D. Mixtures of Nuclides

1. Mixtures of ^{233}U, Carbon, Water, and Thorium

When the ^{233}U-Th reactor fuel cycle was considered more seriously than at present, Thomas made a computational criticality survey of combinations that might be encountered in fuel processing.[90] Thomas covered mixtures of $^{233}UO_2$, ThO_2, carbon and water over a range of ^{233}U densities and at Th/U ratios of 0, 1, and 4. Water-reflected critical spherical masses and radii of infinite cylinders are given as functions of ^{233}U density.

2. ^{235}U-Water-Graphite Mixtures

During the life of the Rover propulsion-reactor project, the need to process graphite-moderated fuel led to a computational criticality survey of U(93) metal-water-graphite mixtures. This survey, reported by Stratton,[81] gives critical sphere masses and volumes, diameters of infinite cylinders, and thicknesses of infinite slabs over ranges of U density, H/U ratio, and C/U ratio, and two thicknesses of water reflector. Subsequently, calculated subcritical limits for U(93.5) metal-water-graphite systems were re-examined and appear in Table 11.

Table 11

Subcritical Limits for Spheres, Cylinders and Slabs of U(93.5) Metal-Water-Graphite Mixtures

H/U	Density (kg U/L)	2.5-cm-Thick Water Reflector				30-cm-Thick Water Reflector			
		Sphere		Cylinder	Slab	Sphere		Cylinder	Slab
		Mass[a] (kg U)	Volume (L)	Diameter (cm)	Thickness (cm)	Mass[a] (kg U)	Volume (L)	Diameter (cm)	Thickness (cm)
					C/U = 0				
0	18.8	29.5	1.56	9.43	3.80	21.0	1.00	7.16	1.31
5	4.09	22.2	5.44	14.7	6.83	13.2	3.24	11.1	2.96
50	0.508	3.55	6.99	16.2	7.80	2.22	4.37	12.6	4.18
300	0.0867	0.991	11.4	19.5	10.0	0.705	8.14	16.4	6.96
1500	0.0174	2.75	158.	49.4	29.2	2.35	135.	46.0	26.3
					C/U = 20				
0	1.69	141.	83.7	39.0	21.7	80.8	47.7	29.6	12.0
5	1.28	46.2	36.1	29.0	15.4	26.4	20.6	21.9	8.13
50	0.399	4.22	10.5	18.8	9.33	2.61	6.55	14.7	5.15
300	0.0828	1.02	12.3	20.0	10.3	0.730	8.81	16.9	7.21
1500	0.0172	2.77	160.	49.5	29.4	2.38	138.	45.5	25.9
					C/U = 100				
0	0.365	104.	285.	59.8	35.1	63.5	174.	47.6	23.0
5	0.341	54.0	158.	48.7	28.0	32.4	95.0	38.3	17.7
50	0.215	5.97	27.8	26.5	14.0	3.70	17.2	20.9	8.48
300	0.0703	1.15	16.3	22.1	11.6	0.822	11.7	18.7	8.23
1500	0.0166	2.83	170.	50.6	30.0	2.42	145.	47.3	27.0
					C/U = 200				
0	0.184	74.8	406.	67.5	40.0	46.8	254.	54.5	27.4
5	0.178	45.3	254.	57.5	33.6	28.1	157.	46.1	22.5
50	0.136	7.05	51.8	33.0	18.1	4.40	32.3	26.4	11.6
300	0.0591	1.29	21.8	24.5	13.1	0.925	15.6	20.8	9.43
1500	0.0159	2.91	183.	51.8	30.9	2.49	156.	50.8	27.7
					C/U = 500				
0	0.0741	38.9	524.	73.7	43.9	25.3	341.	60.8	31.7
5	0.0731	28.7	393.	66.7	39.5	18.6	255.	54.9	28.3
50	0.0649	7.84	120.	44.4	25.3	5.11	78.7	36.4	17.7
300	0.0400	1.62	40.6	30.5	16.8	1.17	29.3	26.1	12.5
1500	0.0141	3.13	222.	55.5	33.2	2.69	191.	51.4	30.0
					C/U = 1000				
0	0.0371	21.4	576.	76.1	45.4	14.4	390.	63.9	34.1
5	0.0369	18.0	488.	71.9	42.7	12.1	330.	60.4	32.0
50	0.0347	7.46	215.	54.2	31.5	5.06	145.	45.5	23.4
300	0.0260	1.99	76.5	38.0	21.5	1.46	56.1	32.9	16.7
1500	0.0119	3.50	295.	61.1	36.8	3.05	257.	56.0	33.4

[a] Mass as total uranium.

3. Plutonium-Uranium Mixtures

Aqueous Mixtures[71, 91]

Standard *ANSI/ANS-8.12* gives subcritical limits for individual units of mixtures of plutonium and natural uranium. It is noted that the subcritical margin of these limits, $\Delta k_{eff} = 0.05$, includes no allowance for contingencies. Consequently, in application, there must be sufficient overall margin to protect against the limit being exceeded accidently.

These subcritical limits appear in Figure 14 for mass, Figure 15 for volume, Figure 16 for cylinder diameter, and Figure 17 for slab thickness. Again, the equivalent of full water reflection is assumed. Solid lines apply to solutions and effectively homogeneous* aqueous mixtures. Dashed lines apply to optimum lattices of rods in water, and may be applied conservatively to other distributions of small pieces in water.

*Particles in a slurry should be uniformly distributed and have a diameter no larger than 0.127 mm (0.005 in.), i.e., are capable of being passed through a 120-mesh screen.[71]

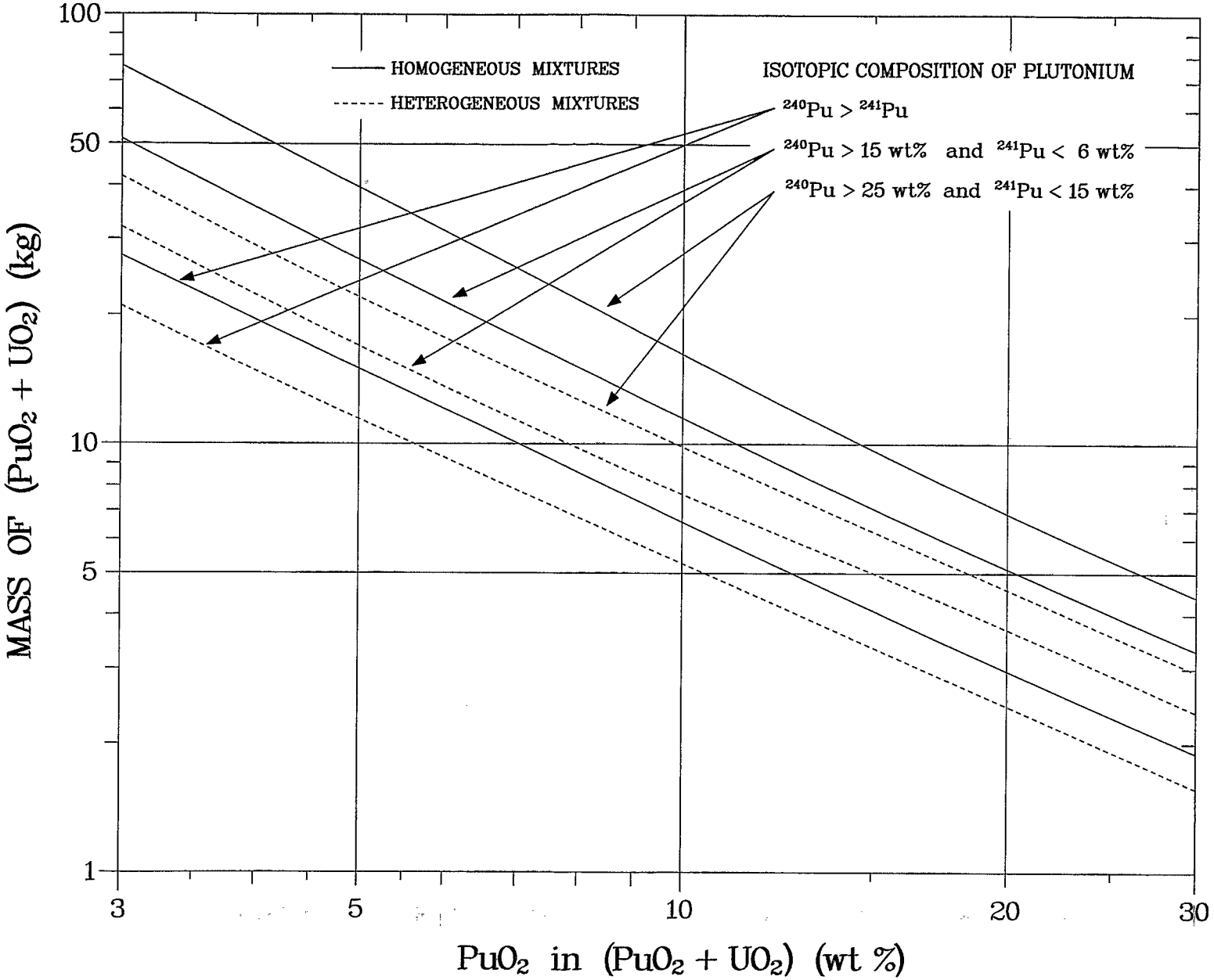

Figure 14

Fig. 14. Subcritical mass limits for water-reflected spheres of aqueous mixtures of PuO_2 and $U(0.7)O_2$.

