Chapter II

SAFETY EXPERIENCE

A. Criticality Accidents

1. General

Present-day criticality controls have been influenced strongly by accidental excursions that have occurred in processing plants in the United States and the United Kingdom. The effectiveness of resulting controls is suggested by the fact that there have been few accidents since the cluster of six that occurred between 1958 and 1964.

There have been eight supercritical accidents in chemical process equipment but none associated with mechanical processing, storage, or transportation. All occurred with aqueous solutions: five involved highly enriched uranium and three involved plutonium. Three of the excursions took place in shielded areas designed for processing irradiated fuel, consequently personnel were protected from the direct radiation.

The consequences of the eight accidents have been two deaths, nineteen significant overexposures of personnel to radiation, no equipment damage, and negligible loss of fissile material. In no case was there any danger to the general public. No incident is attributable to faulty criticality information or to error in its interpretation. Rather, in each case, the cause was related to misuse of equipment, procedural inadequacies or violations, or combinations of these. Most of these accidents resulted in prompt criticality.

Before proceeding from these general remarks to more specific features of the accidents, it may be useful to picture the usual characteristics of a prompt power excursion[8] in a solution.[57] Typically, there is a "fission spike" which may or may not be followed by an oscillatory fluctuation of power. Depending upon the circumstances, secondary spikes or pulses may occur. The fission spike may be described as beginning with an exponential rise in power upon achievement of supercriticality. The rise is arrested by bubbles formed principally by radiolytic dissociation of water, and the solution is driven subcritical causing the power to decrease. The sharp rise and fall in power, i.e., the release of energy at high power but limited to short duration, characterizes the fission spike. If there is no immediate terminating mechanism, this process may be repeated, usually less energetically. Ultimately, upon disappearance of the bubbles, increase in temperature and possible boiling may lead to a quasi-equilibrium condition. This course of events would be affected by changes in conditions that may occur, such as continued addition of material, or loss of material by splashing or evaporation. Of course, loss of solution or redistribution of material may terminate the reaction after the initial burst.

The energy releases associated with the occurrences described below are expressed as numbers of fissions. For convenience, it is noted that 3 x 10^{16} fissions release 1 MW-sec, or 10^6 J, or 240 kcal, or 950 BTU of energy. Most of this energy is deposited in the solution as heat.

A complete listing of criticality accidents before 1990 appears in a review by Stratton, revised by Smith.[30] Details are given in the references cited. Although we will confine our attention to accidents in processing plants, conditions that have led to accidental excursions in critical facilities* are also instructive, and are discussed in the above review. The following accounts of plant accidents are intended to provide not only an idea of the consequences but a general introduction to nuclear criticality safety practices.

2. Criticality Accidents in Processing Plants

The Y-12 Plant, Oak Ridge National Laboratory, TN – June 16, 1958[30, 57–60]

The first of the eight plant excursions was the result of solution leaking into a cleaned cylindrical vessel and being collected with wash water in a 55-gal drum. As a consequence, five persons were exposed severely and three others significantly.

The accident occurred in an area in which highly enriched uranium was being recovered from scrap. In the course of a material inventory, a bank of geometrically subcritical storage vessels had been disassembled and cleaned. Following reassembly, procedures called for leak testing with water, which was to be subsequently drained into a 55-gal drum. In the interval between reassembly and leak testing, uranium solution had accumulated in the vessels through a valve that was supposed to provide isolation from operating equipment upstream. The water being drained into the drum was preceded by this solution. Initial criticality occurred with about 2.1 kg of ^{235}U in 56 liters of solution. A succession of pulses then produced a total of 1.3 x 10^{18} fissions (mostly within 2.8 min) before further dilution decreased the uranium density until the system became subcritical after about 20 min. Although the magnitude of the first and largest pulse was not recorded, subsequent excursion experiments[57] suggest a probable value of about 10^{16} fissions. An initial "blue flash" was observed, and there was no evidence that solution splashed out of the open container.

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*A critical facility is a facility where the critical condition is approached or achieved by plan.

One person who was about 2 meters from the drum at the onset of the excursion received a whole-body dose of ~461 rem[8] (4.61 Sv). Other exposures were 428 rem at ~5.5 m, 413 rem at ~4.9 m, 341 rem at ~4.6 m, 298 rem at 6.7 m, 86.5 rem at 9.4 m, 86.5 rem at 11 m, and 28.8 rem at 15.2 m. These exposures and distances from the drum do not correlate in detail because some exposure may have been incurred during evacuation. Further, it appears that the closest man, who left most rapidly, was exposed for about 5 sec to radiation associated with the initial pulse. Others, responding to the evacuation alarm, presumably were exposed for about 15 sec, which is roughly the interval between the first two pulses. It is apparent that exposures were limited by prompt evacuation.

The following two changes in operating procedures were adopted following the accident. First, whenever transfer lines containing fissile material needed to be isolated from other equipment, they were physically disconnected instead of relying on valves. Second, only vessels that would be subcritical when filled with ^{235}U-enriched uranium solutions, that is, those with favorable geometry,[8] were permitted in solution areas.

The Los Alamos Scientific Laboratory, NM – December 30, 1958[30, 57, 61]

The next accident resulted from concentrated plutonium in a solvent layer which was found in a large tank that was supposed to contain only lean aqueous-organic emulsion. A transient change of shape of the solvent layer when a stirrer was started established criticality of short duration. The result was a fatality and two other significant exposures.

The accident occurred in an area where residual plutonium, usually about 0.1 g/L, and americium were recovered from dilute raffinate. Because the normal plutonium inventory was only 0.1 kg, solvent extraction was conducted in large closed tanks. As at Y-12, a material inventory was in progress and it was intended that the tanks be emptied and cleaned individually. Instead, residues and acidic wash solutions from four vessels were combined in a single 850-liter, 96.5-cm-diameter tank. Many interconnecting transfer lines made this possible. An excursion of 1.5×10^{17} fissions occurred when a stirrer in this tank was started.

As discovered later, a 20.3-cm-thick, 160 liter, organic layer floating on a dilute aqueous solution contained 3.10 kg of plutonium. It is presumed that the source of this plutonium was solids that had accumulated gradually in the tanks and transfer lines during 7.5 years of operations. The initial effect of the stirrer was to disturb the organic layer sufficiently for supercriticality. The stirring rapidly mixed the two phases, diluting the plutonium to a subcritical density.

The operator, who was looking into the tank through a viewport, received an exposure of (12 ± 6) x 10^3 rem and died 36 hours later. Two men who went to aid the victim received doses of 134 and 53 rem. There was neither damage to equipment nor contamination although a shock displaced the tank support about 1 cm. A recording radiation detector[62] 53 meters away was activated* and a flash of light was seen from an adjoining room.

