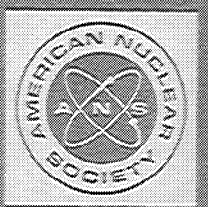
REFERENCE 110

"NUCLEAR CRITICALITY CONTROL AND SAFETY OF PLUTONIUM-URANIUM FUEL MIXTURES OUTSIDE REACTORS," ANSI/ANS-8.12-1987 (AMERICAN NATIONAL STANDARDS INSTITUTE, INC., NEW YORK, 1987).

American Nuclear Society

nuclear criticality control and safety of plutonium-uranium fuel mixtures outside reactors

an American National Standard



published by the
American Nuclear Society
555 North Kensington Avenue
La Grange Park, Illinois 60525 USA

ANSI/ANS-8.12-1987

ANSI/ANS-8.12-1987
Revision of
ANSI/ANS-8.12-1978

American National Standard for Nuclear Criticality Control and Safety of Plutonium-Uranium Fuel Mixtures Outside Reactors

Secretariat
American Nuclear Society

Prepared by the
American Nuclear Society
Standards Committee
Working Group ANS-8.12

Published by the
American Nuclear Society
555 North Kensington Avenue
La Grange Park, Illinois 60525 USA

Approved September 11, 1987
by the
American National Standards Institute, Inc.

American National Standard

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Published by

American Nuclear Society
555 North Kensington Avenue, La Grange Park, Illinois 60525 USA

Printed in the United States of America

Foreword

(This Foreword is not a part of American National Standard for Nuclear Criticality Control and Safety of Plutonium-Uranium Fuel Mixtures Outside Reactors, ANSI/ANS-8.12-1987.)

This standard provides guidance for the prevention of criticality accidents in the handling, storing, processing, and transporting of plutonium-uranium fuel mixtures outside reactors and is applicable to all operations involving mixtures of plutonium and natural uranium. It constitutes an extension of the American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors, ANSI/ANS-8.1-1983.

Under the prescribed five year review of ANSI/ANS-8.12-1978, the standard has been revised to include subcritical limits for heterogeneous lattices of mixed oxide fuel pins in water. The basis for the limits for both homogeneous mixtures and for lattices are calculations done by several members of the work group, which have been published in the open literature. These calculations were done by methods that have been validated by correlations with available experimental data, and an adequate margin of subcriticality was allowed. The revised standard was prepared by Work Group ANS-8.12.1 of Subcommittee 8 of the Standards Committee of the American Nuclear Society. This work group was composed of:

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R. Artigas, *General Electric Company*
C. L. Brown, *Rockwell Hanford Operations*
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Nuclear Criticality Control and Safety of Plutonium-Uranium Fuel Mixtures Outside Reactors

1. Introduction

The American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors, ANSI/ANS-8.1-1983 [1][1] provides single parameter limits for fissionable units of simple shape containing ^{233}U, ^{235}U, and ^{239}Pu [1]. As an example of multi-parameter control it provides an increase in the limits of ^{235}U resulting from control of the amount of ^{238}U associated with ^{235}U in uranium metal and oxide enriched to no more than 5 wt% ^{235}U. Larger limits for plutonium likewise result from placing reliance on, and hence controlling, the isotopic concentraion of ^{240}Pu and the amount of uranium associated with the plutonium. Such increases may prove valuable for operations with mixed oxides of plutonium and uranium encountered in light water, liquid metal fast breeder, and gas-cooled fast reactor fuel cycle operations.

The limits provided here were calculated (E. D. Clayton [2, 3]) by methods satisfying the requirements for validation of a calculational method as set forth in 4.3 of ANSI/ANS-8.1-1983 [1].

The administrative and technical practices for criticality safety and control as embodied in ANSI/ANS-8.1-1983 [1] and in American National Standard Administrative Practices for Nuclear Criticality Safety, ANSI/ANS-8.19-1984 [4] are applicable herein.

2. Scope

This standard is applicable to operations with plutonium-uranium oxide fuel mixtures outside nuclear reactors, except the assembly of these materials under controlled conditions, such as in critical experiments. Basic criteria are presented for plutonium-uranium fuel mixtures in single units of simple shape containing no more than 30 wt% plutonium combined with uranium containing no more than 0.71 wt% ^{235}U. The limits for uniform aqueous mixtures (solution) are applicable to homogeneous mixtures and slurries in which the particles constituting the mixture are uniformly distributed and have a diameter no larger than 127 μm (0.005 in.), i.e., are capable of being passed through 120 mesh screen.[2]

This standard does not include the details of administrative controls, the design of processes or equipment, the description of instrumentation for process control or detailed criteria to be met in transporting fissionable materials.

3. Definitions

3.1 Limitations. The definitions given below are of a restricted nature for the purpose of this standard. Other specialized terms are defined in H. Alter, et al., "Glossary of Terms in Nuclear Science and Technology" [5].

3.2 Shall, Should, and May. The word "shall" is used to denote a requirement, the word "should" to denote a recommendation, and the word "may" to denote permission, neither a requirement nor a recommendation. In order to conform with this standard all operations shall be performed in accordance with its requirements but not necessarily with its recommendations.

[1]Numbers in brackets refer to corresponding numbers in Section 7, References.

[2]Mixtures containing particles 60-100 μm in size have been considered by experimenters to be homogeneous [V. I. NEELEY and H. E. HANDLER, "Measurement of Multiplication Constant for Slightly Enriched Homogeneous UO_3–Water Mixtures and Minimum Enrichment for Criticality," HW-70310 Hanford Atomic Products Operations (August 1961) and V. I. NEELEY, J. A. BERBERET and R. H. MASTERSON, "k∞ of Three Weight Per Cent ^{235}U Enriched UO_3 and $UO_2(NO_3)_2$ Hydrogeneous Systems," HW-66882, Hanford Atomic Products Operations (September 1961)].

3.3 Glossary of Terms.

areal density. The total mass of fissionable material per unit area projected perpendicularly onto a plane. (For an infinite, uniform slab, it is the product of the slab thickness and the concentration of fissionable material within the slab.)

controlled parameter. A parameter that is kept within specified limits.

criticality accident. The release of energy as a result of accidentally producing a self-sustaining or divergent neutron chain reaction.

natural uranium. Reference throughout this standard to natural uranium shall be interpreted to mean uranium in which the concentration of the ^{235}U isotope is equal to or less than 0.71 wt%.

nuclear criticality safety. Protection against the consequences of an inadvertent nuclear chain reaction, preferably by prevention of the reaction.

subcritical limit (limit). The limiting value assigned to a controlled parameter that results in a subcritical system under specified conditions. The subcritical limit allows for uncertainties in the calculations and experimental data used in its derivation but not for contingencies; e.g., double batching or inaccuracies in analytical determinations.