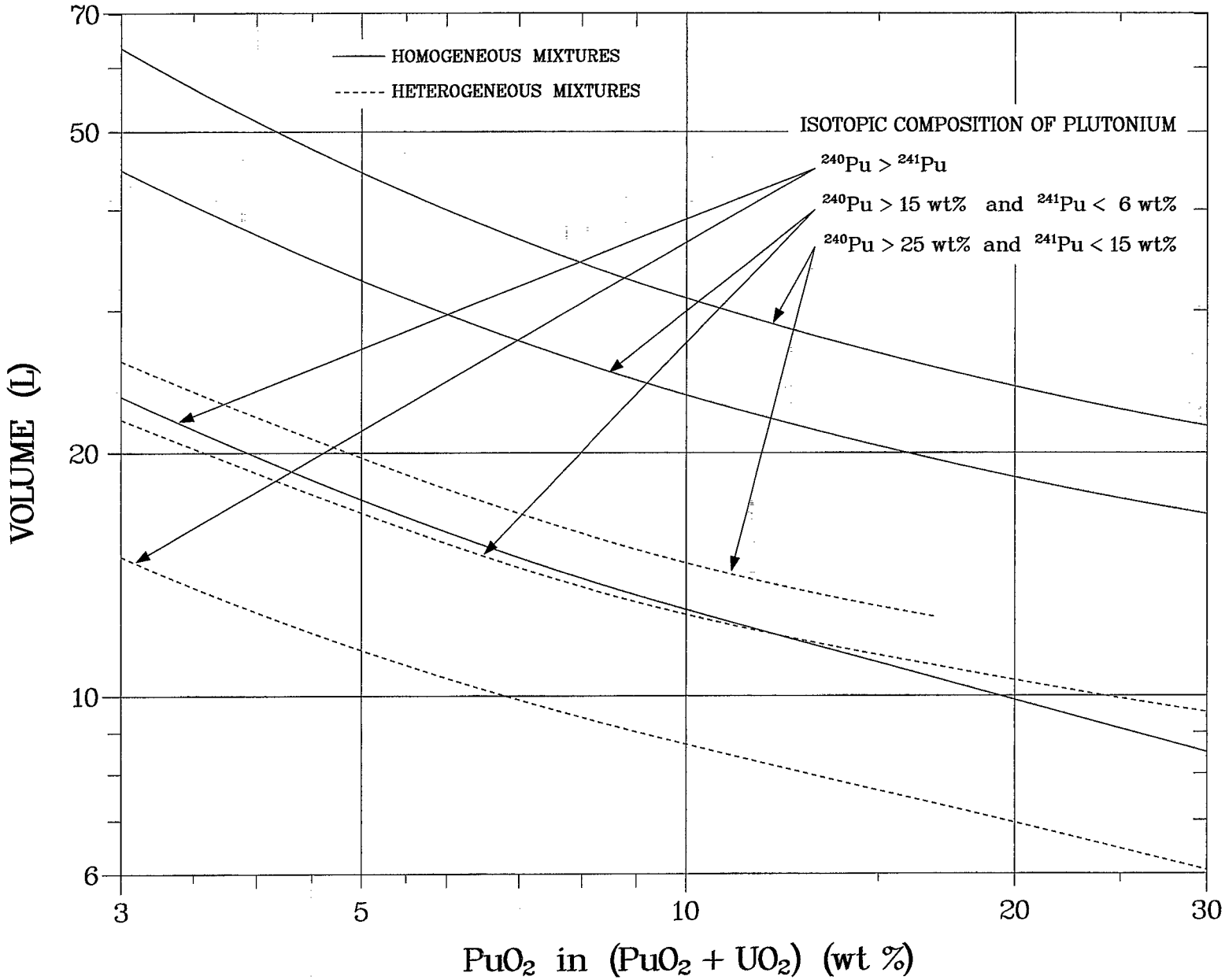

Figure 15

Fig. 15. Subcritical volume limits for water-reflected spheres of aqueous mixtures of PuO_2 and $U(0.7)O_2$.

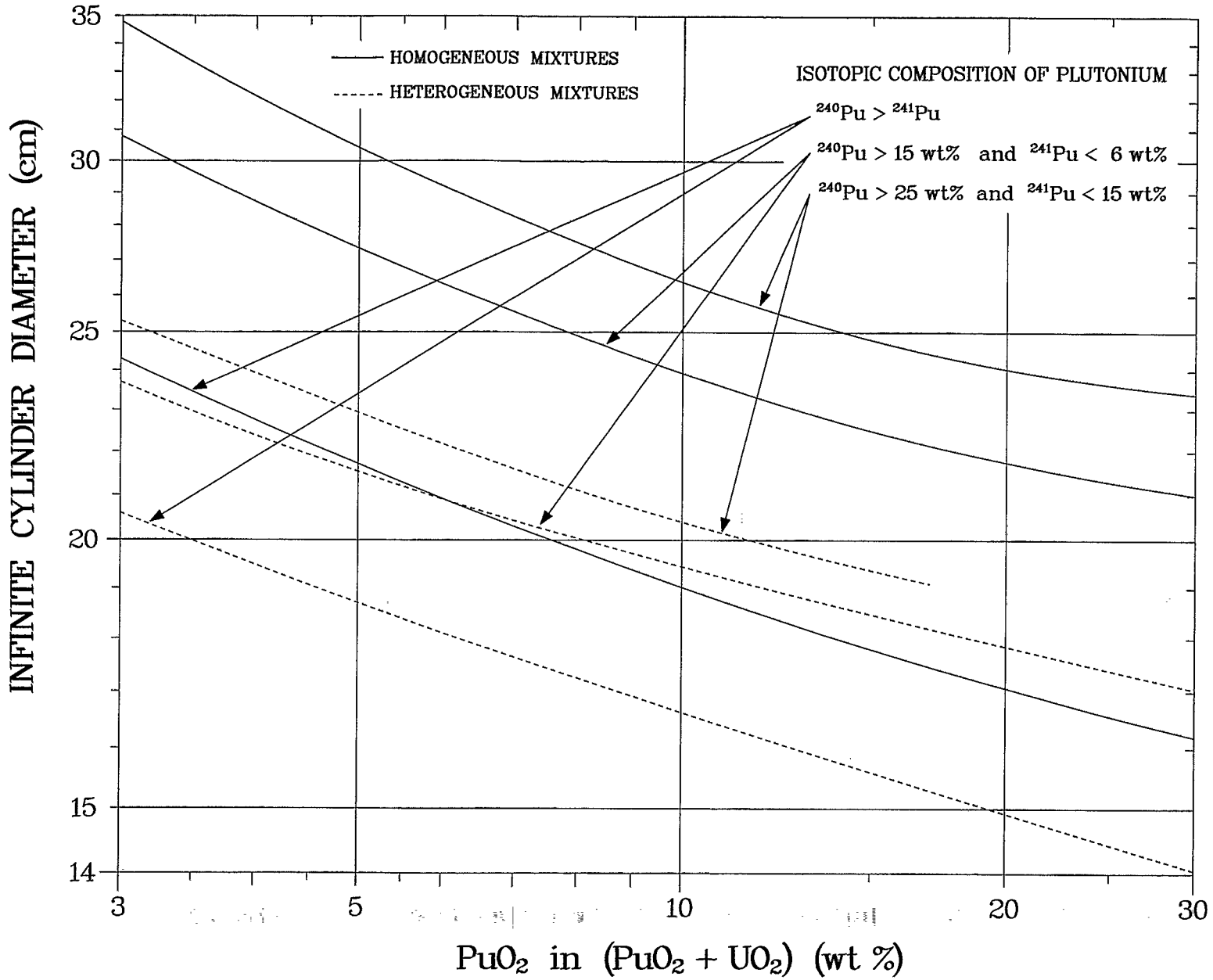

Fig. 16. Subcritical diameter limits for water-reflected infinite cylinders of aqueous mixtures of PuO_2 and $U(0.7)O_2$.

Figure 16

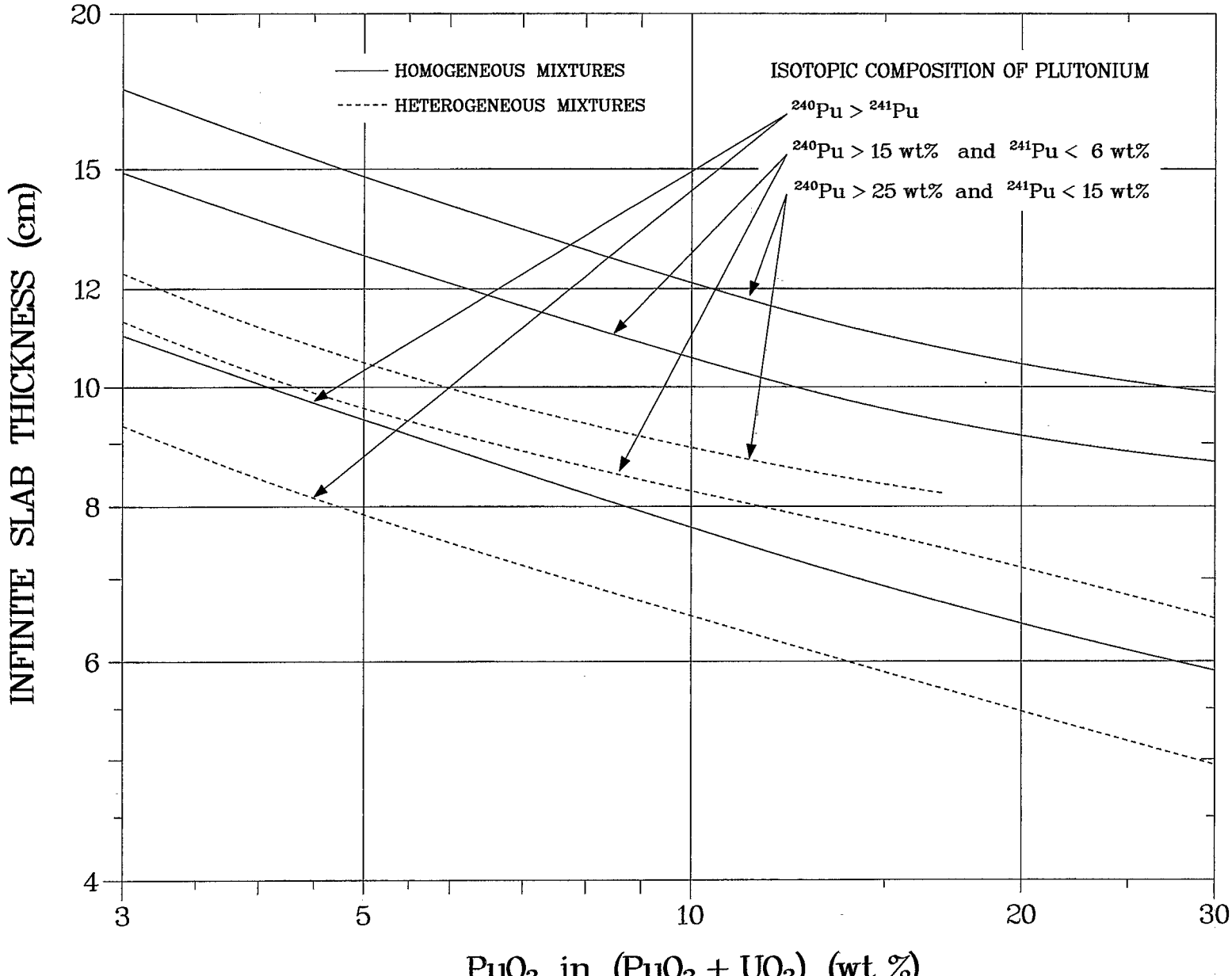

Fig. 17. Subcritical thickness limits for water-reflected infinite slabs of aqueous mixtures of PuO_2 and $U(0.7)O_2$.