The entire recovery plant, which had been scheduled for rebuilding after another six months of operation, was retired immediately. After ultimate conversion to geometrically subcritical equipment, the following corrective measures were adopted. Unnecessary solution-transfer lines were avoided, and auxiliary vessels such as vent tanks and vacuum-buffer tanks were "poisoned" with borosilicate glass Raschig rings. Additionally, written procedures and nuclear-safety training were improved. Periodic surveys with portable neutron detectors to locate abnormal plutonium deposits were instituted. The accident also led to more complete coverage of process areas by improved gamma-ray-sensing radiation alarms.

The Idaho Chemical Processing Plant, Idaho Reactor Testing Area, ID – October 16, 1959[30, 57]

This excursion was the result of inadvertently siphoning highly enriched uranium solution from a bank of geometrically subcritical storage cylinders into a large waste tank. Although heavy shielding required for irradiated-fuel processing protected personnel from direct radiation, fission products vented into working areas resulted in two significant dosages, of 50 and 32 R(roentgen)[8], mostly as beta radiation to the skin.

The siphoning, through a trapped vent system to the waste tank, started as a result of air sparging the storage cylinders. About 200 liters of solution containing 34 kg of ^{235}U transferred into about 600 liters of water in the 1.9 x 10^4-liter waste tank. Criticality in this tank led to a total of 4 x 10^{19} fissions over a period of about 20 min. It is postulated that an initial spike of $\sim 10^{17}$ fissions was followed by smaller pulses, then by more-or-less stable boiling that distilled 400 liters of water into another tank. The exceptionally large yield was the result of the large solution volume and long duration of the reaction, not of the intensity of the excursion.

The incident disclosed the need for improved evacuation procedures and demonstrated the value of radiation alarms in areas that might be affected by an excursion elsewhere. Equipment and operating procedures were modified to establish several lines of defense against inadvertent transfer of fissile material.

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*The radiation detector did not produce an audible warning sound and hence was not a criticality accident alarm.

The Idaho Chemical Processing Plant, Idaho Reactor Testing Area, ID – January 25, 1961[30, 57]

This excursion occurred when a large air bubble forced enriched-uranium solution out the top of a 12.7-cm-diameter section of an evaporator and into a 61-cm-diameter vapor-disengagement cylinder above the normal solution level. The heavy concrete shielding required for irradiated-fuel processing protected personnel from direct radiation, the ventilation system prevented airborne activity from entering work areas, and equipment design excluded the possibility of a destructive or persistent excursion. Nevertheless, this incident is instructive because consequences could have been serious in an unshielded area.

Apparently air used to clear a plugged line and to improve operation of two pumps was the source of the bubble that forced 40 liters of solution containing 8 kg of ^{235}U into the larger-diameter section. The resulting excursion, probably a single pulse, had a magnitude of 6 x 10^{17} fissions. Operation was resumed within an hour.

Because the possibility of an excursion in the vapor-disengagement cylinder had been foreseen, there was provision for drainage into a subcritical configuration, which prevented both pressure buildup and a sustained reaction. Although consequences were minor, the 61-cm-diameter cylinder ultimately was "poisoned" by a grid of stainless steel plates containing 1 wt% natural boron. Steps were also taken to prevent the introduction of air into solution lines where the effect could be undesirable.

The Recuplex Plant, Hanford Works, WA – April 7, 1962[30, 57, 63]

This incident occurred when liquid from a sump was collected in a 69-liter, 45.7-cm-diameter vessel. The liquid, unidentified at the time, was subsequently shown to contain between 1400 and 1500 g of plutonium in a volume of about 46 liters after the addition of lean solutions. The only significant exposures were 110, 43, and 19 rem, received by personnel at distances from the excursion of about 2.1, 3.2, and 7 m, respectively.

The accident took place in plutonium-recovery equipment located in a room-size glove box. The vessel in which the excursion occurred was normally used for transfer of a dilute sidestream from solvent-extraction columns to a secondary recovery process, similar to the raffinate-treatment process of the Los Alamos accident. Apparently the concentrated solution had overflowed from a favorable geometry tank and was sucked into the 45.7-cm-diameter vessel through a temporary line used for cleanup operations that were in progress. A total yield of 8.2 x 10^{17} fissions occurred over 37 hours, with about 20% of the energy released in the first half hour. An initial pulse of no more than 10^{16} fissions was followed by smaller pulses for about 20 min., after which boiling ultimately distilled off enough water to stop the reaction.

The initial pulse, accompanied by the usual blue flash, triggered a criticality accident alarm, and the area was evacuated promptly, presumably before a second pulse. A unique feature of the analysis of events was the use of a small, remotely controlled robot developed for handling irradiated fuel. By means of this device, the excursion site was located, meters were positioned and read, and valves were operated without exposures to personnel.

A new plant to replace Recuplex had been authorized before the accident, and operations were not resumed until it became available. In the modern plant, vessels that are not subcritical by favorable geometry usually contain neutron absorbers, the system is adaptable to a variety of uses without improvisation, and equipment is easier to keep clean.

Wood River Junction Plant, RI – July 24, 1964[30, 57]

This accident was initiated when concentrated enriched-uranium solution was inadvertently poured into a 46-cm-diameter tank. The first of two excursions resulted in a lethal exposure and the second, about 2 hours later, was primarily responsible for two other significant radiation doses.

Startup difficulties in this plant for recovering highly enriched uranium from scrap led to an unusual accumulation of trichloroethane (TCE) solution of low uranium density. Small amounts of uranium were recovered by tedious hand agitation of the TCE with sodium-carbonate solution. An easier process was improvised, in which the TCE was treated in the 46-cm-diameter tank that had been intended only for the makeup of sodium-carbonate solution used in the normal recovery process. Neither the plant superintendent nor one of three shift supervisors was aware of this practice. Meanwhile, solutions of unusually high ^{235}U density, resulting from cleanout of plugged equipment, had been stored in 11-liter, 12.7-cm-diameter bottles identical to those that contained the contaminated TCE. Apparently, a bottle of the concentrated solution was mistaken for TCE and was poured into the sodium-carbonate solution being stirred in the makeup tank. The shock from a single pulse of $\sim 10^{17}$ fissions knocked the operator onto the floor and splashed part of the solution out of the tank. A flash of light was observed. The victim received an exposure estimated to be 10,000 rad[8] and died 49 hours later.