4. Nuclear Criticality Safety Practices

4.1 Administrative and Technical Practices. Operations within the scope of this standard shall be conducted in accordance with ANSI/ANS-8.1-1983 [1] except that the limits for plutonium-uranium mixtures given herein may be utilized. Attention shall be given to credible abnormal conditions such as those listed in the Appendix A of ANSI/ANS-8.1-1983 [1].

4.2 Homogeneity Control. Consideration shall be given to the possibility of preferential separation of plutonium from uranium. Neither the limits for homogeneous systems nor those for heterogeneous systems apply unless the plutonium and uranium oxides remain intimately and homogeneously mixed.

5. Subcritical Limits for Uniform, Homogeneous Plutonium-Uranium Mixtures in Water

Operations with the fuel mixtures specified may be performed safely by complying with any one of the subcritical limits given in Tables 1, 2, or 3, provided the conditions under which it applies are maintained.

A limit shall be applied only when the effect of neutron reflectors and other nearby fissionable materials is no greater than that of a contiguous water reflector of effectively infinite thickness.

Process specifications shall incorporate margins to protect against uncertainties in process variables and against a limit being accidentally exceeded.

5.1 Homogeneous Aqueous Mixtures. The limits of Table 1 are applicable to homogeneous mixtures of oxides of natural uranium and plutonium provided the plutonium oxide content of the mixture of oxides is between 3 wt% and 30 wt%. Limits are provided for three isotopic compositions of plutonium. In applying these limits, any ^{238}Pu and ^{242}Pu present shall be omitted in computing the isotopic composition. All limits are valid for uranium containing no more than 0.71 wt% ^{235}U with ^{239}Pu.

5.2 Variation of Subcritical Limits with PuO_2 Content. The limits of Table 1 are appropriate for criticality prevention. For greater flexibility, plots of the subcritical limits of Table 1 are provided in Figs. 1 through 5, from which values appropriate to any PuO_2 content in the range 3 to 30 wt% may be read. These figures give, respectively, limits on the total mass of the oxides ($PuO_2 + UO_2$), on volume, on cylinder diameter, on slab thickness, and on areal density.

5.3 Dry and Damp Mixed-Oxide Powders. The limits given in Table 2 apply to dry and damp mixtures of the oxides of plutonium and natural uranium provided the concentration of ^{240}Pu exceeds that of ^{241}Pu. Because completely dry oxide may be difficult to ensure, limits are provided for damp oxides in which $0 < H:(Pu + U) \leq 0.45$ [1.48 wt% water corresponds to an $H:(Pu + U) \simeq 0.45$]. Limits are also provided for oxide powders with 50% voids.

5.4 Limiting Concentrations of Plutonium in Unlimited Quantities of Mixtures of the Oxides and Nitrates of Plutonium and Natural Uranium. The maximum safe concentrations of plutonium in (PuO_2 + UO_2) or in [$Pu(NO_3)_4$ + $UO_2(NO_3)_2$] in unlimited quantity under the stated conditions are given in Table 3. In each case the value of the infinite multiplication factor, $k\infty$ is less than unity for these conditions and is independent of the density of the mixture. For example, criticality cannot be achieved in a homogeneous mixture of (PuO_2 + UO_2) in water if the plutonium content is $\leq$ 0.13 wt% of the combined mass of the (Pu +U) in the oxides. These limits are not applicable to mixtures of elemental plutonium and uranium in water.

6. Subcritical Limits for Mixed-Oxide Heterogeneous Systems.

Lumping the mixed-oxide fuel produces an effect on criticality similar to that which occurs in natural or moderately enriched uranium. The total absorption in ^{238}U and ^{240}Pu resonances in the mixed oxide will be minimized if the mixed oxide is separated from the moderator, e.g., in a lattice assembly of fuel rods in water, as compared to an aqueous homogeneous mixture of the oxides. Because of the decreased absorption of the ^{240}Pu as a result of lumping, the effect of ^{240}Pu on the subcritical limit also can be expected to be smaller than in the case of aqueous homogeneous mixtures.

Subcritical limits are specified in Fig. 6 through 9 as a function of both the PuO_2 content in the mixed oxides and the ^{240}Pu content of the plutonium. In applying these limits, any ^{238}Pu and ^{242}Pu present shall be excluded when computing isotopic composition. The limits of these figures are applicable to heterogeneous systems regardless of the size or shape of the mixed oxide pieces or degree of moderation by water.

CAUTION: Curves III of Fig. 7 through 9, which apply to $^{240}Pu \geq 25$ wt% and $^{241}Pu \leq 15$ wt%, shall not be extended by extrapolation beyond the 18 wt% PuO_2 limit shown. Curve II may be used above 18 wt% for higher PuO_2 concentrations. Alternatively, the subcritical limits may be calculated and separately validated or determined by measurement.

Process specifications shall incorporate margins to protect against uncertainties in process variables and against a limit being accidentally exceeded.

7. References

[1] American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors, ANSI/ANS-8.1-1983. American Nuclear Society, LaGrange Park, IL.

[2] E. D. CLAYTON, et. al., "Basis for Subcritical Limits in Proposed Criticality Safety Standard for Mixed Oxides," *Nucl. Tech. 35,* 97, 111 (1977).

[3] E. D. CLAYTON, et. al., "Basis for Extending Limits in ANSI Standard for Mixed Oxides to Heterogeneous System," *Nucl. Tech.* (1986).

[4] American National Standard Administrative Practices for Nuclear Criticality Safety ANSI/ANS-8.19-1984. American Nuclear Society, LaGrange Park, IL.

[5] H. ALTER, et. al., Glossary of Terms in Nuclear Science and Technology. American Nuclear Society, LaGrange Park, IL.

Only the standards explicitly referred to in this document qualify as references. Subsequent revisions of these standards shall not be substituted.

