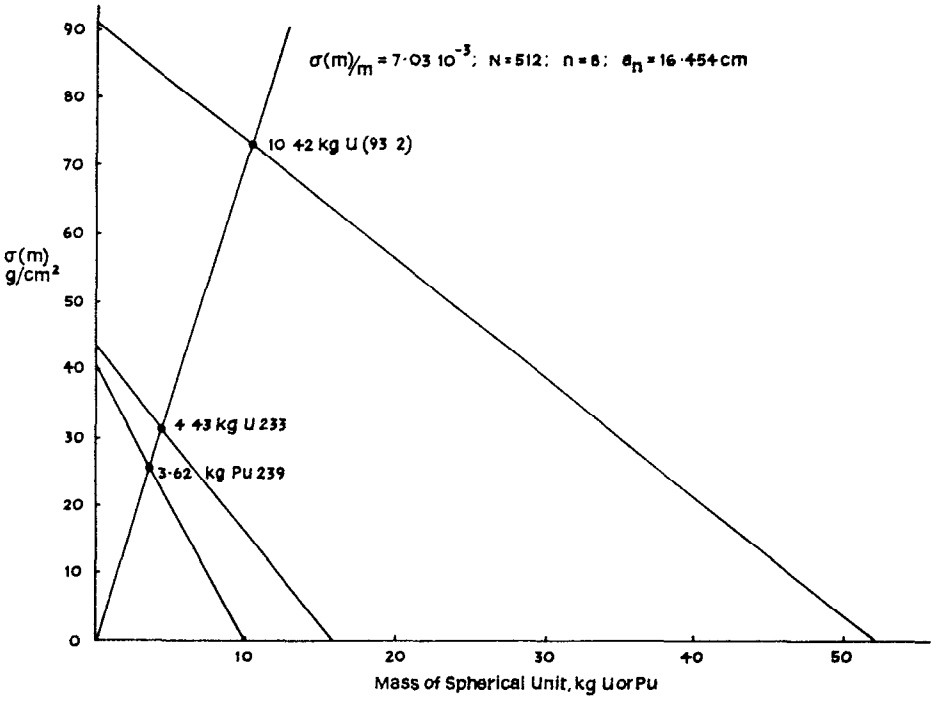
[a] kg U + Pu
[b] k-effective 0.86 by Limiting Surface Density
[c] $Pu(100)O_2$: k-effective 0.9
[d] PuO_2/UO_2, 40% Pu/(U+Pu) by weight: k-effective 0.9

TABLE 7: Array k-effective values

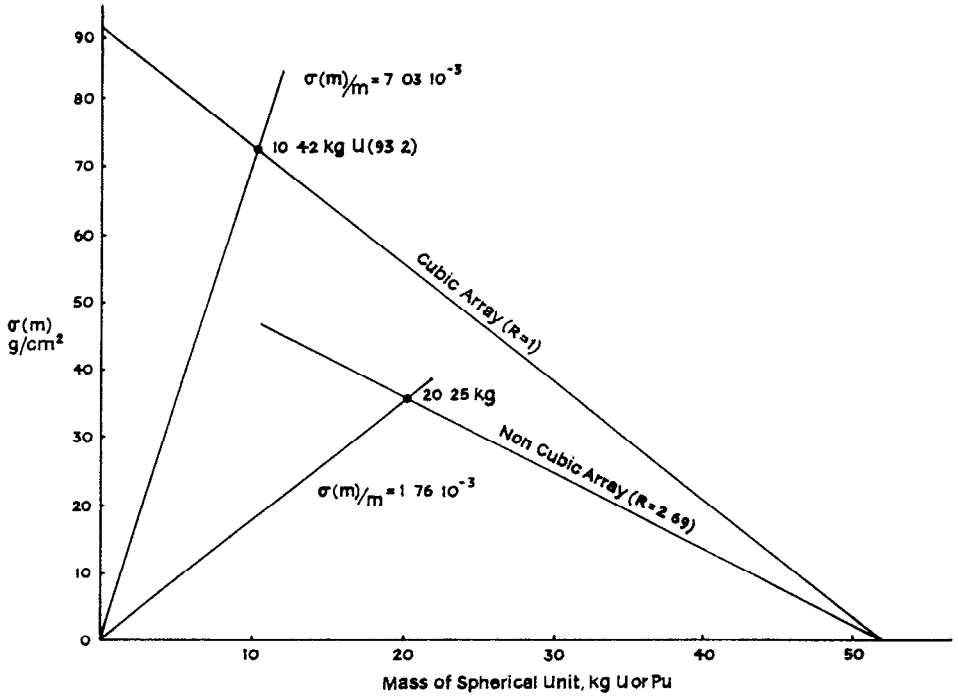
Oxide density[a] g cm^{-3}	k-effective		
	Limiting Surface Density	KENO4 (16)	MONK6.3 (pt)
2	0.71	0.681 (0.007)	0.67 (0.005)
3	0.82	0.807 (0.005)	0.793 (0.006)
4	0.92	0.929 (0.008)	0.906 (0.005)
5	1.00	1.033 (0.008)	1.016 (0.006)