It appears that enough solution was ejected from the tank (the final content of the vessel was 2 kg of uranium in about 40 liters) so that the stirrer vortex was sufficient to maintain subcriticality. Two hours after the first excursion, however, two men entered the area, stopped the stirrer and restarted it some minutes later, after which they drained the tank. These two received radiation doses between 60 and 100 rad. Evidence of neutron exposure suggested a second less violent excursion while the stirrer was off, which was not detected because the radiation alarm continued to sound after the first excursion. The combined yield of both excursions was 1.3×10^{17} fissions.

Before operation was resumed, there were extensive analyses of the process. These included penetrating reviews and modifications of operating and emergency procedures, criticality limits and controls, uranium accountability and material balance practices, health physics procedures and controls, and training. Favorable geometry equipment for recovering uranium from TCE, which had been planned previously, was put into operation.

UKAEA Windscale Works, Great Britain – August 24, 1970[30, 57, 64]

This excursion is reminiscent of the Los Alamos accident, but without severe consequence. Similarities are the unsuspected buildup of plutonium in an organic solvent layer and a transient change of geometry that led to criticality of short duration. The total fission yield was on the order of 10^{15} fissions, and exposures were negligible – less that 2 rad for the two closest workers, who were protected somewhat by shielding.

The excursion, detected by the criticality alarm system, took place at the head end of a process for recovering plutonium by solvent extraction. Normally, aqueous solution having a density of ~6 g Pu/L from a dissolver and a "conditioner" for feed adjustment was raised by vacuum into a transfer vessel, then flowed by gravity through a trap and into a tank that supplied metered solution to a favorable-geometry extraction column. When 40 liters of organic solvent from an unknown source entered the vacuum transfer vessel, the trap isolated the floating layer of solvent instead of permitting it to drain. So instead of serving the intended safety purpose, the trap allowed the solvent to accumulate plutonium in the transfer vessel, little by little, from aqueous batches pouring through it. At the final density of 55 g Pu/L in the solvent, it appears that an emulsion band between the solvent and aqueous solutions led to criticality during the brief period after the flow stopped and before the two phases of emulsion separated. This sequence of events was reconstructed and demonstrated by means of an inactive transparent replica of the transfer system.

Before the plant was returned to service, neutron monitors to detect plutonium accumulations were installed on all vessels without favorable geometry. Furthermore, the drain traps were modified to permit positive drainage and to facilitate washout procedures.

The Idaho Chemical Processing Plant, Idaho Reactor Testing Area, ID – October 17, 1978[30]

The most recent of eight process excursions occurred in a shielded cell of a plant for reprocessing irradiated fuel. The dissolved fuel, as dilute aqueous solution, was introduced into the first of a series of pulsed columns for extracting and purifying the highly enriched uranium.

Normally, in the first column the uranium is extracted into an organic stream which enters the second column for the removal of fission products by a stream of water. The water is buffered with aluminum nitrate to prevent significant takeup of uranium, then reintroduced along with feed to the first column to remove traces of uranium. Most of the uranium, in the organic stream, leaves the second column for further processing.

Abnormal operation occurred as a result of water leaking into the aluminum nitrate makeup tank, which was not detected because of malfunctioning instrumentation. As a result, the solution of aluminum nitrate entering the second column was too dilute to prevent appreciable uptake of uranium. Instead of leaving with the organic, the uranium recycled successively through the first and second columns building up to an estimated 10 kg in the second column.

This was sufficient to cause an excursion of about 2.7 x 10^{18} fissions extending over one-half hour until the reaction was terminated by improved mixing after feed was turned off.

Although there was insignificant personnel exposure and no damage of equipment, the plant underwent an extended and expensive shutdown. Operating procedures were revised, and there was increased emphasis on plant maintenance and operator training. Further, a highly instrumented plant protection system with automatic controls was installed.

3. Examples of Radioactive Contamination from Chemical Explosions

Explosion at Kyshtym, USSR – 1957[65–66]

Chemical accidents, not just criticality accidents, have resulted in severe human exposure to nuclear radiation. A chemical explosion at the Kyshtym waste repository, which did not involve the potential for criticality, serves to illustrate this point. The explosion occurred in a concrete tank containing nitrate-acetate fission-product waste. A fault developed in the cooling system used to offset fission-product heating. As a result, in the mistaken belief that coolant was no longer necessary, its flow was stopped. The wastes then dried, leaving a highly explosive mixture of nitrate salts and acetate, which ignited as the temperature increased.

As a result of the accident, 10,180 people were evacuated throughout an area of roughly 1000 km^2. There were no fatalities. After 25 years of surveillance, the greater part of this area had been repopulated. Health effects were summarized as follows. "In conclusion, we may note that observations on health, morbidity and mortality among the population subjected to the accidental release of radiation – with whole body exposure doses from 1 to 52 cSv* and irradiation of individual organs up to 150 cSv – have revealed no significant deviations from the comparable values found among healthy unexposed individuals."

This incident provides a reminder that attention to criticality safety does not eliminate the need to guard against other types of radiation accidents.

Explosion of Ion-Exchange Column, Hanford, WA – 1976[67]

In the United States, chemical explosions capable of spreading contamination have been much less severe than that at Kyshtym. That, however, is no reason for complacency, for they have occurred and might happen where consequences could be more serious.

The 1976 explosion at Hanford was in a cation exchange column for americium recovery. The steel column ruptured, shattering windows of a glove box in which it was contained, injuring and contaminating an operator, and contaminating others nearby. The violent

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*One cSv equals one rem; stated doses were received before evacuation.

reaction was presumed to entail nitric acid and products of cation-resin degradation. The degradation resulted from exposure of the resin to radiation from 100 g of americium for more than five months.

The report of this accident refers to an earlier ion-exchange column explosion at another site, but no detail is given.

B. General Observations

Because of timely evacuations initiated by criticality alarms, exposures of personnel to criticality events in unshielded facilities were limited to the direct radiation from the initial pulse or two. The exposure limitations of eleven individuals from the two prolonged reactions are attributable to their evacuation following alarms. It may be concluded that lives were saved by immediate evacuation, showing the value of radiation-initiated alarms installed where the potential for an accidental excursion is significant. An American National Standard addresses this subject.[46]

The two fatalities were suffered by persons within a few feet of an excursion; significant exposures were received by others at distances extending to 15 m (50 ft). This observation may be generalized to a certain extent by Figure 1. This figure shows that personnel doses normalized to excursions of 10^{17} fissions and crudely adjusted to exposure times of $\sim$15 sec correlate roughly with distances from the source. For the typical excursion of 10^{17} fissions, the distances resulting in early fatalities are similar to those of a moderate chemical explosion.