Figure 17

Dry and Damp Mixed-Oxide Powders[71]

The subcritical mass limits given in Table 12 apply to dry and damp mixed oxides of plutonium and natural uranium. Again, the limits for damp oxide are provided because completely dry oxide may be difficult to maintain. These are for H/(Pu+U) $\leq$ 0.45 (1.48 wt% water). Also, limits are provided for oxides of half-theoretical density.

Table 12

Subcritical Mass Limits for Single Units of Mixed Oxides of Plutonium and Natural Uranium, Thick Water Reflector[a]

Material	PuO_2 in (PuO_2 + UO_2) (wt%)	Subcritical Limit	
		Mass of Pu (kg)	Mass of Oxides (kg)
Dry mixed oxides at density $\leq$ 11.0 g/cm^3	3	–[b]	–[b]
	8	122.0	1729.0
	15	47.0	355.0
	30	26.1	98.6
Damp mixed oxides at density $\leq$ 9.4 g/cm^3 H/(Pu + U) $\leq$ 0.45	3	236.0	8919.0
	8	49.4	700.0
	15	32.9	249.0
	30	23.3	88.1
Damp mixed oxides at half density[c] $\leq$ 4.7 g/cm^3 H/(Pu + U) $\leq$ 0.45	3	885.0	33,447.0
	8	161.0	2282.0
	15	102.0	771.0
	30	67.9	256.6

[a] Masses given are for the Pu contained in the mixed oxide, and for the permissible quantity of PuO_2 + UO_2. The limits apply to mixed oxides of ^{239}Pu and natural uranium (^{235}U $\leq$ 0.72 wt%).

[b] Subcritical in any amount

[c] CAUTION: Application of these limits requires that the total oxide density be less than 4.7 g/cm^3.

Subcritical Plutonium Content for Unlimited Quantities of Plutonium and Natural Uranium Mixtures[71]

Either of two conditions results in subcriticality for unlimited quantities of plutonium-natural uranium mixtures. One condition is sufficient dilution of plutonium by uranium for $k_\infty \leq 1$. Material for which k_∞ (Ref. 9) is less than unity will be subcritical regardless of the mass, volume, shape, or reflector condition. Table 13 gives subcritical limits of Pu in (Pu + U) for unlimited quantities of dry and aqueous oxide mixtures and nitrate solutions. For example, a homogeneous mixture of PuO_2 and UO_2 in water cannot achieve criticality if the plutonium content does not exceed 0.13 wt% of the total (Pu + U). Table 13 is not applicable to (Pu + U) metal and water mixtures.

Table 13

Subcritical Limits for Plutonium in Plutonium and Natural Uranium Mixtures of Unlimited Mass

Mixture of Pu +U	wt% Pu in (Pu + U)
Dry oxides, H/(Pu + U) = 0	4.4
Damp oxides, H/(Pu + U) $\leq$ 0.45	1.8
Oxides in water	0.13
Nitrate solution	0.65

The second condition is the dilution of plutonium by sufficient water that neutron absorption by hydrogen will maintain $k_\infty < 1$. Guidance for uniform aqueous mixtures of the oxides of natural uranium and plutonium is provided in Table 14 for three isotopic compositions of plutonium. The particle size limitation stated earlier applies; i.e., less than 0.127 mm. The limits are given for four compositions of plutonium expressed as wt% PuO_2 in the oxides and are specified for each of three controllable parameters.

These parameters are the mass of plutonium per unit volume, the minimum H/Pu atomic ratio, and the mass of combined oxides per unit volume. When there is less than 3 wt% PuO_2 in the oxides, the subcritical limit of 6.8 g Pu/L in Table 14 must be reduced because of the increased relative importance of ^{235}U as the proportion of uranium increases. Oxides having compositions between 0.13 and 3 wt% PuO_2 must be treated as special cases. If the Pu in (Pu + U) composition of the oxides is less than 0.13 wt%, criticality is not possible, as noted in Table 13.

Table 14

Limiting Subcritical Densities of Unlimited Volumes of Uniform Aqueous Mixtures[a] of PuO_2 and UO_2($^{235}U \leq 0.72$ wt%)

PuO_2 in ($PuO_2 + UO_2$) (wt%)	Pu Isotopic Content[b]	H/Pu Atomic Ratio[c]	Pu Density (g/L)	($PuO_2 + UO_2$) Density (g/L)
3	I	3780	6.8[d]	257.
	II	3203	8.1	305.
	III	2780	9.3	351.
8	I	3780	6.9	97.
	II	3210	8.2	116.
	III	2780	9.4	134.
15	I	3780	7.0	52.9
	II	3237	8.2	61.7
	III	2818	9.4	71.0
30	I	3780	7.0	26.5
	II	3253	8.1	30.7
	III	2848	9.3	35.2

[a] These limits also apply to solutions of plutonium and natural uranium compounds, provided all specified conditions are satisfied.

[b] Plutonium isotopic content:

I $\rightarrow$ $^{240}Pu > {}^{241}Pu$

II $\rightarrow$ $^{240}Pu \geq 15$ wt% and $^{241}Pu \leq 6$ wt%

III $\rightarrow$ $^{240}Pu \geq 25$ wt% and $^{241}Pu \leq 15$ wt%

[c] Lower limit.

[d] This density limit is not applicable to oxide mixtures in which the $PuO_2/(PuO_2 + UO_2)$ ratio is less than 3 wt%.

E. Special Geometries

1. Annular Cylinders

Experiments at Oak Ridge[92–93] and Valduc, France,[94] on the criticality of solutions in annuli provide the basis for potential applications described in Section C of Chapter IV, **Storage of Solutions**. All annuli consisted of the space between two coaxial cylinders, with the central cylinder lined with cadmium and filled with water. The Oak Ridge experiments established critical heights of $U(93)O_2F_2$ solutions at $H/^{235}U = 50.4$ or 309 in annuli of various thicknesses and ranging from 25.4-cm to 76.2-cm-o.d. These experiments were carried out with and without an external water reflector.

At Valduc, critical heights of plutonium solutions in water-reflected 50 cm o.d. annuli were determined. One set of data applies to low-^{240}Pu solution at densities from 130 to 190 g ^{239}Pu/L in an annulus of 30-cm-i.d. Another set applies to solutions of Pu containing 19 wt% ^{240}Pu, at about 50 to 165 g ^{239}Pu/L, in a 20-cm-i.d. annulus.

Critical experiments at Los Alamos on a tall 76-cm-o.d. and 57-cm-i.d. tank with various reflectors[95] were in support of upgraded fuel processing equipment at the Idaho Chemical Processing Plant.

Attention is called to the publication, *Criticality Experiments with Mixed Plutonium-Uranium Nitrate Solution at Plutonium Fractions of 0.2, 0.5, and 1.0 in Annular Cylindrical Geometry.*[96] The 21-inch-o.d. by 10-inch-i.d. annulus was reflected externally by water. Various inserts included bottles of solution surrounded by a variety of absorbers.

2. Pipe Intersections

Subcritical manifolds, consisting of pipe (arms) intersecting a larger diameter pipe (a column), are described in *American National Standard Nuclear Criticality Safety Criteria for Steel-Pipe Intersections Containing Aqueous Solutions of Fissile Material, ANSI/ANS-8.9.*[97] This Standard applies to ^{233}U solutions in 4-inch maximum pipe, ^{235}U or ^{239}Pu solutions in branched columns of 6-inch or less Schedule-10 or heavier pipe, and U(5) solutions in columns as large as 10-inch pipe. Reflector conditions applied to locations within a small enclosure with concrete walls, or complete water immersion. The Standard considers only single columns with intersections, and states, "Multiple columns or columns in the vicinity of other fissionable materials ... shall be investigated by experiment or by a validated computational technique."

Critical experiments with intersecting cylinders, at Rocky Flats and Oak Ridge, were involved in the validation of calculations providing data for *ANSI/ANS-8.9*. The Rocky Flats measurements all were with $U(93.1)O_2(NO_3)_2$ solution in 17.8-cm square columns with as many as 12 arms in three layers.[98] At Oak Ridge, concentrated $U(5.0)O_2F_2$ solution was in a 30° aluminum "Y" (both legs 27.9-cm-i.d.), or in Plexiglas "crosses" (26.7-cm and 27.3-cm-i.d.).[99] Results of earlier Oak Ridge measurements[92] with U(93) solution in a cross and "Y" (both 12.3-cm-i.d.) apparently were not used for validation. Any validation of calculations going beyond the Standard should be based on data from these experiments.

The Monte Carlo calculations[100] with 123-group cross sections that provided data for the Standard, averaged $k_{eff} = 0.9994 \pm 0.0027$ for the Rocky Flats critical experiments and 0.9999 ± 0.0022 for the Oak Ridge U(5) experiments. Calculations for ^{233}U and ^{239}Pu relied upon data for simple cylinders. The geometries that appear in the Standard were adjusted to $k_{eff} = 0.85$ for reflection by concrete walls and $k_{eff} = 0.90$ for water immersion.

F. Factors Affecting Limits of Individual Units

1. Abnormal Conditions

Appearing in Appendix A of Standard *ANSI/ANS-8.1*, and reproduced below, are examples of abnormal variations in process conditions. Such variations should be considered in establishing limits for criticality control. Contingencies, the dominant items, usually lead to practical limits with subcritical margins significantly greater than in the stated subcritical limits. The examples of contingencies, and other abnormal conditions to be considered, follow.*

- A change in intended shape or dimensions resulting from bulging, corrosion, or bursting of a container, or failure to meet specifications in fabrication.
- An increase in the mass of fissionable material in a location as the result of operational error, improper labeling, equipment failure, or failure of analytical techniques.
- A change in the ratio of moderator to fissionable material resulting from
    1. Inaccuracies in instruments or chemical analyses,
    2. Evaporating or displacing moderator,
    3. Precipitating fissionable material from solutions,
    4. Diluting concentrated solutions with additional moderator.
- A change in the fraction of the neutron population lost by absorption resulting from
    1. Loss of solid absorber by corrosion or by leaching,
    2. Loss of moderator,
    3. Redistribution of absorber and fissionable material by precipitation of one but not the other from solution,
    4. Redistribution of solid absorber within a matrix of moderator or solution by clumping,
    5. Failure to add the intended amount of absorber to a solution or failure to add it with the intended distribution,
    6. Failure of analytical techniques to yield correct ... concentrations.

*A group of examples applicable to multiple units has been deleted.