Table 1
Subcritical Limits for Uniform Aqueous Mixtures of the Oxides of Plutonium and Natural Uranium

All values are upper limits except atomic ratios which are lower limits

PuO_2 in (PuO_2 + UO_2), wt%	3			8			15			30[b]		
Plutonium Isotopic Composition[a]	I	II	III	I	II	III	I	II	III	I	II	III
Mass of plutonium in oxide mixture, kg	0.73	1.35	2.00	0.61	1.06	1.53	0.54	0.94	1.28	0.50	0.87	1.16
Mass of (PuO_2 + UO_2), kg	27.5	51.3	75.9	8.6	15.1	21.7	4.1	7.1	9.7	1.9	3.3	4.4
Diameter of infinite cylinder, cm	24.3	30.8	34.8	19.8	24.9	27.5	17.8	22.5	24.8	16.2	21.0	23.4
Thickness of infinite slab, cm	11.0	14.9	17.4	8.2	11.2	12.9	6.9	9.6	11.0	5.9	8.7	9.9
Volume of oxide mixture, liter	23.5	44.8	63.4	14.0	25.9	34.4	11.0	20.4	26.6	8.5	16.8	21.6
Concentration of plutonium, g Pu/liter	6.8[c]	8.1	9.3	6.9	8.2	9.4	7.0	8.2	9.4	7.0	8.1	9.3
Concentration of oxides, g(PuO_2 + UO_2)/liter	257[c]	305	351	97.3	116	134	52.9	61.7	71.0	26.5	30.7	35.2
H:Pu atomic ratio	3780	3203	2780	3780	3210	2790	3780	3237	2818	3780	3253	2848
Areal density of plutonium, g Pu/cm^2	0.27	0.38	0.47	0.25	0.34	0.42	0.25	0.33	0.41	0.24	0.32	0.37
Areal density of oxides, g(PuO_2 + UO_2)/cm^2	10.2	14.4	17.7	3.5	4.8	5.9	1.9	2.5	3.1	0.9	1.2	1.4

a. Plutonium isotopic composition:
   I — $^{240}Pu > {}^{241}Pu$
   II — $^{240}Pu \geq 15$ wt% and $^{241}Pu \leq 6$ wt%
   III — $^{240}Pu \geq 25$ wt% and $^{241}Pu \leq 15$ wt%

b. Dimensional and volume limits do not apply for isotopic compositions II and III unless, for II, the concentration of oxides is less than 5700 g/liter and, for III, less than 4500 g/liter.

c. This concentration limit is not applicable to oxide mixtures in which the $PuO_2/(PuO_2 + UO_2)$ ratio is less than 3 wt% because of the increased relative importance of ^{235}U in high-uranium-bearing materials. Limited guidance for material of less than 3 wt% PuO_2 may be found in Table 3.

Table 2
Subcritical Mass Limits for Single Units of Homogeneously Mixed Oxides of Plutonium and Natural Uranium at Low Moderation

The limits apply to combinations of plutonium isotopes provided $^{240}Pu > {}^{241}Pu$.

PuO_2 in (PuO_2 + UO_2), wt%	3		8		15		30	
	Mass of Plutonium, kg	Mass of (PuO_2+UO_2), kg	Mass of Plutonium, kg	Mass of (PuO_2+UO_2), kg	Mass of Plutonium, kg	Mass of (PuO_2+UO_2), kg	Mass of Plutonium, kg	Mass of (PuO_2+UO_2), kg
Dry mixed oxides at theoretical density Density of (PuO_2 + UO_2) ≤ 11 g/cm³ H:(Pu + U) = 0	Subcritical in any amount		122	1729	47.0	355	26.1	98.6
Damp mixed oxides at theoretical density Density of (PuO_2 + UO_2) ≤ 9.4 g/cm³ 0 < H:(Pu + U) ≤ 0.45	236	8919	49.4	700	32.9	249	23.3	88.1
Damp mixed oxides with 50% voids Density of (PuO_2 + UO_2) ≤ 4.7 g/cm³ 0 < H:(Pu + U) ≤ 0.45	885	33,447	161	2282	102	771	67.9	256.6

Table 3
Subcritical Concentration Limits for Plutonium in Homogeneous Mixtures of Plutonium and Natural Uranium of Unlimited Mass[a]

These limits apply to combinations of plutonium isotopes provided $^{240}Pu > {}^{241}Pu$.

PuO_2 in (PuO_2 + UO_2)	Plutonium Content Pu/(Pu + U), wt%
Dry Mixed Oxides, H:(Pu + U) = 0	4.4
Damp Mixed Oxides, 0 < H:(Pu + U) ≤ 0.45	1.8
Oxides in Water	0.13
$Pu(NO_3)_4$ in [$Pu(NO_3)_4$ + $UO_2(NO_3)_2$]	0.65

[a] These limits are not applicable to atom mixtures of elemental plutonium and uranium.