The relatively large number of plant accidents, six between 1958 and 1964, calls for some explanation. An increased demand for plutonium and enriched-uranium production without a corresponding reassessment of criticality control in existing processing facilities had some impact. Plants that had been designed for moderate capacity and were operating with minimal criticality safety guidance were called upon to increase throughput and perform a wider variety of tasks. Even though the potential for accidents had increased, a long accident-free period prior to 1958 made it difficult to justify an accelerated effort to improve criticality safety. On the other hand, the occurrence of a criticality accident provides an immediate incentive to improve criticality control. For example, the plutonium recovery plants at Los Alamos and Hanford were not modernized until the accidents occurred there. As might be expected, the cluster of accidents between 1958 and 1964 emphasized the need to improve nuclear criticality safety. Most importantly, additional experiments were performed to determine critical configurations for a larger range of materials and geometries. The resulting body of experimental data led to more definitive guidance and enhanced techniques for criticality control. This effort led to a significant reduction in the accident rate after 1964 in that only one accident has occurred since then.

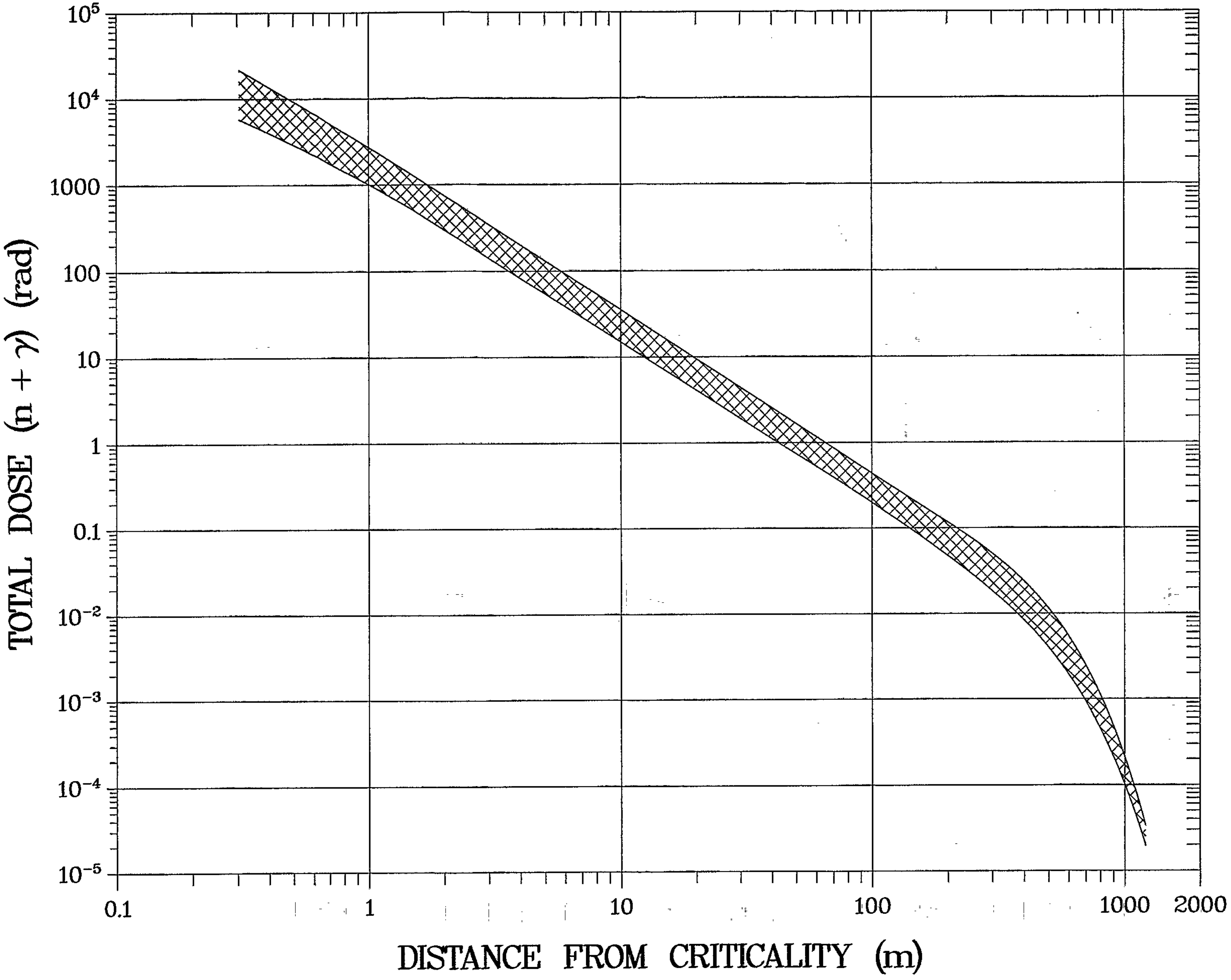

Figure 1

Fig. 1. Approximate correlations of dose with distance from excursions normalized to 10^{17} fissions. This was adapted from Ref. 68 and incorporates the doses received upon evacuation as given in this chapter.

The fact that all the accidental excursions involved solutions of plutonium or highly enriched uranium is not surprising. Small critical mass and the mobility and ease of solute exchange that make solutions so desirable in chemical processing, invite criticality in unexpected locations. By contrast, the movement of solids is more apparent, more easily controlled, and the amount of fissile material needed to achieve a critical configuration is much larger. Containers sufficiently larger than their solid contents can prevent criticality in the event that they fall together as a result of structural collapse.* As we shall see, it is more important that criticality control be effective for certain solids than for solutions, although the problems with solutions are much more subtle.

Typical accident experience with solutions of fissile material shows minimal damage to equipment and no exposure of the public to radiation. Disruptive pressures resulting in dispersion of radioactive contamination would require unusual circumstances, such as containment without pressure relief. Properties of solution excursions are illustrated further by an extensive series of kinetic experiments conducted at the Dijon Laboratory of the French Commissariat a l'Energie Atomique.[70] Certain types of conceptual accidents with solid fissile material, notably with ^{235}U metal, are more likely to be violent.[30] Fortunately, as noted above, it is not difficult to foresee the conditions, such as large pieces of metal falling together, that might lead to an extreme accident. Control of these conditions is usually straightforward and is emphasized in plant operations; for example, by limiting the location and movement of massive pieces in a single plane.

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*One hundred twenty-five units, each consisting of 10 kg of enriched uranium metal in a convenient 20.3-cm-diameter x 24.1-cm-deep can, would remain subcritical if gathered together on a concrete floor.[69]

Chapter III

LIMITS FOR INDIVIDUAL UNITS

A. Subcritical Limits of Controlling Parameters

Subcritical limits for individual units appear in *American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors, ANSI/ANS-8.1*, and *American National Standard for Nuclear Criticality Control and Safety of Plutonium-Uranium Fuel Mixtures Outside Reactors, ANSI/ANS-8.12.*[71] The former source, *ANSI/ANS-8.1*, includes all such limits except those for the plutonium-natural uranium combinations of *ANSI/ANS-8.12*, which are appropiate for the fissionable material of mixed-oxide reactors.