- A change in the amount of neutron reflection resulting from

    1. An increase in reflector thickness by adding ... material (e.g., water or personnel),
    2. A change in reflector composition such as loss of absorber (e.g., by corrosion of an outer casing of absorber).

The remainder of this section discusses the influence on criticality limits of conditions that may be normal in processes but are not included in any Standard.

2. Extended Subcritical Limits

Reduced Density of Fissile Cores

For a highly enriched uranium metal core in the equivalent of a thick water reflector, the critical mass varies as the core density (ρ) to the -1.4 power.[11] This relationship also applies to cores of plutonium metal and of the uranium compounds listed in Table 4. Under rare circumstances, subcritical masses of Tables 3 and 4 may be increased in accordance with $(\rho/\rho_o)^{-1.4}$ when ρ is less than the normal density ρ_o. Conditions that must be satisfied are that ρ differs from ρ_o only as a result of free space, that no moisture or other moderating material can enter the core, and that the unit cannot be compressed, for example, by compaction as a result of vibration.

Dilution of ^{235}U Metal Core

The relation discussed above does not apply when the density of fissile metal, ρ, is reduced by uniformly replacing a volume fraction of the metal (F), with an inert element. The volume fraction of the remaining fissile metal $(1 - F)$ equals ρ/ρ_o as defined above. If the diluting element has an atomic number Z within the range $11 \leq Z \leq 83$, the subcritical mass for ^{235}U in Table 3 may be increased[101] by the factor $(1 - F)^{-1.0}$, i.e., $(\rho/\rho_o)^{-1.0}$. This factor cannot be used if a moderating material is introduced into the mixture. This relation is a lower envelope for the diluting elements in ^{235}U with a natural uranium reflector and so would be conservative with a water reflector. With natural uranium as a diluting element, the measured factor $(1 - F)^{-0.7}$ increases with large F.[102]

3. Neutron Absorbers in Solutions of Fissile Material

As discussed in Chapter I, criticality in solutions of fissile material may be prevented by the proper addition of either solid or soluble neutron absorbers. In either case, it is important that intended distributions and densities of the absorbers be maintained. Examples of some elements that can be used as neutron absorbers are boron, chlorine, cadmium, and gadolinium.

Solid Neutron Absorbers

American National Standard Use of Borosilicate-Glass Raschig Rings as a Neutron Absorber in Solutions of Fissile Material, ANSI/ANS-8.5, provides guidance on the use of borosilicate-glass Raschig rings as neutron absorbers for criticality control in plants processing fissile material. It specifies subcritical density limits for uranium and plutonium in vessels of unlimited size when packed with borosilicate-glass rings. The recommended limits are summarized in Table 15. Several examples of systems that go beyond the limits of Table 15, including plutonium-uranium solution mixtures, are mentioned in Chapter V.

Possible use of tanks loaded with borosilicate-glass Raschig rings for solution storage is discussed in Section C of Chapter IV and an example is provided in Chapter V. Also in Chapter V is an example of boron introduced heterogeneously as boron-loaded stainless steel rings.

Although not usually thought an absorber, the borosilicate-glass pipe commonly used for solution storage columns reduces the effectiveness of a surrounding water reflector, as does steel pipe. The specified minimum wall thickness of nominally 6-inch-diameter Pyrex pipe is 0.71 cm. Assuming this value and that thick water is the only external reflector to be considered, the value of the limit on cylinder diameter (from Table 1) may be increased to 18.5 cm for ^{235}U. The value for ^{239}Pu may be increased to 20.7 cm provided the nitrogen to plutonium atomic ratio is no less than 4. It may be noted that the 15.8 cm (6.20 inch) maximum inside diameter of nominally 6-inch-diameter Pyrex pipe is well below these limits.

Table 15

Maximum Permissible Densities of Solutions[a] of Fissile Material in Vessels of Unlimited Size Packed With Borosilicate-Glass Raschig Rings

Isotopic Composition	Maximum Density[b] in Vessels With Minimum Glass Content of:		
	24 vol %	28 vol %	32 vol %
1. 0.0 wt% $<$ ^{233}U $\leq$ 100 wt% (g U/L)	150[c]	180[c]	200[c]
2. 5.0 wt% $<$ ^{235}U $\leq$ 100 wt%; ^{233}U $\leq$ 1 wt% (g U/L)	270	330	400
3. 0.7 wt% $\leq$ ^{235}U $\leq$ 5.0 wt%; ^{233}U = 0 wt% (g U/L)	unrestricted	unrestricted	unrestricted
4. ^{239}Pu $\geq$ 50 wt%, ^{241}Pu $\leq$ 15 wt% and ^{240}Pu $>$ ^{241}Pu			
a. $\leq$ 5 wt% ^{240}Pu (g Pu/L)	115	140	180
b. $>$ 5 wt% ^{240}Pu (g Pu/L)	140	170	220

[a] The density of the hydrogen in the solution shall be not less than 75 g/L and not greater than 115 g/L.

[b] Any fissile material deposited as solids shall be included.

[c] These limits also apply to mixtures of ^{233}U and other uranium isotopes, including ^{235}U, provided the ^{233}U content is greater than 1 wt% of all the uranium.

Soluble Neutron Absorbers

Any use of a soluble absorber for criticality prevention requires confirmation that the absorber be uniformly distributed in the fissile solution and that it cannot precipitate.

Calculations based on experiments carried out at Hanford,[103–104] provide densities of gadolinium in plutonium solutions required for k_∞ to be less than unity.

Boron content in aqueous uranium solutions required to reduce k_∞ to less than unity also have been calculated. The calculations were substantiated by experiments[105–106] with enrichments not exceeding 5 wt% ^{235}U. Figure 18 gives the minimum $B/^{235}U$ atomic ratio required to result in k_∞ less than unity for any moderation and for any ^{235}U enrichment up to 5 wt%.

Calculations[81] based on experiment[107] indicate that the presence of one atom of boron for each atom of ^{235}U will maintain large volumes of solution subcritical for ^{235}U densities less than 400 g/L. A boron-to-^{235}U atomic ratio of 1.5 is sufficient to maintain subcriticality up to a ^{235}U density of 1000 g/L.

Hanford critical experiments in support of the design of dissolvers at the Idaho Chemical Processing Plant[108] established the effect of cadmium nitrate dissolved in $U(85)O_2(NO_3)_2$ solution or its water reflector.

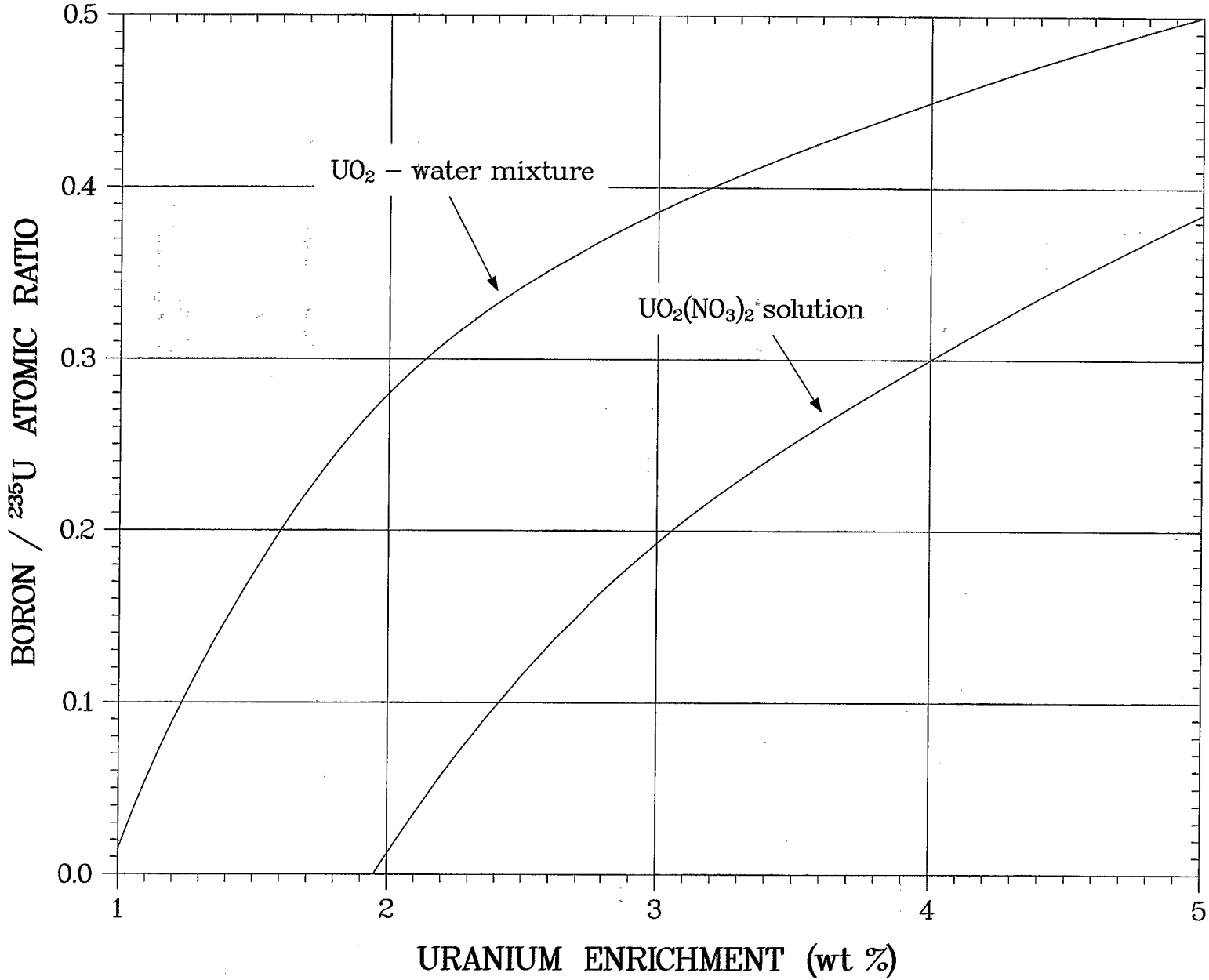

Figure 18

Fig. 18. Boron-to-^{235}U atomic ratio for subcriticality of solutions of $UO_2(NO_3)_2$ and UO_3-water mixtures for uranium containing no more than 5 wt% ^{235}U.