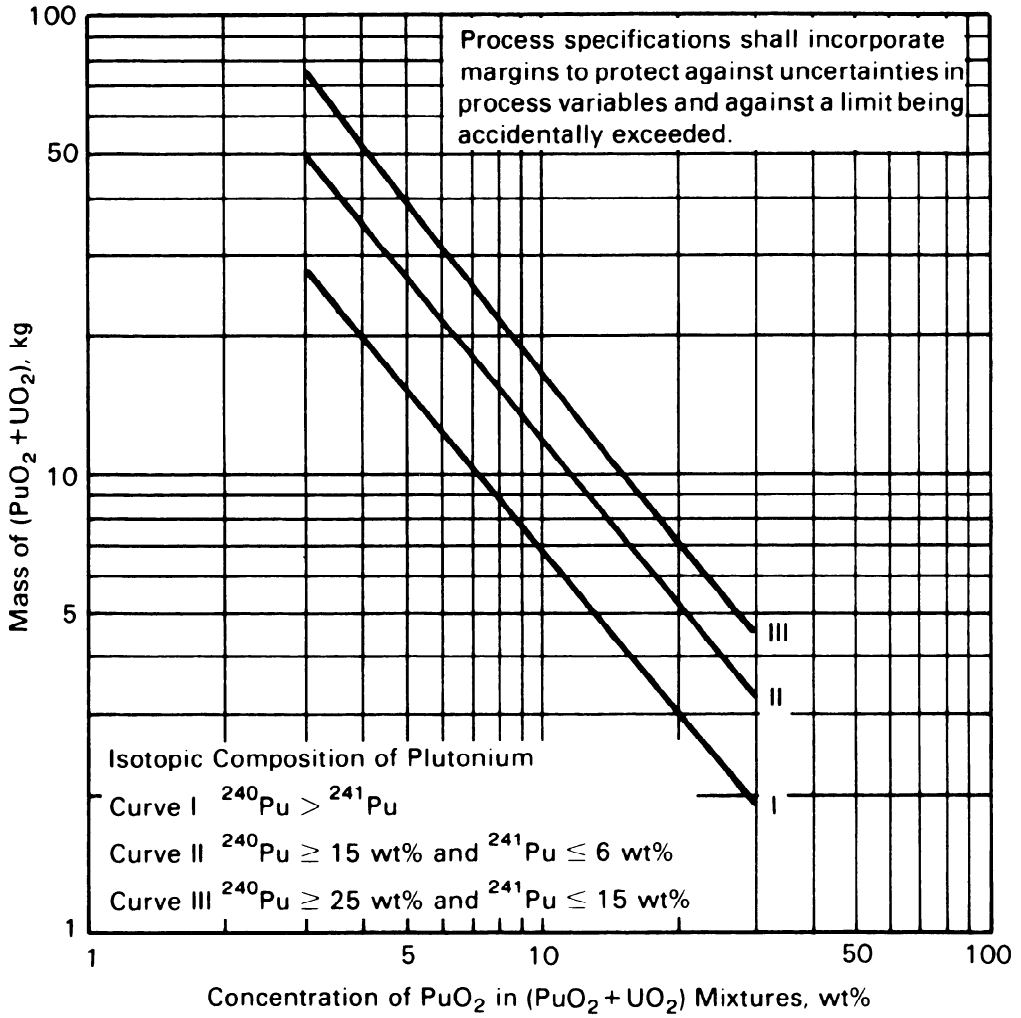

Figure 1. Limiting Mass of Homogeneous Mixtures of the Oxides of Plutonium and Natural Uranium in Water as a Function of the Plutonium Oxide Content.

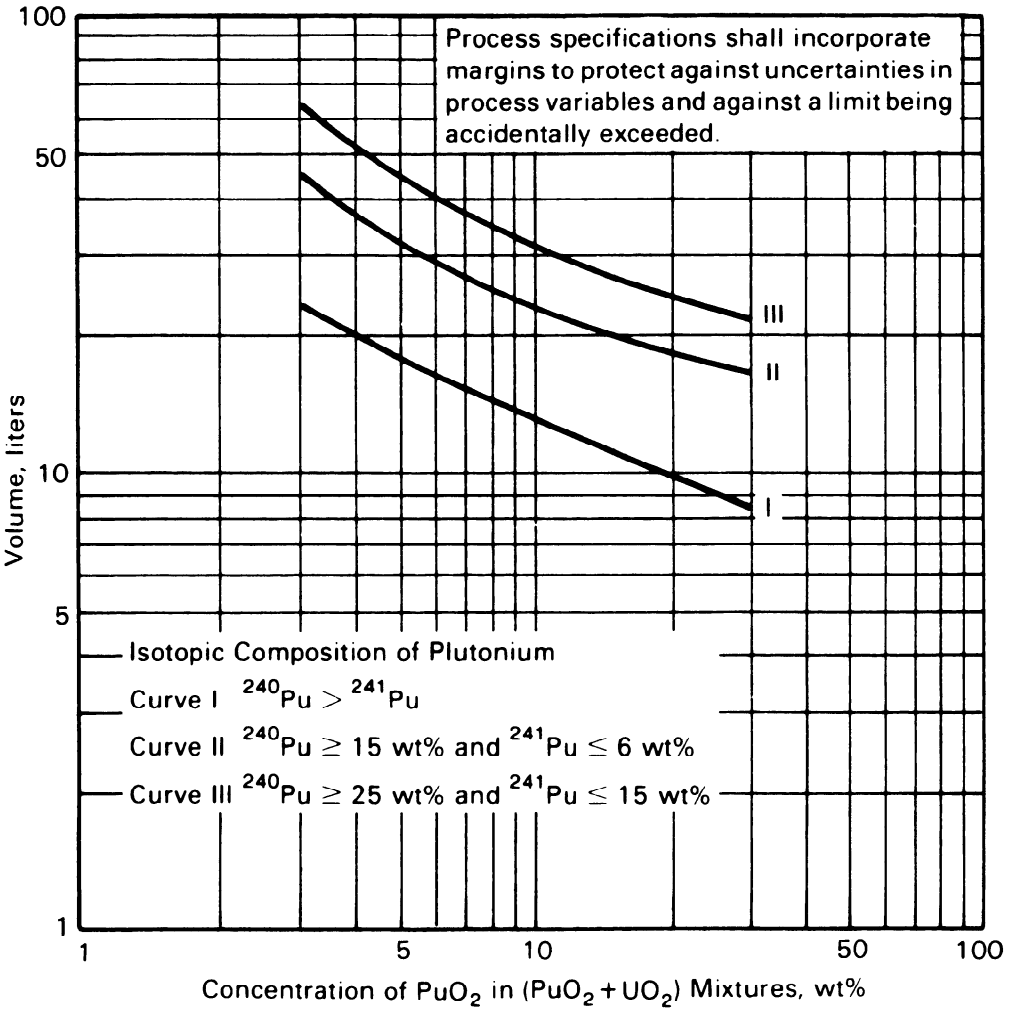

Figure 2. Limiting Volume of Homogeneous Mixtures of the Oxides of Plutonium and Natural Uranium in Water as a Function of the Plutonium Oxide Content. (See Table 1, Footnote b.)