The subcritical limits throughout *ANSI/ANS-8.1* result from calculations validated by comparison with experimental data. The computational bias is incorporated in a subcritical margin of $\Delta k_{eff} = 0.02$ below minimum indicated criticality.[72–74] The subcritical limits apply to units with full water reflection. This small margin is effectively increased if reflection is limited, for example, to that of a container, a person nearby, objects more than one-meter distant, and parts of a room (i.e., incidental reflection). Regardless, contingencies that include the effects of more probable deviations must be considered in applying any of these limits. In other words, the following requirement of the Standard must be satisfied.

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

For subcritical limits in *ANSI/ANS-8.12*, the margin is $\Delta k_{eff} = 0.05$, and full water reflection is again specified.[75] The greater margin demonstrates judgment as to the adequacy of the range of supporting experimental data and in no way reduces the need to consider contingencies. See Section F of this chapter for a discussion of typical contingencies.

B. Subcritical Limits for Systems of Fissile Nuclides

1. Introduction

This section is an expansion of Section 5 of Standard *ANSI/ANS-8.1*, which bears the same title. The section gives the subcritical limits for various parameters, such as the mass or volume of fissile material, any one of which is sufficient for implementing criticality control. Subcritical limits, as discussed in Section A of this chapter, are implied. The subcritical limits of Tables 1 through 5 are taken directly from Standard *ANSI/ANS-8.1*.

2. Hydrogen-Moderated Systems

Aqueous Solutions[72–74, 76]

The subcritical limits of Tables 1 and 2 apply to solutions reflected by the equivalent of an unlimited thickness of water. Masses and volumes apply to spheres and so are conservative for other shapes. The limits expressed for cylinder and slab dimensions apply, respectively, to the diameter of a uniform circular cylinder of unlimited length and to the thickness of a uniform slab of unlimited area. These dimensions are conservative for a cylinder of finite length or a slab of finite area. Areal density is defined as the product of the thickness of a uniform slab and the density of fissile material within the slab; hence, it is the mass of fissile material per unit area of the slab.

For plutonium in which the content of ^{240}Pu exceeds that of ^{241}Pu, the mass, density, and areal density limits of Table 1 apply to the sum of ^{239}Pu and ^{241}Pu. It should be noted that the content of ^{240}Pu exceeds that of ^{241}Pu in typical materials encountered in a reactor fuel cycle. The limits of Tables 1 and 2 are appropriate for many commonly encountered reflector conditions. An example of a reflector other than thick water is the metal-water combination of a cooling jacket and a steel wall of moderate thickness. Sometimes water-flooding may be a reasonable assumed contingency, but, where this is not the case, the adoption of values for complete water reflection allows for unknown neutron reflecting properties of nearby concrete walls, floors, neighboring water lines and process vessels, and transient personnel. Closely fitting reflectors of thick beryllium, beryllium oxide, heavy water, concrete, lead, or graphite are examples of exceptions for which the listed limits are not appropriate. Composite reflectors, e.g., thick steel outside a thin hydrogenous reflector, may be very effective, thus requiring explicit evaluation.

Organic Mixtures and Uniform Slurries

The limits of Tables 1 and 2 may be used for effectively homogeneous hydrogen-moderated mixtures, e.g., organic solutions and macroscopically uniform slurries, provided the atomic ratio of hydrogen-to-fissile-material does not exceed that of a water solution having the same density of fissile material.* Note that experiments at Hanford[77] show nearly identical critical numbers of reactor fuel pins latticed in water and in organic (dodecane) moderator at the same value of H/Pu.

This provision is satisfied by most common mixtures, such as oxides combined with organic material. For plutonium, an additional restriction is that the nitrogen-to-plutonium atomic ratio be at least 4.0 throughout.

Nonuniform Slurries

Single-parameter limits may be assigned to certain nonuniform slurries, provided the restrictions for uniform slurries are satisfied at all locations within the slurry. In that case, the subcritical mass limits for ^{233}U, ^{235}U, and ^{239}Pu are 0.52, 0.70, and 0.45 kg, respectively.[78] For vertical cylinders or slabs on edge, where density gradients arise entirely from gravitational settling (i.e., a gradient along the cylinder axis or parallel to the slab face), the limits of Table 1 on cylinder diameter and slab thickness may be used. The areal density limits of that table are valid for a horizontal slab subject only to gravitational settling, provided the restrictions for uniform slurries are met throughout. Where there are variations in the areal density, the maximum value shall not exceed the limit.

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*Quantities characteristic of solutions appear in Table 7 of Reference 11.

Table 1

Single-Parameter Subcritical Limits for Uranium and Plutonium Solutions, Reflected by an Effectively Infinite Thickness of Water

	Subcritical Limit						
Fissile Solute	Mass of Fissile Nuclide (kg)	Diameter of Cylinder of Solution (cm)	Thickness of Slab of Solution (cm)	Volume of Solution (L)	Density of Fissile Nuclide (g/L)	Atomic Ratio[a] of Hydrogen to Fissile Nuclide	Areal Density of Fissile Nuclide (g/cm^2)
$^{233}UO_2F_2$	0.54	10.5	2.5	2.8	10.8	2390	0.35
$^{233}UO_2(NO_3)_2$	0.55	11.7	3.1	3.6	10.8	2390	0.35
$^{235}UO_2F_2$	0.76	13.7	4.4	5.5	11.6	2250	0.40
$^{235}UO_2(NO_3)_2$	0.78	14.4	4.9	6.2	11.6	2250	0.40
$^{239}Pu(NO_3)_4$	0.48	15.4	5.5	7.3	7.3	3630	0.25

Table 2

Subcritical Limits for Solutions of $Pu(NO_3)_4$ Containing ^{240}Pu, Reflected by an Effectively Infinite Thickness of Water

	Subcritical Limit						
Solution	Mass of Pu (kg)	Cylinder Diameter (cm)	Slab Thickness (cm)	Volume (L)	Density of Pu (g/L)	Atomic Ratio[a] (H/Pu)	Pu Areal Density (g/cm^2)
$\geq$ 5 wt% ^{240}Pu $\leq$ 1 wt% ^{241}Pu	0.57	17.4	6.7	10.0	7.8	3400	0.28
$\geq$ 15 wt% ^{240}Pu $\leq$ 6 wt% ^{241}Pu	0.78	19.5	8.0	13.6	8.9	2980	0.34
$\geq$ 25 wt% ^{240}Pu $\leq$ 15 wt% ^{241}Pu	1.02	21.3	9.2	17.2	10.2	2600	0.40

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[a] Lower limit.