4. Concrete as a Reflector

As noted before, concrete as a neutron reflector is significant because its effectiveness may exceed that of water.[109–111] Although the composition of concrete is variable, changes in its effectiveness as a reflector are minor within the typical density range of 2.1 to 2.4 g/cm^3 and as the water content ranges from 3 to 10 wt%. As a reflector, water is more effective for thicknesses less than 10 cm whereas concrete is more effective for thicknesses greater than 10 cm. Consequently, for closely fitting concrete 10 cm or less in thickness, the single unit limits specified in Chapter III for thick water reflection may be used.

Closely fitting concrete greater than 10 cm in thickness requires a reduction of the thick water reflector limits. The limits should be multiplied by the following factors:

- 0.90 for mass and volume,
- 0.80 for diameter of infinite cylinder, and
- $0.44\rho^{-0.155}$ for thickness of infinite slab, where ρ is the fissile material density in g/cm^3.

A vessel often may be located in a concrete cell or in the vicinity of a concrete wall. Guidance is available for the location and dimensions of spherical and cylindrical vessels depending on the density of the fissile material in solution.[109] For fissile material densities not exceeding 0.5 g/cm^3, the thick water reflector limits may be used, provided a surface separation between the vessel and concrete walls or floor is not less than 0.5 times the vessel diameter. For higher densities, the minimal surface separation should be 0.6 times the diameter.

Chapter IV

MULTIPLE UNITS

A. Neutron Interaction

1. Surface-Density, Density-Analog, and Solid-Angle Methods

The approximate methods for the calculation of interacting fissile units described in this section were conceived before accurate experimental or computational information existed.[112–113] These methods can still serve to separate hypothetical arrays which are clearly subcritical from those that may be supercritical. Models may also serve to identify borderline cases that require investigation by more reliable methods. The solid-angle method, and in some cases the surface-density method, may be particularly useful for estimating the acceptable interaction of vessels within processing plants where information on uniform arrays is inapplicable. The density-analog method was developed to crudely represent regular three-dimensional arrays of fissile units.

Surface-Density[114–116]

This interaction method considers the average surface density of an array of fissile units projected onto an appropriate plane such as a floor or wall. An acceptable value for this surface density is related to the surface density of a subcritical infinite slab of the fissile material by an empirical expression that depends upon the magnitude of an individual unit. For example, Figure 2 of Reference 115 shows such relationships for planar (two-dimensional) arrays of ^{235}U metal spheres and cubes, and of elongated cylinders of uranium solutions at two different ^{235}U densities.

For arrays that are not cubic, the surface density is not unique, but depends upon the plane of projection. The plane giving the maximum value, which is most restrictive, usually will be apparent, e.g., the base of an array of vertical cylinders.

Although there may be cases where application of the surface-density method makes a Monte Carlo calculation unnecessary, the method is by no means universally applicable. There is, for example, no experimental basis for analyzing a planar array of horizontal cylinders such as used for solution storage.

Density-Analog[113, 117–118]

The initial density-analog approach was an attempt to apply to a cubic array a relation similar to that relating the critical mass and density of a single unit. For an isolated unit, the critical mass is proportional to the density to the -2(1-γ) power where γ is zero for an unreflected unit and 0.2 to 0.4 for full water reflection. Early crude subcritical measurements suggested that for arrays, γ could be replaced by f, the "fraction critical" of an isolated unit as defined in the discussion of the surface-density model.

More refined measurements of arrays show that this model is much too conservative for arrays of significant size. As a result, Thomas[114–115] has devised an improved density-analog model that is actually based more nearly on consideration of surface density than overall density. An expression from this version appears in *Nuclear Safety Guide, Revision 2*, but it is not pursued here because tabulations of data from experiments and Monte Carlo calculations are more reliable.

Solid-Angle

This method was developed[112] as a quick, empirical means of evaluating interaction among small numbers of moderated fissile units. It is based on data from experiments with solutions. The technique has been extended in practice to arrays containing large numbers of units. Application of the method to units characterized by a fast neutron spectrum would result in nonconservative spacing if it were not for a required minimum spacing of 0.3 m between units. Thus, guidance for the storage of these units can best be obtained from *American National Standard Guide for Nuclear Criticality Safety in the Storage of Fissile Materials, ANSI/ANS-8.7.*[119]

Application of the solid-angle method may be convenient for special cases such as judging the safety of crowded equipment on a plant floor.

2. Other Methods

Models and methods shown to be consistent with requirements of Standard *ANSI/ANS-8.7* may be used to establish nuclear criticality safety limits. It is emphasized that the concept of the method, its parameter dependence, and its area of applicability must be clearly understood. Appropriately, users should document, for themselves and for others, their ability to apply the method.

A method, well-described and extensively correlated with the results of critical experiments, is Clark's albedo method.[120] Various tables and graphs of parameters have been published[121–122] which facilitate these hand calculations.

Another semi-empirical scheme for evaluating the interaction of fissile units, the interaction parameter method, is reported by Thomas and Scriven of the United Kingdom.[123]

B. Storage of Metal and Compounds

1. Introduction

In addressing the criticality safety of fissile material storage, consideration must be given to the purpose of the storage area. It may be a staging area providing temporary storage for materials in process, it may be an area for transient materials in transport, or it may be an area for long-term storage. Each use presents different problems. The number of units, their mass, surroundings, the necessary accessibility, and the desired margin of subcriticality determine the spacing between units.

Storage specifications of this section are based on descriptions of critical uniform near-cubic arrays that became available after the approximate methods of Section A of this chapter were developed. These descriptions are either experimental,[69, 124–125] from NB_n^2 extrapolations of experimental data, or from validated Monte Carlo calculations.[126–127]

American National Standard Guide for Nuclear Criticality Safety in the Storage of Fissile Materials, ANSI/ANS-8.7, presents subcritical mass limits for spherical units of fissile material assembled in uniform cubic arrays reflected by thick water or its equivalent. The tabulated arrays have a neutron multiplication factor not exceeding 0.95. Although it does not answer all questions, this Standard is directly applicable to many storage problems.

The materials to which the Standard is directly applicable are ^{233}U, uranium containing 30 wt% ^{235}U or more, and plutonium as metals and as dry and wet oxides. The water content of the oxides varies between about 1.4 and 40 wt% (e.g., $0.4 \leq H/U$ or $H/Pu \leq 20$). For each species of fissile material, allowable masses of units are tabulated for cubic arrays of four to ten units on an edge and a range of cubic cell sizes. The limits are also conservatively applicable to units not spherical in shape and to arrays that are not cubic. It should be clear that cubic arrays for which data are tabulated in the Standard and the arrays in this section do not represent most practical storage arrangements because of need for access to interior units.

The Standard does not provide for the introduction of hydrogenous material into the space between units. If such moderation is present, the effect must be evaluated by a validated computational technique. The effect on array reactivity due to the introduction of water, as for example from fire protection systems, is strongly dependent on the form of the fissile material, and on the mass and spacing of the units. There is, however, an adequate margin in the limits to accommodate incidental moderation such as would result from enclosing the units in plastic bags that introduce no more than 10 g of polyethylene per kilogram of fissile material. As Reference 128 shows, there is extreme sensitivity to hydrogenous moderation between units, which becomes even greater if the density of units is decreased. Because this effect can easily override the margin $\Delta k_{eff} = 0.05$, interstitial moderation would become an important contingency.

Guidance for increasing the subcritical margin of an array of Standard *ANSI/ANS-8.7* beyond the existing $\Delta k_{eff} = 0.05$ may be obtained from Figure 3 of Reference 127. This figure gives values of k_{eff} for an extensive range of subcritical reflected cubic arrays of spherical units. All data in the range of k_{eff} above about 0.75 satisfy the relation $k_{eff} = r/r_c$, where r is the radius of a unit in the subcritical array and r_c is the radius that would make the array critical. As an example, a change of $\Delta k_{eff} = 0.1$ below $k_{eff} = 0.95$ corresponds to a decrease of r/r_c from 0.95 to 0.85. This change in radius is equivalent to a 28% decrease in mass.

The Standard provides factors for reducing the mass limits for arrays closely reflected by concrete. The limits are reduced to 75% of their tabulated values if the concrete thickness is between 5 and 8 inches and to 60% for greater thicknesses. Criteria are presented for pairs of arrays in concrete enclosures. (Slight neutron coupling of arrays separated by 50-cm-thick concrete has been observed experimentally.)[129] Each unit of an array must remain subcritical if immersed in water. The possibility of double batching of the units in a storage cell should be considered when establishing safety limits and operating procedures. Administrative controls, appropriately-sized containers, and storage cell design may be useful for the prevention of double batching.

Consideration should be given to other normal and credible abnormal storage conditions that may affect the margin by which the array is subcritical. Typical examples[4, 23] of changes in operating conditions that should be considered are

- flooding, spraying, or the presence of water, oil, snow (i.e., low-density water), cardboard, wood, or other moderating materials;
- the introduction of additional units or reflectors;
- improper placement of units;
- loss of moderator and neutron absorber between units;
- collapse of a framework used to space units;
- a change in the density of fissile material during storage;
- the substitution of units containing more fissile material than permitted in operations as a result of operational error or improper labeling.

The Standard associates each unit with a cubic (or near-cubic) cell, but does not specify the means of establishing the cell. Although the cell can be visualized as an imaginary cuboidal volume, in practice it needs to be defined by hardware. For example, cells may be subdivisions of sturdy shelving, be maintained by compartments, or be defined by appropriately sized containers. Alternatively, the cell may be determined by a "birdcage" consisting, typically, of a container centered in a cell-size framework constructed of tubing or angle iron. Although the birdcage may be an open structure, it must be capable of

assuring separation of the containers. Birdcages may be on shelving or simply distributed or stacked on a floor. An alternative to shelving, suitable for long-term storage, may be an array of tubes extending through a concrete matrix as reported from the Y-12 Plant.[130] As a technical practice, the Standard states "Storage of fissile materials shall be such as to obviate concern with accidental nuclear criticality in event of fire, flood, earthquake or other natural calamities."

2. Commingling of Dissimilar Units

The Standard allows for commingling of dissimilar units in an array under the following conditions. If cell size is maintained by a container or birdcage about each unit, a criticality indicator CI may be assigned to each container or birdcage with its unit such that CI = 100/N, where N is the number of cells in an appropriate allowable array.[131] Then, dissimilar units in containers may be commingled, provided the summation of CIs of all cells within the resultant array does not exceed 100.

3. Alternative Representation of Storage Arrays

Figures 19 through 23 are graphic representations of selected tables in Standard *ANSI/ANS-8.7* for enriched uranium and plutonium. Graphs for ^{233}U are not included because of the generally small available quantity of this material. These figures simplify the interpolation of allowable numbers of units to non-cubic three-dimensional arrays and to cell sizes other than those tabulated.