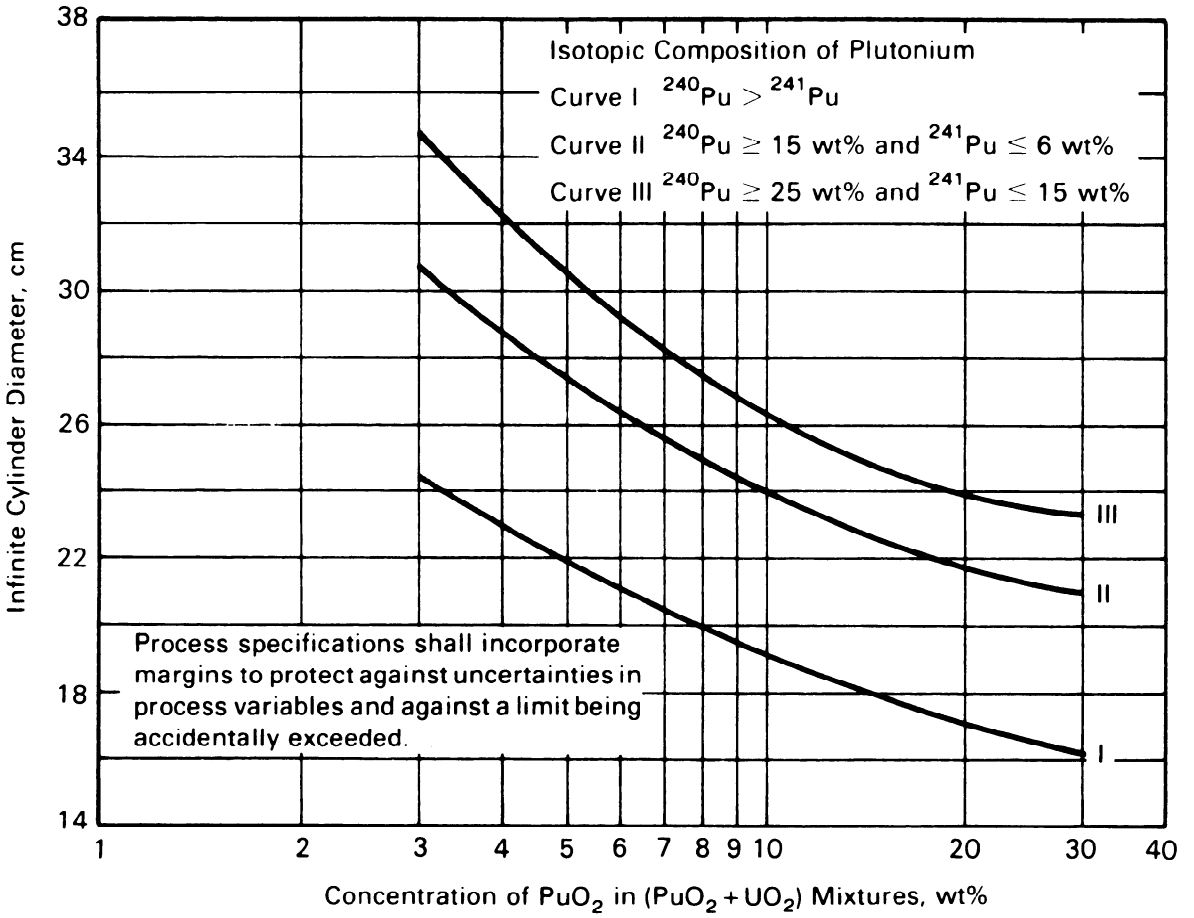

Figure 3. Limiting Diameter of Cylinders Containing Homogeneous Mixtures of the Oxides of Plutonium and Natural Uranium in Water as a Function of the Plutonium Oxide Content. (See Table 1, Footnote b.)

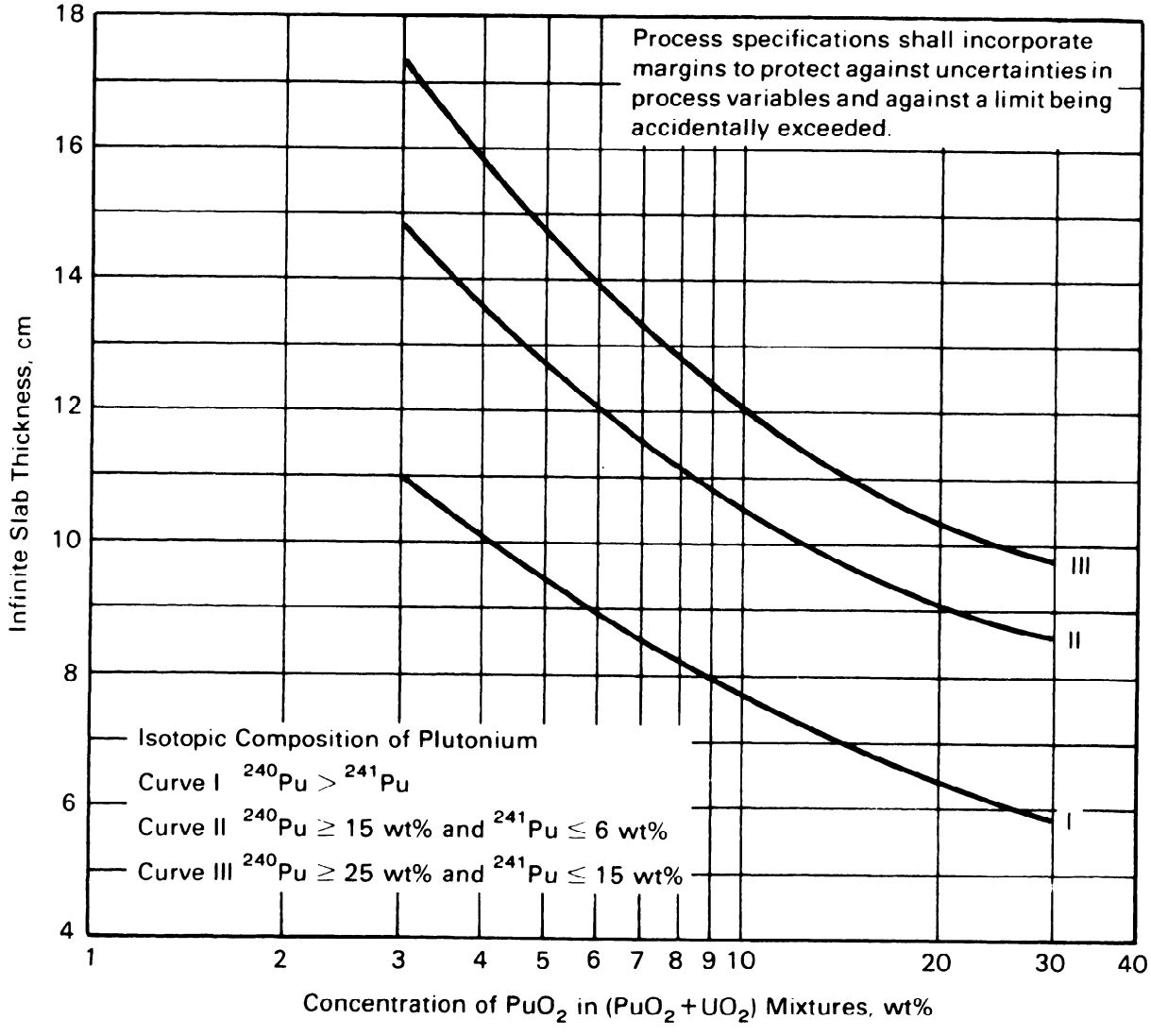

Figure 4. Limiting Thickness of Slabs Containing Homogeneous Mixtures of the Oxides of Plutonium and Natural Uranium in Water as a Function of the Plutonium Oxide Content. (See Table 1, Footnote b.)