3. Metal Units

Single-parameter subcritical limits[4, 79–80] for units of water-reflected fissile metal appear in Table 3. The mass limits and the ^{235}U enrichment limit for uranium metal apply to a unit without reentrant void that can be occupied by water or other moderator. These limits may be extended to a group of small pieces having the same total mass, provided there can be no moderator between the pieces. The limits for ^{233}U and ^{235}U of Table 3 may be applied to uranium containing ^{234}U, ^{236}U, and ^{238}U, provided the masses of ^{234}U and ^{236}U are included with that of ^{233}U or ^{235}U.

4. Oxide Units

The single-parameter limits of Tables 4 and 5 apply to oxides that have less than 1.5 wt% water.[4] The 1.5 wt% water is chosen because absolutely dry oxide can seldom be guaranteed. Table 4 holds for oxides compacted to as much as the stated maximum density. As footnote 10 of Reference 4 points out, it is possible for this density to be exceeded in a highly compacted oxide. Table 5 applies to uncompacted oxides at no more than one-half maximum density.

Table 3

Single-Parameter Subcritical Limits for Metal Units, Reflected by an Effectively Infinite Thickness of Water

	Subcritical Limit			
Nuclide	Mass of Fissile Nuclide (kg)	Cylinder Diameter (cm)	Slab Thickness (cm)	Uranium Enrichment (wt% ^{235}U)
^{233}U	6.0	4.5	0.38	–
^{235}U	20.1	7.3	1.30	5.0
^{239}Pu	5.0	4.4	0.65	–

Table 4

Subcritical Limits for Oxides Containing No More Than 1.5 wt% Water, at Full Density, Reflected by an Effectively Infinite Thickness of Water

Compound	Subcritical Limit				
	Mass of fissile nuclide, (kg)	Mass of oxide,[a] (kg)	Cylinder diameter, (cm)	Slab thickness, (cm)	Maximum dry bulk density for which limits are valid (g/cm^3)
$^{233}UO_2$	10.1	11.7	7.2	0.8	10.75
$^{233}U_3O_8$	13.4	16.0	9.0	1.1	8.15
$^{233}UO_3$	15.2	18.7	9.9	1.3	7.16
$^{235}UO_2$	32.3	37.2	11.6	2.9	10.84
$^{235}U_3O_8$	44.0	52.8	14.6	4.0	8.21
$^{235}UO_3$	51.2	62.6	16.2	4.6	7.22
$^{239}PuO_2$	10.2	11.5	7.2	1.4	11.49

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[a] These values include the mass of any associated water up to the limiting value of 1.5 wt%.

Table 5

Subcritical Limits for Oxides Containing No More Than 1.5 wt% Water, at No More Than Half Density,[a] Reflected by an Effectively Infinite Thickness of Water

	Subcritical Limit			
Compound	Mass of Fissile Nuclide (kg)	Mass of Oxide[b] (kg)	Cylinder Diameter (cm)	Slab Thickness (cm)
$^{233}UO_2$	23.4	27.0	11.9	1.6
$^{233}U_3O_8$	30.5	36.6	14.8	2.2
$^{233}UO_3$	34.7	42.4	16.3	2.6
$^{235}UO_2$	88.0	102.0	20.4	5.8
$^{235}U_3O_8$	122.0	146.0	26.0	8.0
$^{235}UO_3$	142.0	174.0	28.8	9.3
$^{239}PuO_2$	27.0	30.0	12.6	2.8

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[a] These are half the maximum bulk densities of Table 4.

[b] These values include the mass of any associated water up to the limiting value of 1.5 wt%.

C. Fissile Density Dependent Limits

1. Solutions and Metal-Water Mixtures

The overall acceptable limit for a solution parameter applies to the minimum value over the solution density range. If the solution density range is controlled to exclude the value for which the parameter is minimum, an increased limit may be valid. From Figures 2-13, acceptable limits for restricted density ranges may be established. Curves of critical and subcritical values as functions of the density of ^{233}U, ^{235}U or ^{239}Pu are given for

- spherical mass in Figures 2, 6, and 10;
- spherical volume in Figures 3, 7, and 11;
- infinite cylinder diameter in Figures 4, 8, and 12;
- infinite slab thickness in Figures 5, 9, and 13.

The curves labeled k=1.0 (to avoid cluttering figures, k appears instead of k_{eff}) represent calculated critical conditions for water reflected metal-water mixtures. The Figures also display experimental results for solutions. Critical parameters for experimental results are consistently larger than the corresponding values for the metal-water mixtures. The Appendix describes the Monte Carlo computational techniques that were used, and it documents the calculational method used to obtain the results. Curves at $k = 0.9$ and $k = 0.8$, calculated by the same means are included in each figure to show adjustments that would correspond to desired margins in Δk.

By means of the curves for different values of k, subcritical margins that allow for assumed contingencies can be selected. These margins, of course, go beyond the computational bias incorporated in the subcritical limits that have been discussed. If, for example, the dominant contingency could increase k to 1.02, a point on or near the curve for $k = 0.9$ may be acceptably conservative.

The 15-cm-thick (effectively infinite) water reflector used for the curves is the most effective reflector material commonly encountered outside reactors. As stated earlier, some reflector materials, when closely fitting and of sufficient thicknesses, are more effective than ordinary water. Thicknesses of these materials equivalent to 15-cm-thick water about fissile metal are listed in Tables 6 and 7. Unlike water, these materials, including closely fitting concrete, are rarely encountered, never accidentally, and call for special evaluation if incorporated into a design. Water is indeed one of the most effective reflectors in thicknesses of 7.5 cm or less. In general, like water, the effectiveness of hydrocarbons as reflectors saturates at thicknesses of about 10 cm.[81]

Table 6

Thickness of Reflectors Required for the Criticality of a 21.24-kg ^{235}U Metal Sphere at 18.81 g/cm^3

Reflector Material	Density (g/cm^3)	Thickness (cm)
Water	1.00	15.0
Iron	7.86	17.6
Heavy Water	1.10	7.2
Carbon	1.90	8.4
Beryllium	1.80	3.8
Plexiglas[a]	1.20	5.1

Table 7

Thickness of Reflectors Required for the Criticality of a 5.32-kg ^{239}Pu Metal Sphere at 19.85 g/cm^3

Reflector Material	Density (g/cm^3)	Thickness (cm)
Water	1.00	15.0
Iron	7.86	16.2
Heavy Water	1.10	7.6
Carbon	1.90	8.1
Beryllium	1.80	3.2
Plexiglas[a]	1.20	6.1

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[a] Methacrylate plastic, $C_5H_8O_2$.