[a] $Pu(100)O_2$ H:Pu = 1

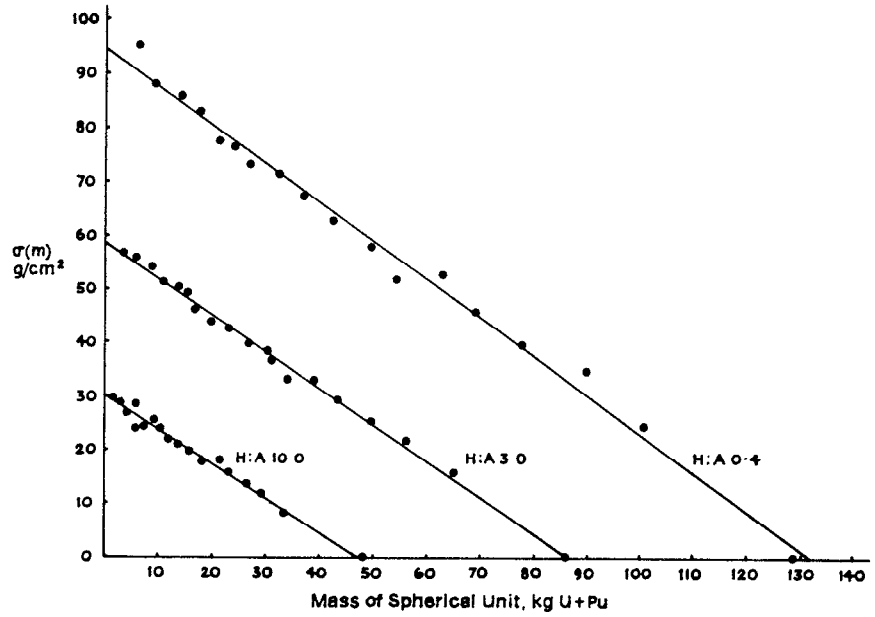
1 Equivalent masses of U(93.2), ^{233}U and ^{239}Pu metal spherical units.



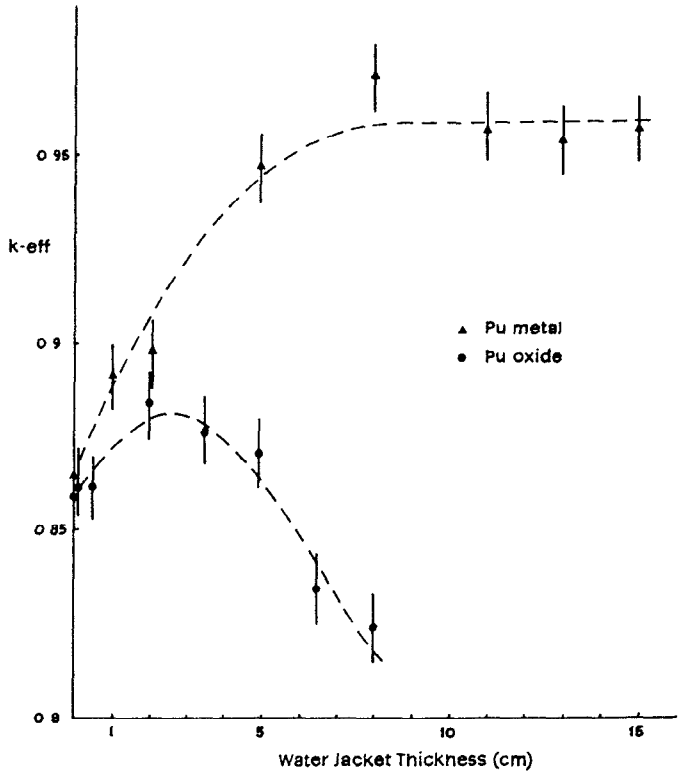
2 Representation of a change in array shape.



3 Mixed Oxide cubic array calculations in the σ(m),m plane.



4 k-effective as a function of water jacket thickness for limiting cases.



5 Cell k-effective as a function of package diameter, density, moisture and ^{240}Pu content.