It is considered more useful to give minimum cell dimensions in inches instead of millimeters. Further, equivalent cell volumes in US gallons are indicated in Figure 24. These include capacities of steel drums that may be used as containers of units. Often such drums are outer containers of units packaged for transportation. The graphs facilitate establishing values of CI for units in drums.

It must be emphasized that the conditions which pertain to Standard *ANSI/ANS-8.7* apply to Figures 19 to 23. Specifically, the arrays are reflected, and no significant amount of interstitial moderating material is present.

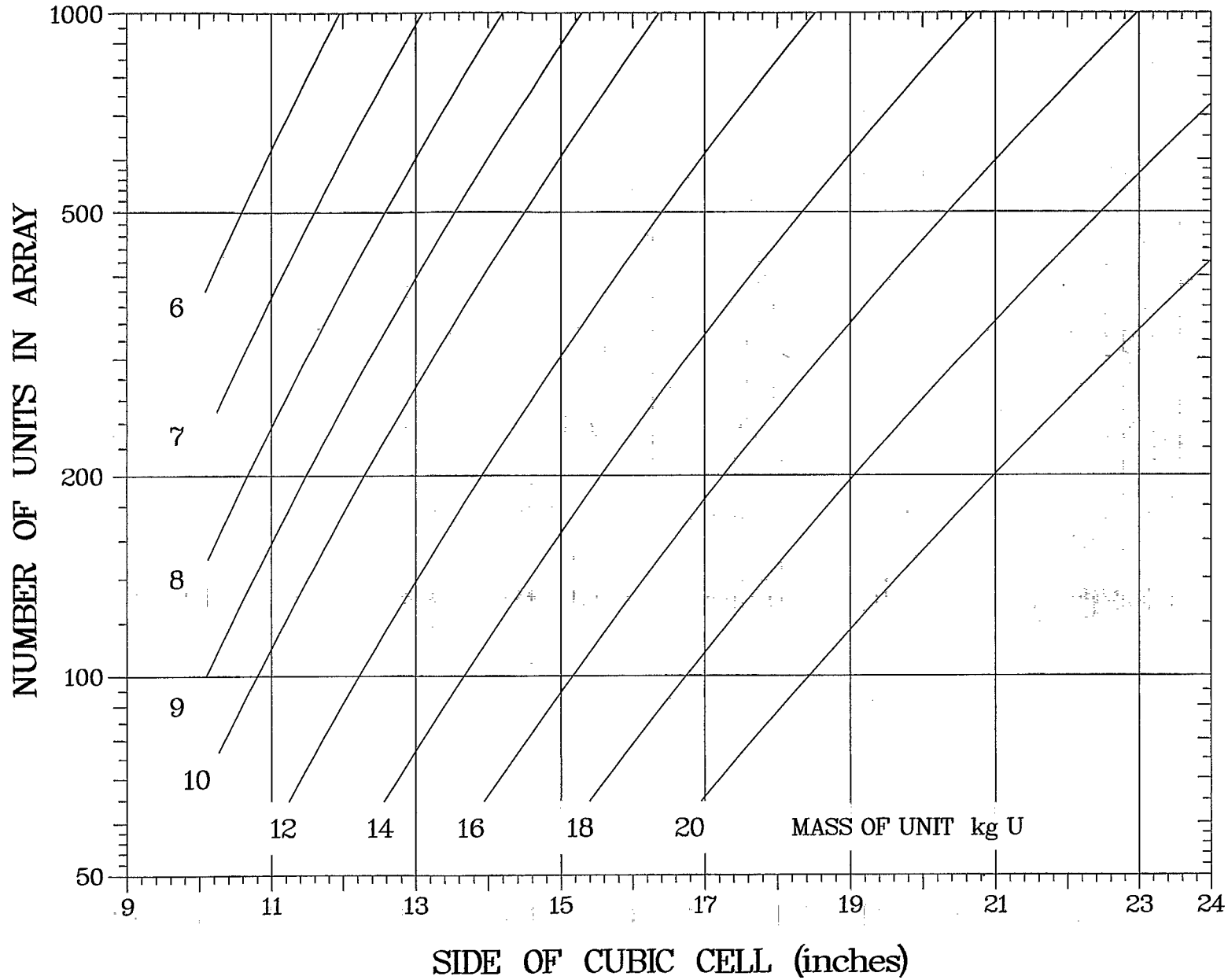

Figure 19

Fig. 19. Subcritical reflected arrays of spherical units of U(93.2) metal.

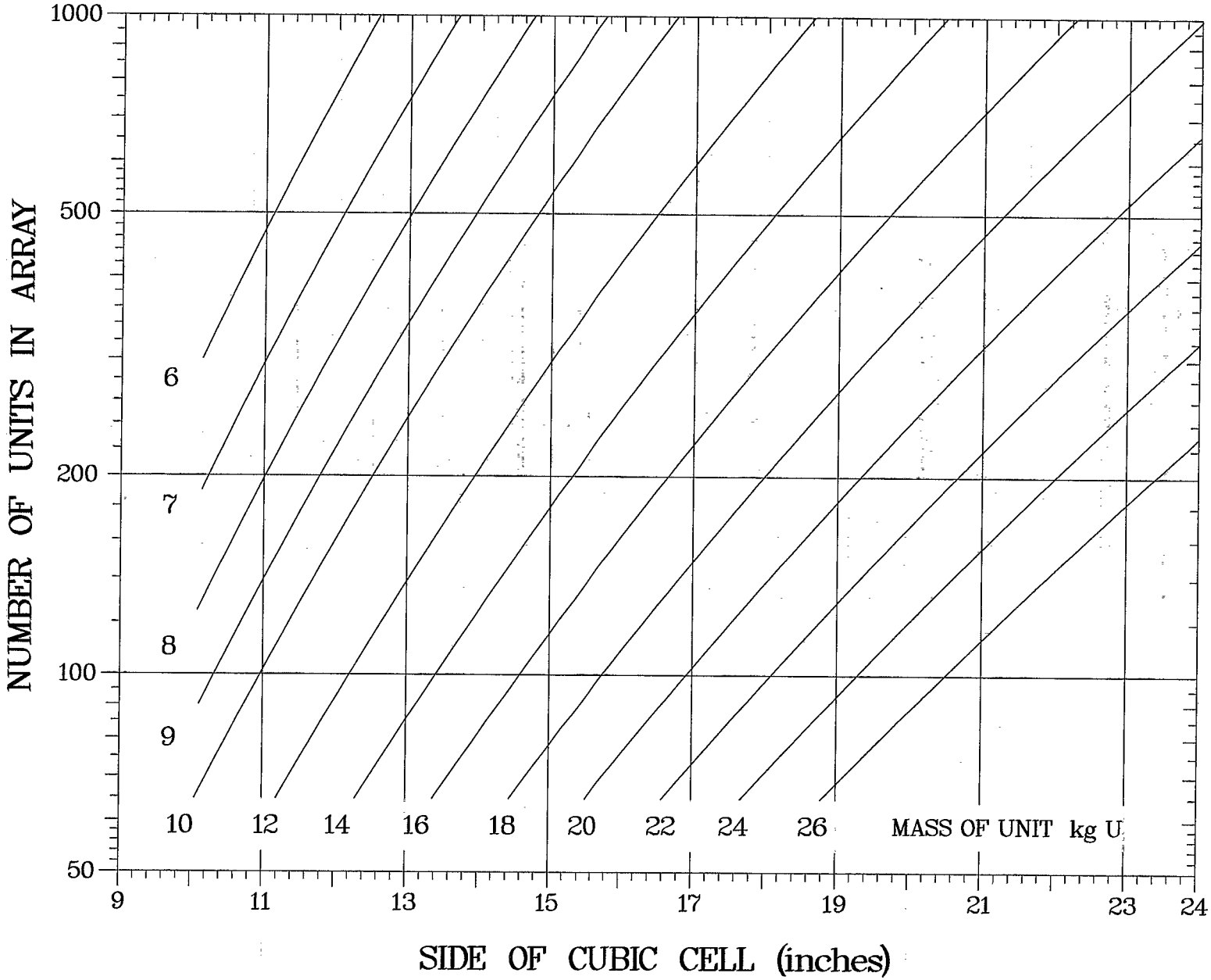

Figure 20

Fig. 20. Subcritical reflected arrays of spherical units of U(93.2) oxide at H/U $\leq$ 0.40.