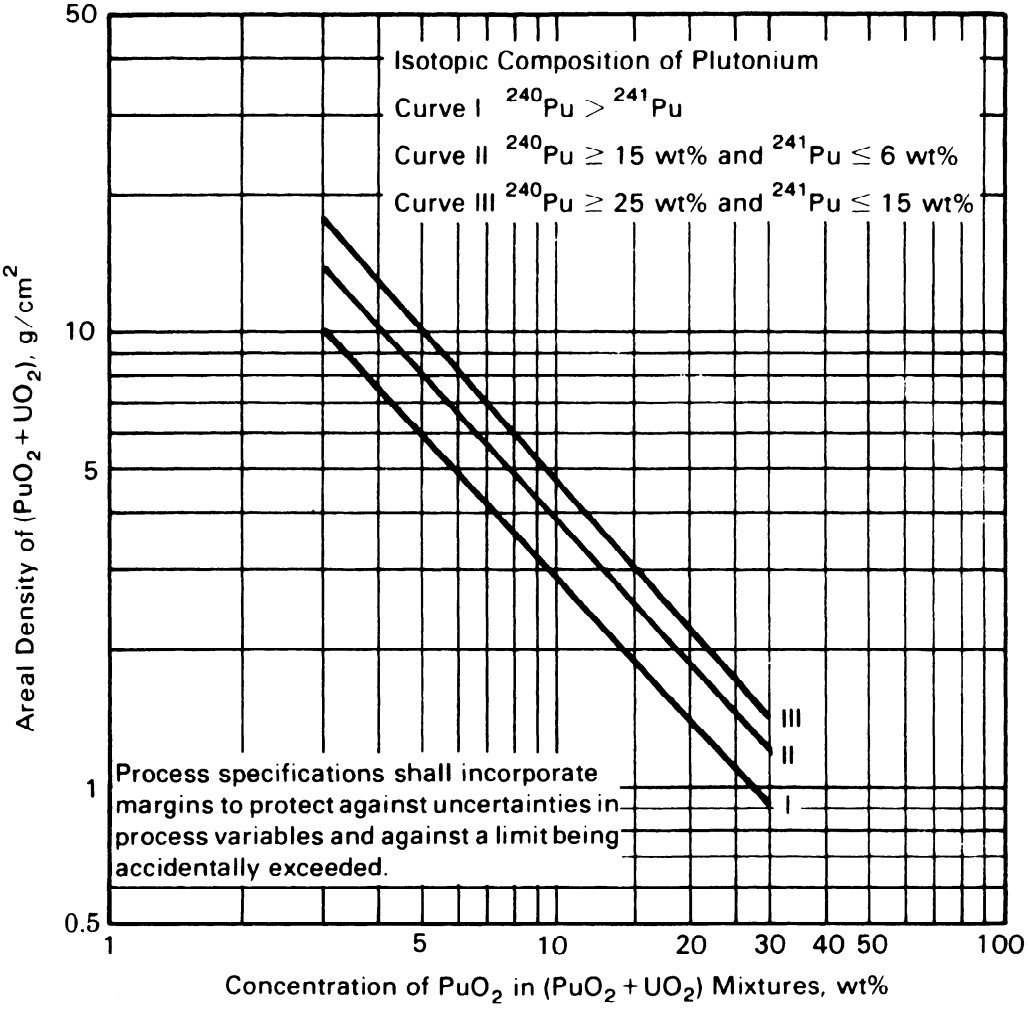

Figure 5. Limiting Areal Density of Homogeneous Mixtures of the Oxides of Plutonium and Natural Uranium in Water as a Function of the Plutonium Oxide Content.

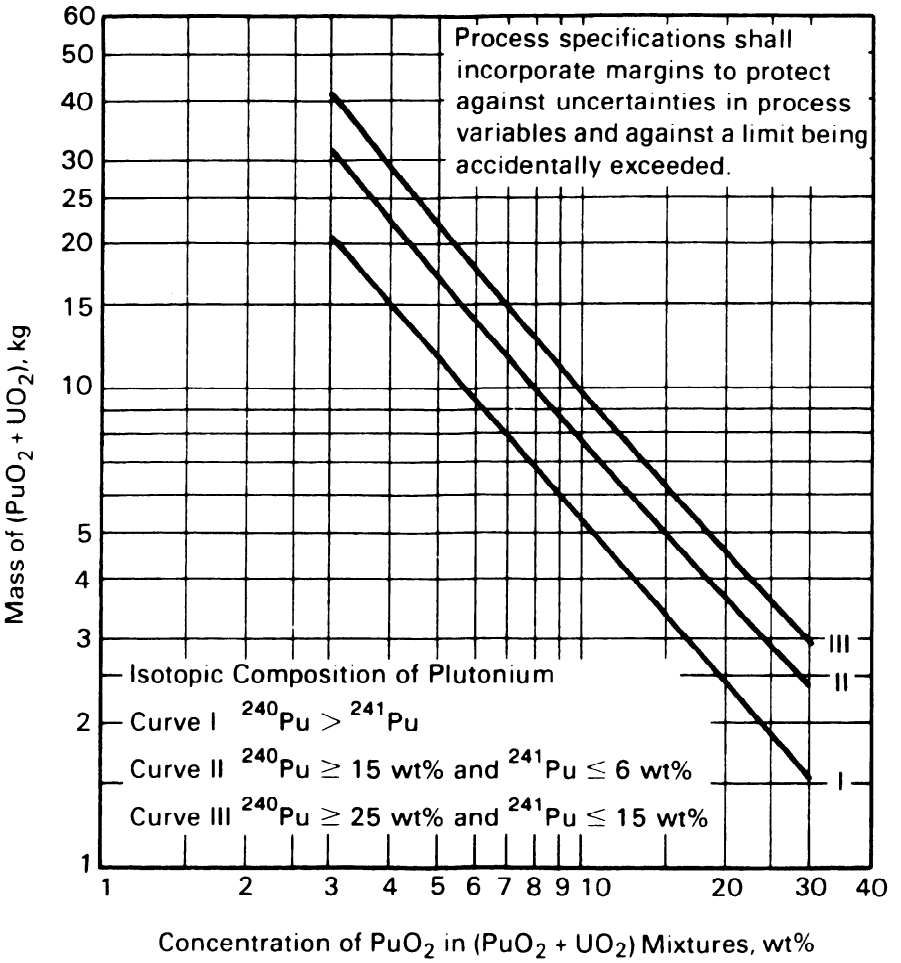

Figure 6. Limiting Mass of Heterogeneous Mixtures of the Oxides of Plutonium and Natural Uranium in Water as a Function of the Plutonium Oxide Content.