The upper dashed curves of Figures 2 to 13 display critical values, provided the only reflection is by a thin aluminum or steel container. This reflection condition is seldom encountered in processing plants because it is nearly always augmented by reflection from external objects (incidental reflection).* The purpose of these curves is to provide a reference for showing the effect of assuming full water reflection when it does not actually occur. It may be noted that this effect corresponds roughly to a shift in k of 0.1.

The metal-water curves, of course, apply conservatively to solutions. An appropriate adjustment between curves at k = 1.0 and k = 0.9 can establish subcritical limits. The curves at k = 0.9 and k = 0.8 can be used to adjust safety margins for operating contingencies to which general subcritical limits would not apply. Because of this flexibility, the figures do not include specific values of subcritical limits similar to the single-parameter limits of Section A of this chapter. The following may be pertinent if there should be interest in curves that appear in *Nuclear Safety Guide, Revision 2* for a 2.5-cm thick water reflector. It is seen that the subcritical limits for this reflector very nearly coincide with the k = 1 curves with thick water reflection in each figure.

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*Except as a limit, a minimally reflected infinite critical slab (Figs. 5, 9, and 13) would be fictional. If truly of infinite extent, it could not escape full reflection.

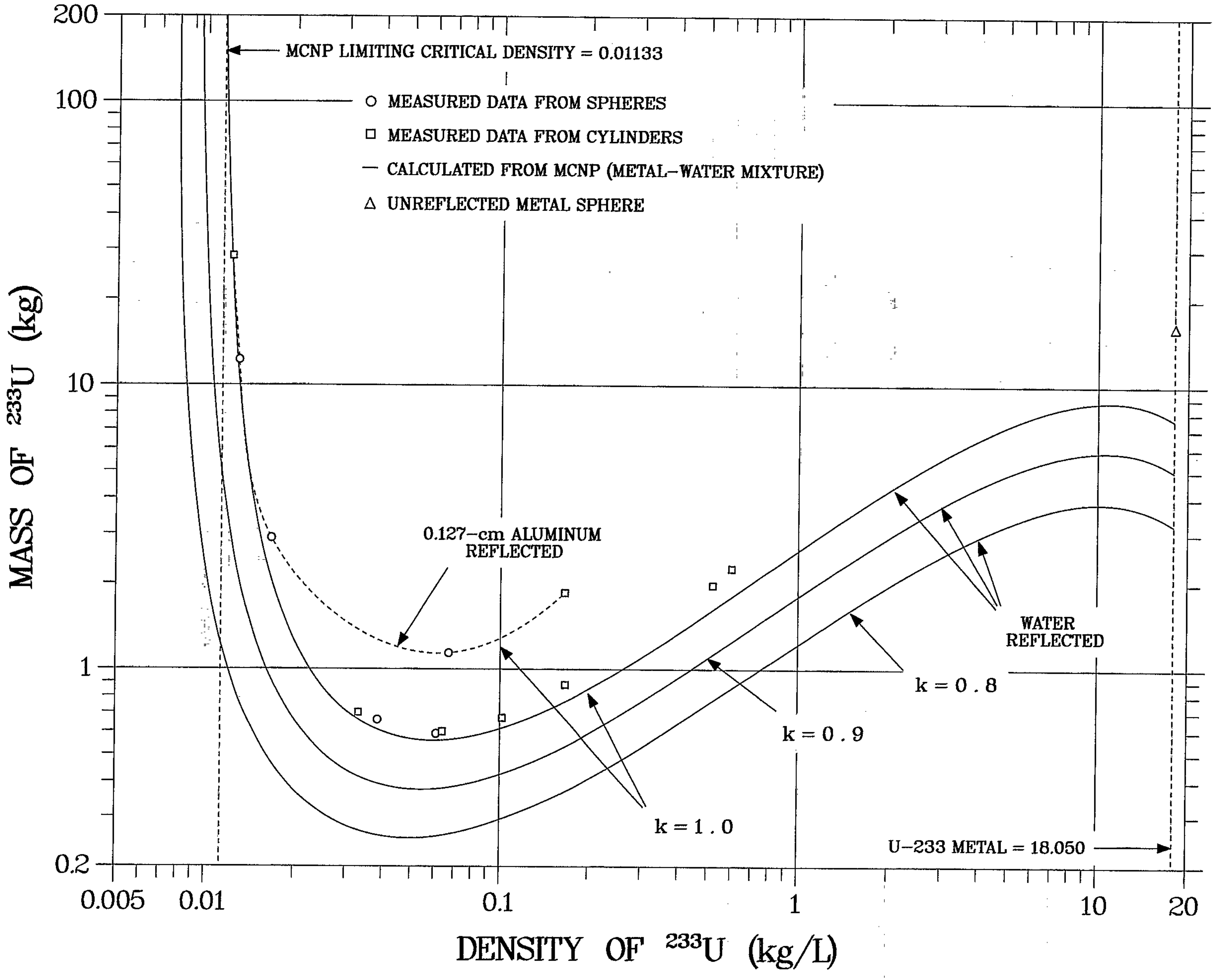

Fig. 2. Masses of spheres of homogeneous water-moderated ^{233}U as functions of ^{233}U density.