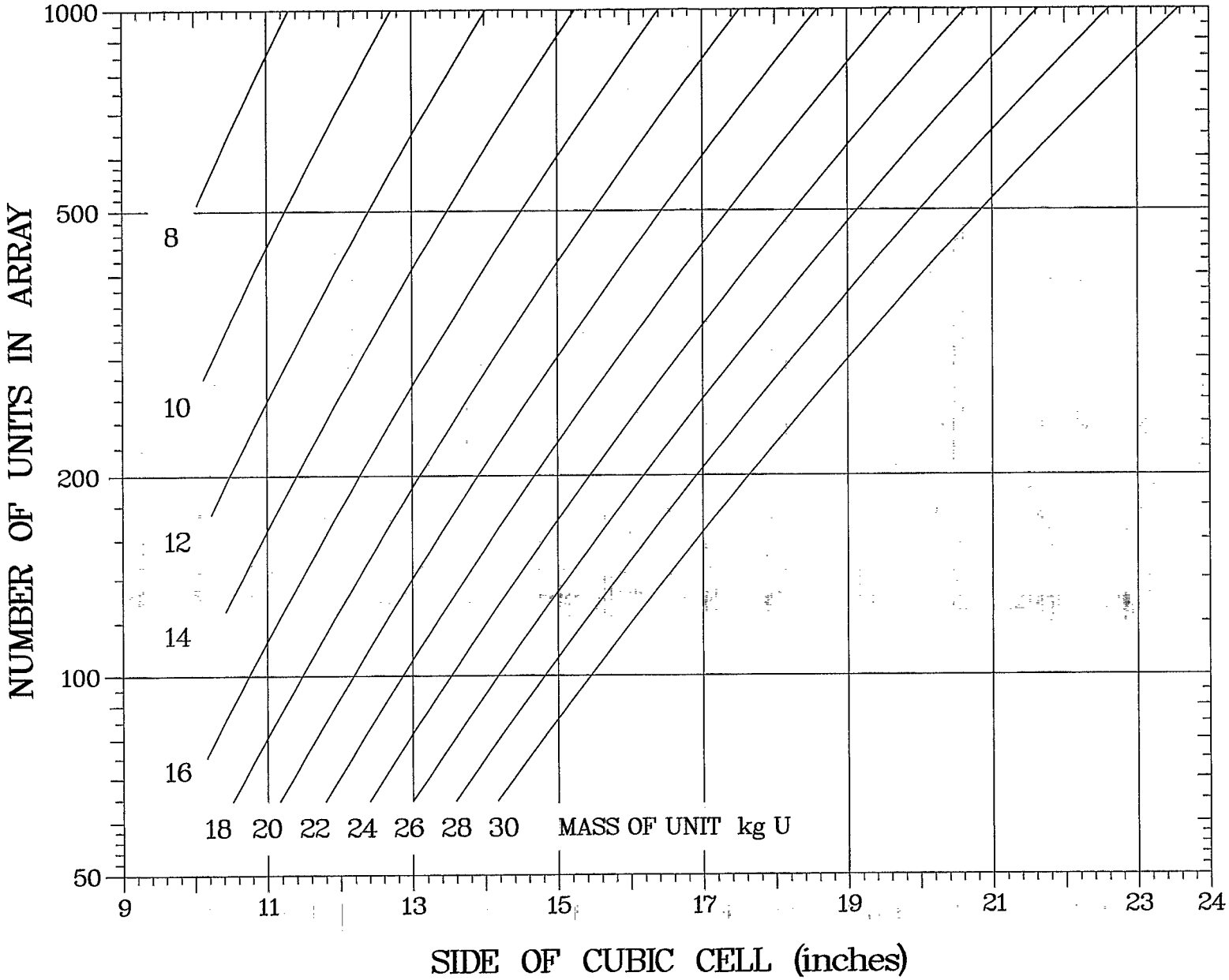

Figure 21

Fig. 21. Subcritical reflected arrays of spherical units of U(50) oxide at H/U $\leq$ 0.40.

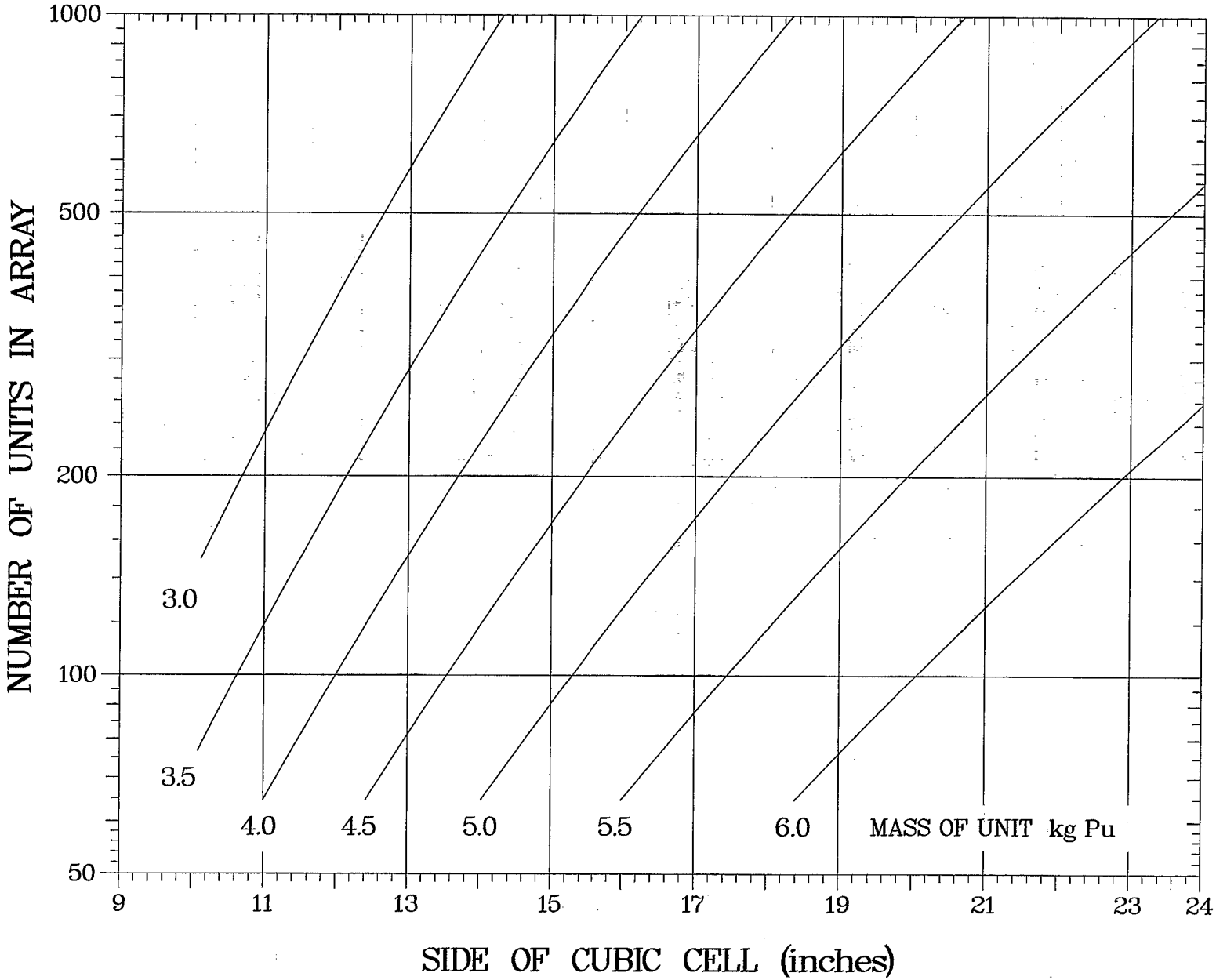

Figure 22

Fig. 22. Subcritical reflected arrays of spherical units of Pu(94.8) metal.

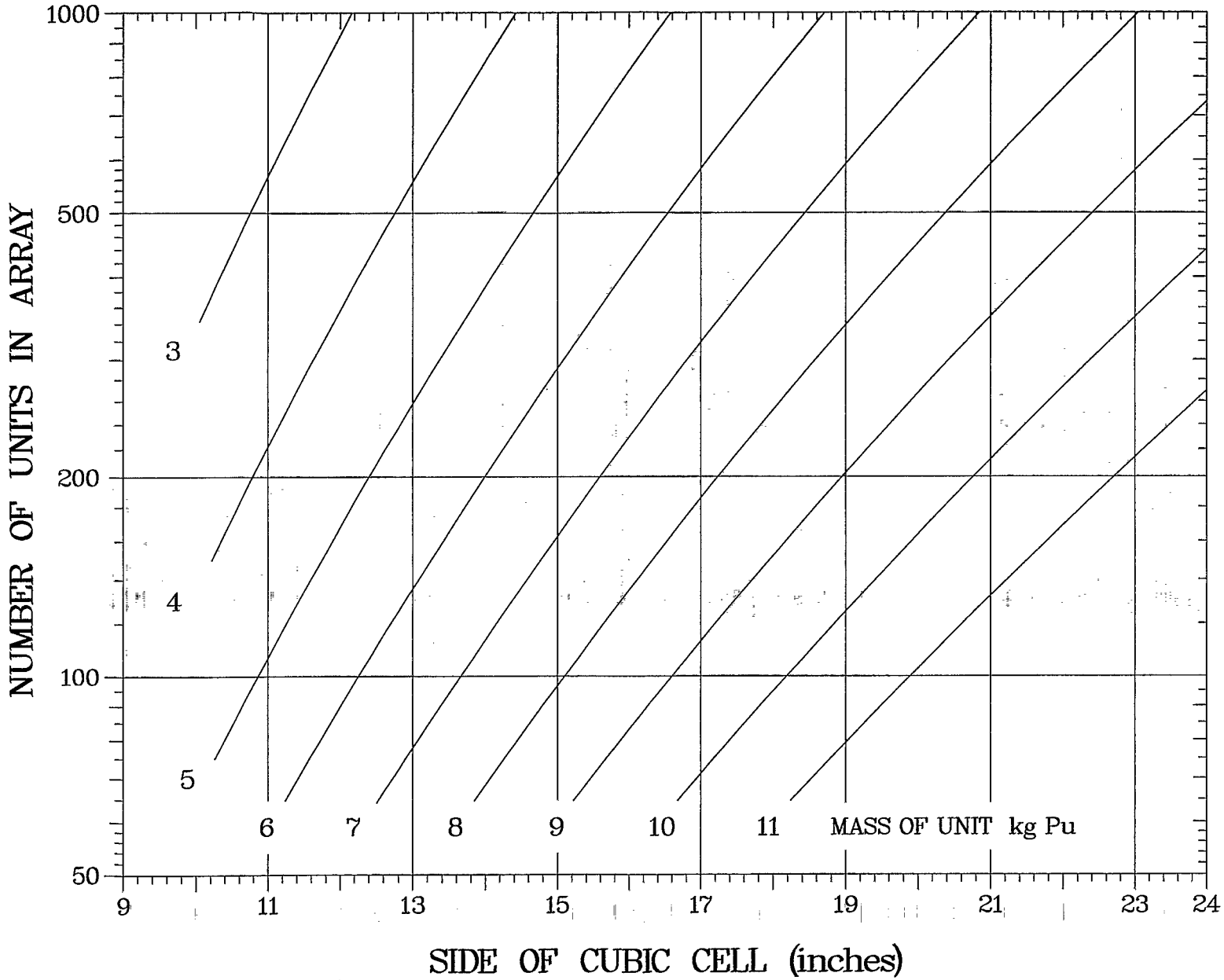

Figure 23

Fig. 23. Subcritical reflected arrays of spherical units of Pu(94.8) oxide at H/Pu $\leq$ 0.40.