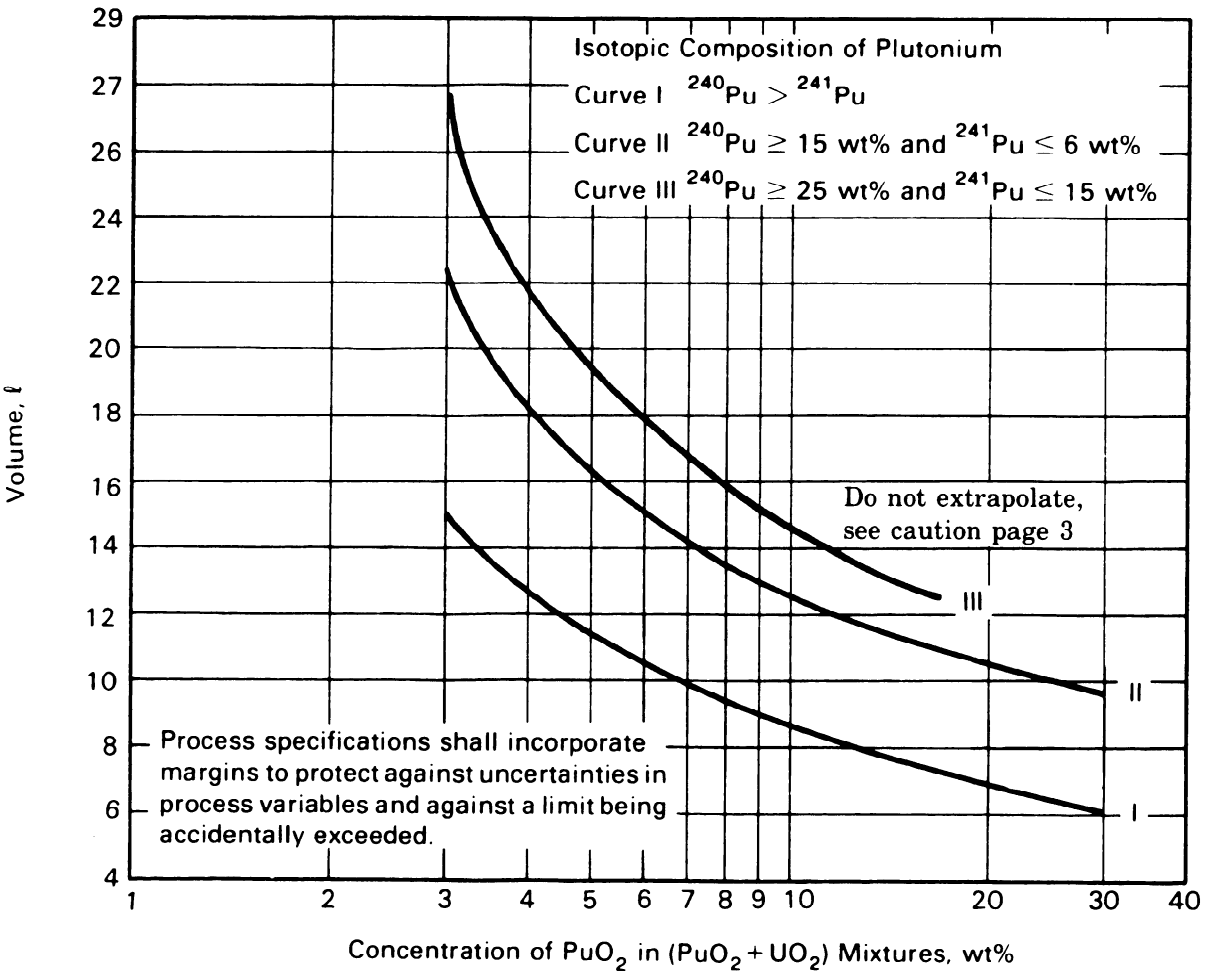

Figure 7. Limiting Volume of Heterogeneous Mixtures of the Oxides of Plutonium and Natural Uranium in Water as a Function of the Plutonium Oxide Content.

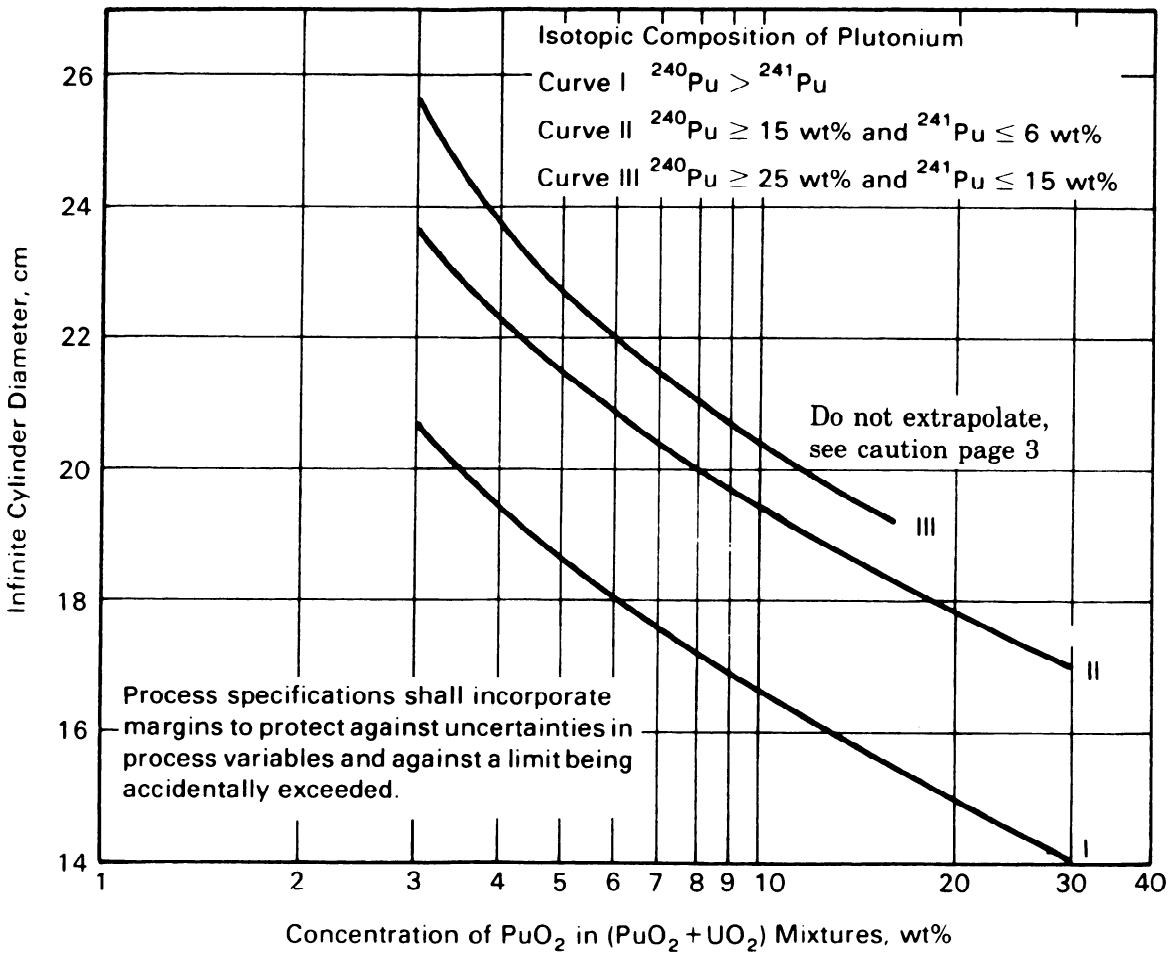

Figure 8. Limiting Diameter of Cylinders Containing Heterogeneous Mixtures of the Oxides of Plutonium and Natural Uranium in Water as a Function of the Plutonium Oxide Content.

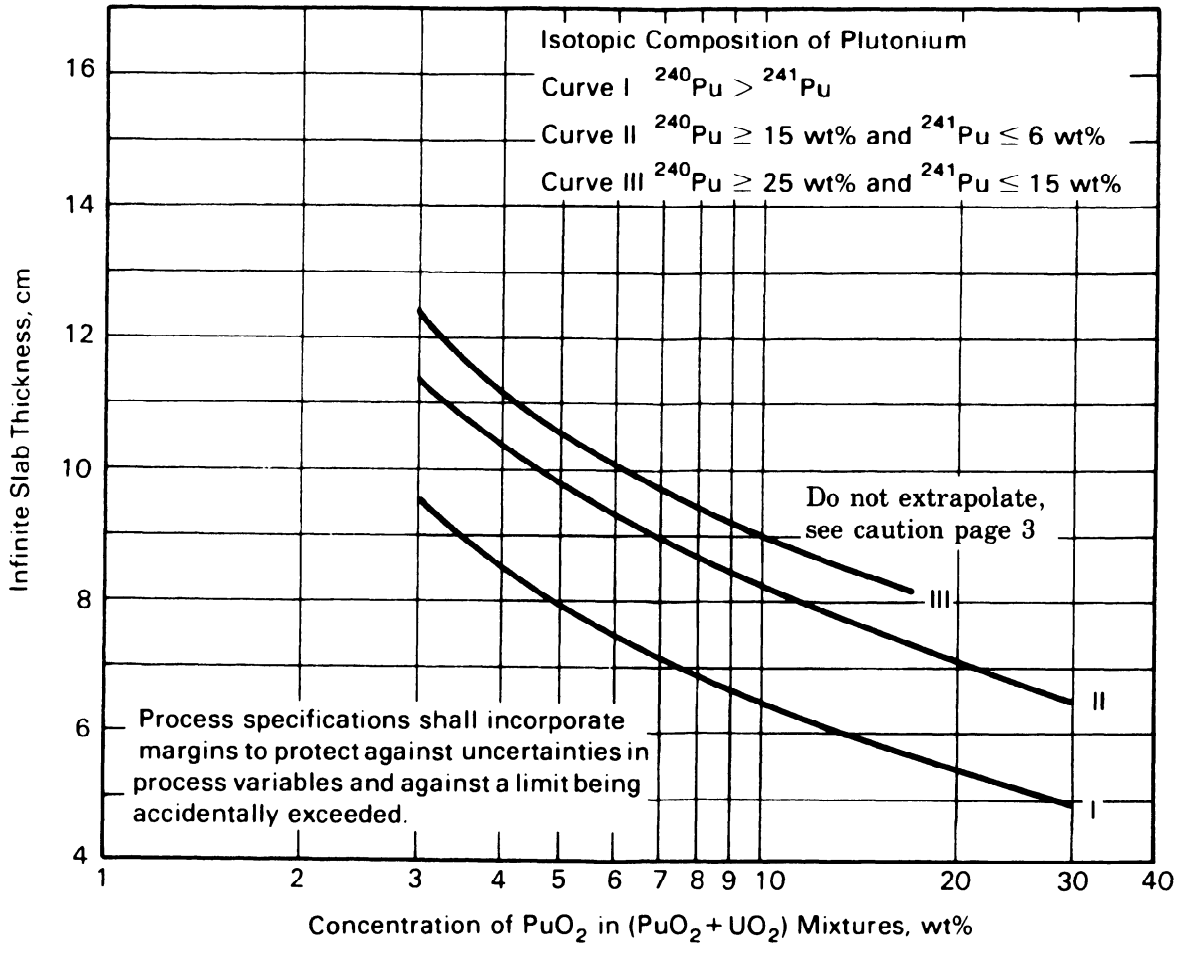

Figure 9. Limiting Thickness of Slabs Containing Heterogeneous Mixtures of the Oxides of Plutonium and Natural Uranium in Water as a Function of the Plutonium Oxide Content.

Appendix

(This Appendix is not a part of American National Standard for Nuclear Criticality Control and Safety of Plutonium-Uranium Fuel Mixtures Outside Reactors, ANSI/ANS-8.12-1987, but is included for information only.)

References to Criticality Data on Plutonium-Uranium Fuel Mixtures

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