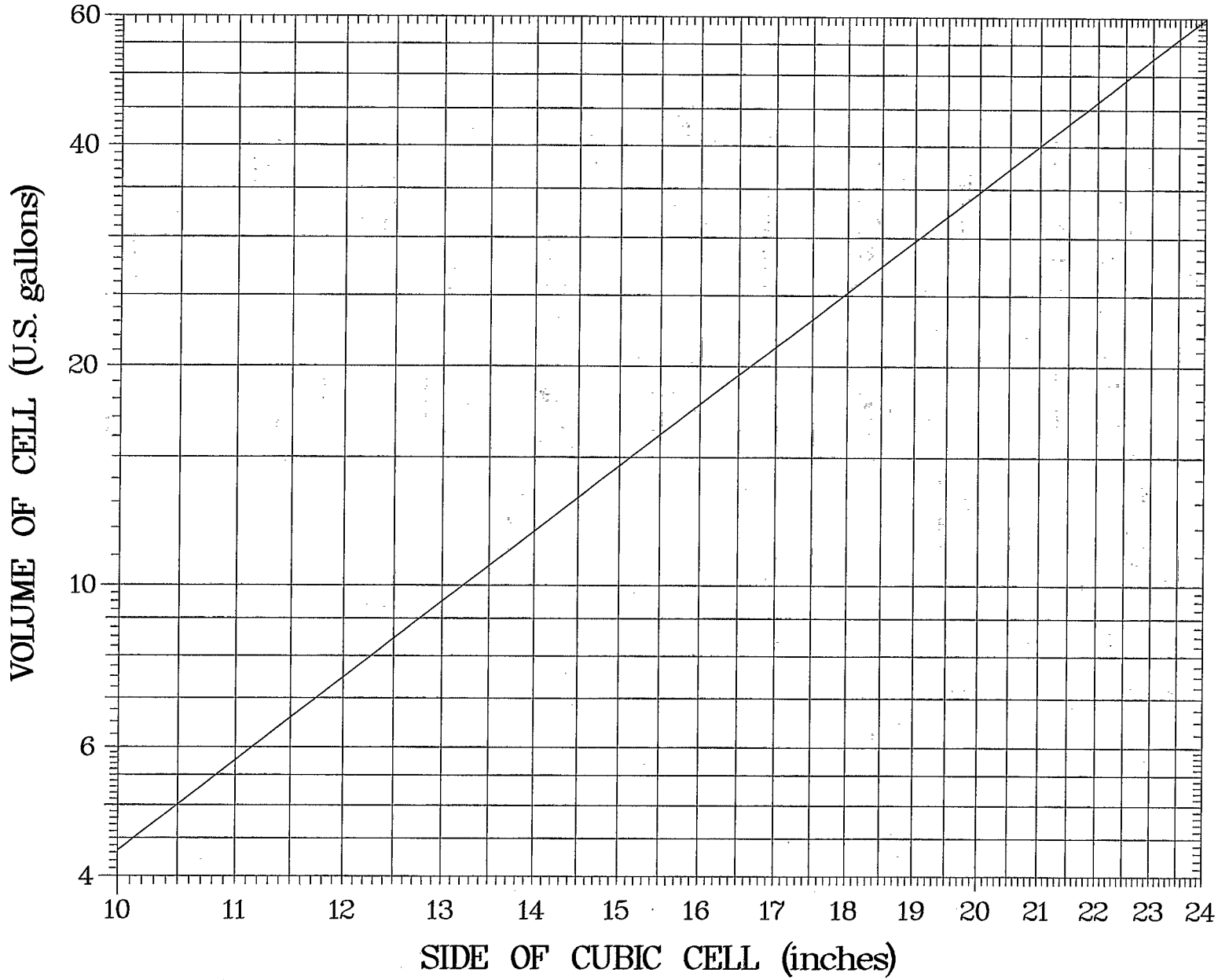

Figure 24

Fig. 24. Relationship between cell volume and edge length of cubic cell.

C. Storage of Solutions

1. Tanks and Bottles

Experimental data on critical near-cubic arrays of five-liter ^{235}U solution units[126] suggest that it might be possible to include storage arrays of those units in the format of Standard *ANSI/ANS-8.7*. Such an approach would not be of much value, however, because there are more practical ways to store appreciable volumes of solution than in small containers.* The absence of a general standard on solution storage may be, in part, because the choice among tanks with Raschig rings, banks of pipe or tubing, annular tanks, slab tanks, or even elongated polyethylene bottles depends on which fits best into the operational and physical features of each plant. Regardless, general solution-storage criteria are not available. Each of the available options will be considered, with the background experimental information, and something about adaptation to plant conditions. There is no study that uses a consistent set of criteria to evaluate the relative costs of these types of storage. Cost of solution storage can be quite high.

Tanks with Raschig Rings[31]

Storage of solutions in large tanks containing Raschig rings has the attractive feature of minimizing floor area. Offsetting this, however, are time-consuming and expensive inspections and tests called for in the Standard *ANSI/ANS-8.5* as a result of the large surface area in contact with solution, which invites deposits, and the possibility of damage to the rings.[134] Consequently tanks packed with Raschig rings are now used more commonly in auxiliary vessels where solution can be introduced only by accident than for actual storage.

The Raschig ring Standard[31] is supported by critical experiments at Oak Ridge with $U(93)O_2(NO_3)_2$ solution[135] and $^{233}UO_2(NO_3)_2$ solution.[136] Experiments at Hanford included solutions of plutonium nitrate[137] and mixtures of plutonium and uranium nitrate.[138] A problem with computation is the difficulty of realistically modeling the random array of Raschig rings with Monte Carlo techniques. The possible use of vessels packed with Raschig rings for storing solutions is discussed further in Chapter V.

*Because of the limited availability of ^{233}U solution, storage in small containers may be practical. Guiding experimental data are available for critical arrays of 4.3-liter[132] and 3.0-liter units of ^{233}U solution.[133]

Elongated Polyethylene Bottles

The largest critical array of uranyl nitrate solution units, reported from Oak Ridge,[139] is so impressive that its photograph has appeared widely, for example on p. 137 of Reference 11 and p. 607 of Reference 140. The solution is contained in 98 polyethylene bottles, each 13.7-cm-o.d. and of about 13-liter capacity. The wall thickness varies from 0.51 cm at the top to 1.14 cm near the base and the inside height is about 122 cm.

These containers, designated "Type A" in the Oak Ridge report, have been used in several plants for transferring solution from one location to another and for at least temporary storage. A cart for transfer holds the container upright and spaces it from other objects. A fixture secures the container at an appropriate storage location, properly separated from other objects. This arrangement may be useful for regular uranium solution storage if the total inventory does not exceed 100 L or so.

Some type A containers have embrittled after a couple years of service and in one instance are being replaced by smaller cylinders with a more resistant polyethylene composition.

Slab Tanks

Thin slab tanks have been used to store solutions of both plutonium and enriched uranium. They have been used for both plutonium and enriched uranium solutions. Welded spacers prevent thickness increase as a result of hydrostatic pressure or relaxation of walls. Slab tanks have not been used in plants for large-scale storage partially because of the moderator-absorber between parallel tanks that would be required to reduce interaction in an array.

Annular Cylinders

Although experiments with cylindrical annuli[92–93, 96] were conceived as contributing to solution storage, this type of container has not been used for large-volume storage. At the Valduc facility in France, solution for CRAC experiments was stored in annular containers.[94]

2. Solution Storage using Pipe or Tubing

Parallel lengths of 6-inch pipe or tubing are used for storing very large volumes of fissile solutions at the Oak Ridge Y-12 Plant and at Los Alamos. Principally because of sensitivity to surroundings, generally applicable criteria for such storage do not exist. Therefore, each new application should be examined in detail by a validated method.

Apart from solution properties, significant considerations include spacing of units, distance from concrete walls and other massive or fissionable objects, provisions for handling solution leakage, and the possibility of water flooding. It should be noted that effects of even small depths of water can be significant. At neither Y-12 nor Los Alamos is flooding a contingency, although the influence of low-density water from fire-protection sprinklers is considered.

With solutions, control of fissile-material density and stability are important. The possibility and consequences of precipitation and settling must be considered.

Effective procedures for transfer of solution to and from storage arrays must be established.

Y-12 Practice

At the Y-12 Plant, solution volumes are thousands of liters even though practice is to concentrate dilute solutions. The practical upper limit of uranium density is 450 g/liter, and solutes are uranyl nitrate. Containers are lengths (up to 40 feet) of 6-inch stainless-steel pipe, generally Schedule-40 or greater to allow for corrosion. 6-inch stainless-steel tubing, presently with one-quarter inch wall, is substituted in some cases. Pipes of smaller standard size are ruled out because of the large numbers that would be required.

In storage arrays, the minimum axial spacing of containers is 24 inches, and separation of axes from wall or floor is at least one-half of this. Each array consists of containers in a single planar configuration. Most arrays have horizontal containers, which are awkward to clean thoroughly. To simplify sparging, plans call for replacing all horizontal containers with vertical containers. There is allowance for the effect of sprinkler water having a density of 0.015 g/cm^3 between units, which is three times the expected value.

The floor of each storage room is lined with a stainless steel pan to accommodate solution in a subcritical slab in the event of severe leakage.

These conditions prove to be conservative throughout the Y-12 Plant, which might not be the case for an array consisting of more than one plane of containers, if water-flooding were a contingency, or if extremely concentrated solutions or mixtures were permitted.

Los Alamos Practice

Los Alamos has been required to accommodate hundreds of liters of plutonium solution in a facility designed originally for research in which solution storage was expected to be minimal. As at Y-12, 6-inch pipe is the practical size for storage containers, but space limitations call for 10-foot lengths in more compact arrays than those at Y-12. This compact configuration is made possible by a stringent limitation on plutonium density.

Supported by Monte Carlo calculations,[141] solutions with plutonium density not to exceed 20 g/liter are stored in 6-inch Schedule-10 pipes with minimal axial spacings of 18 inches. Smaller spacings with intervening fixed neutron absorbers are used for dilute waste solutions held to determine whether further plutonium recovery is required.

As planned at Y-12, horizontal tanks are being replaced by vertical tanks for ease of cleaning. Deposits as a result of precipitation are more often encountered with plutonium solutions than with uranium.

Plutonium contamination is controlled by plastic wrappings of potential leakage points. This approach eliminates the need for the floor catch pans such as those in use at Y-12.

Comments

These examples show what can be done for large-volume solution storage when validated Monte Carlo calculations are available for guidance and confirmation. With smaller volumes, for which containers of diameter no greater than 5 inches are practical, generous spacing of vessels, or a readily observed limit on density of fissile material, may reduce or eliminate the need for Monte Carlo confirmation of safe conditions. Administrative controls on solution transfer, handling of leakage or spillage, and material accountability should contribute reassuringly to criticality safety. The possibility of precipitation and settling or deposit on surfaces would still be a consideration.

D. Transportation

Transport of fissile material is addressed by regulations of the International Atomic Energy Agency (IAEA),[142–143] the US Department of Transportation,[144] the Nuclear Regulatory Commission (NRC),[145] and the Department of Energy (DOE).[146]

Requirements of transport regulation for criticality safety necessarily go beyond experimental data and computational results. Some requirements result from consensus among national and international authorities on judgmental matters. These matters include interaction of shipments, effects of accidents on packages, and design of specification containers. Approved US practice, also a matter of judgment, is even more limited than allowed by regulation, e.g., the general avoidance of shipment by common carrier. Since the requirements for transport of fissile material are more legalistic than technical, discussion of these requirements is beyond the scope of this document.