Figure 2

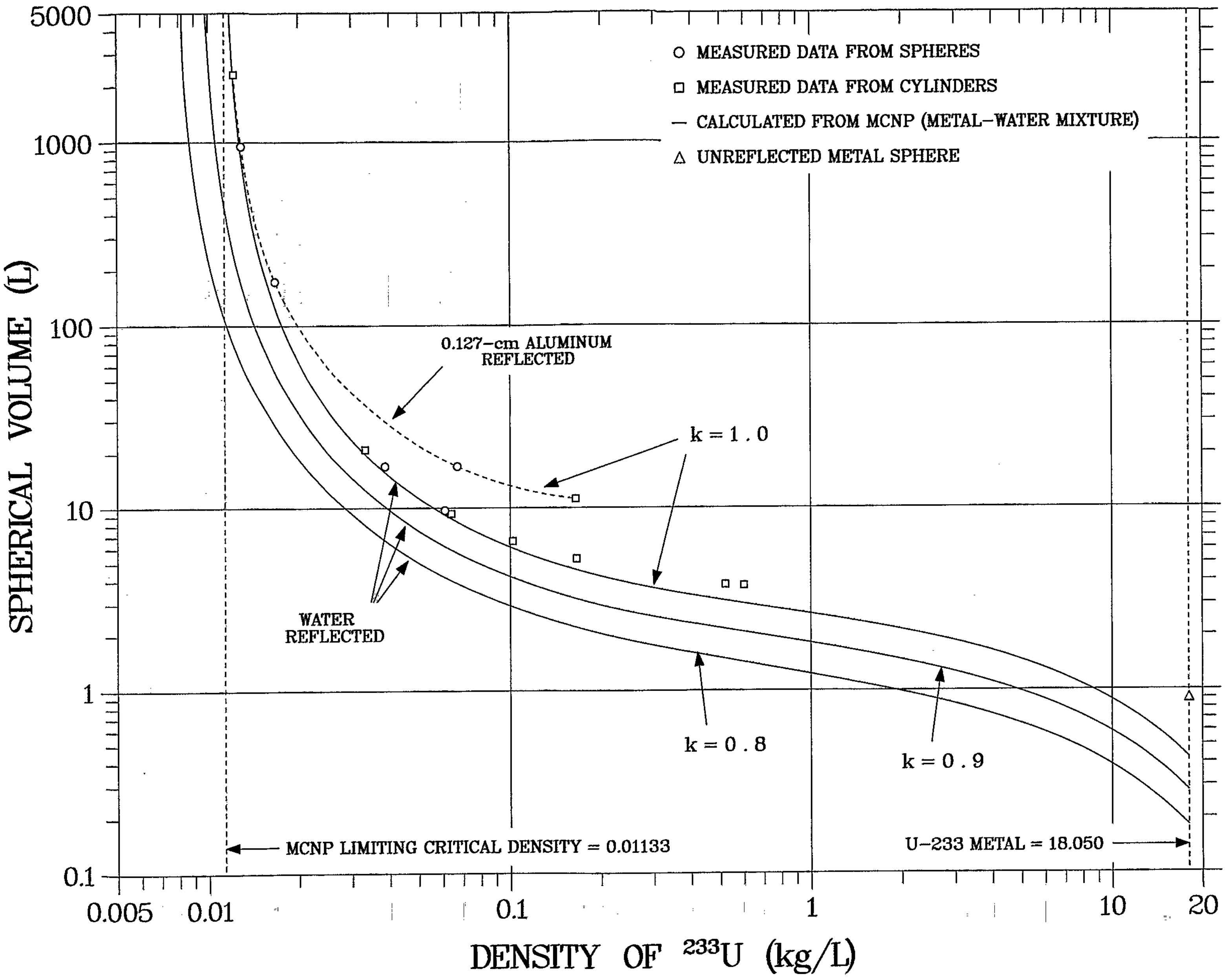

Fig. 3. Volumes of spheres of homogeneous water-moderated ^{233}U as functions of ^{233}U density.

Figure 3

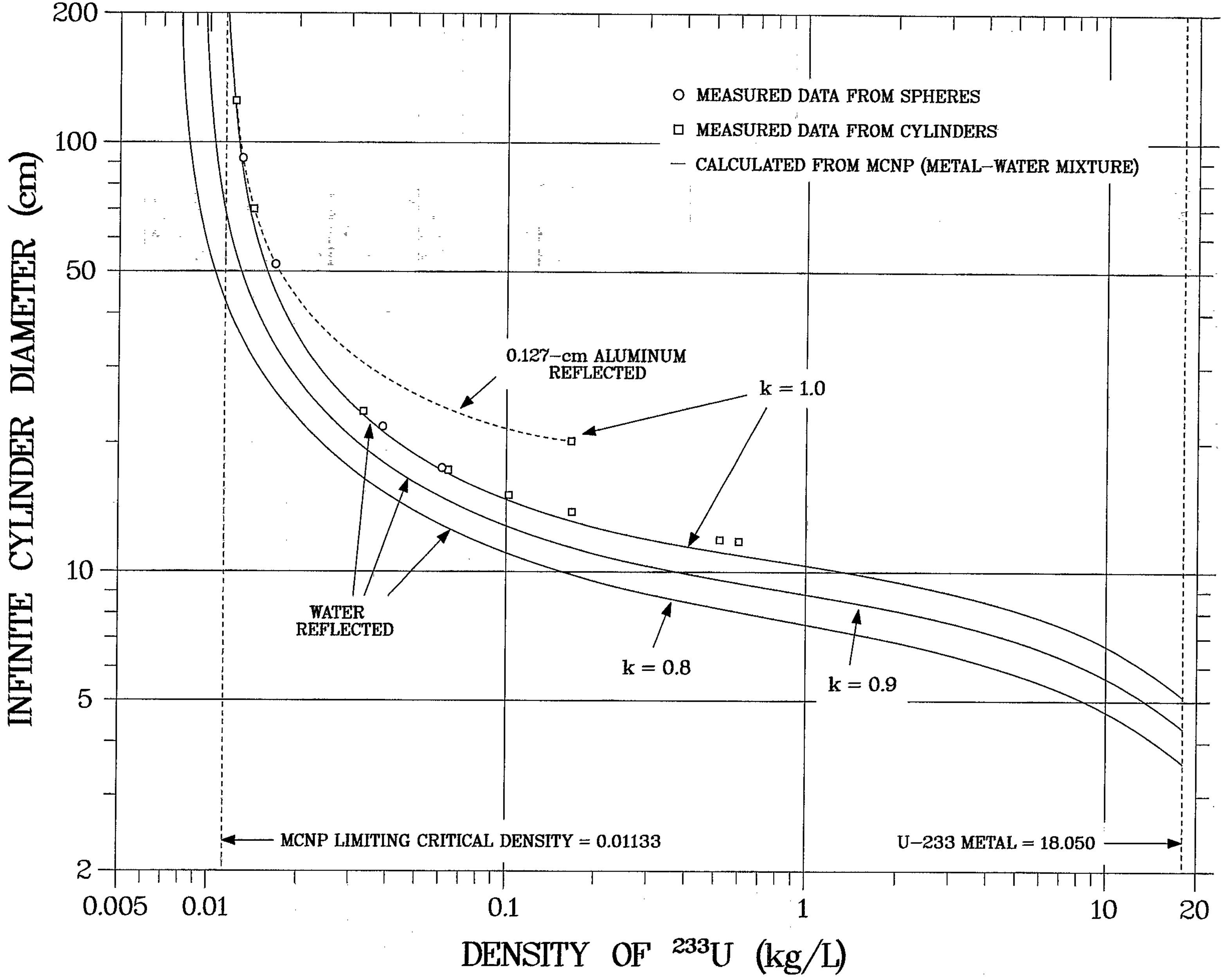

Fig. 4. Diameters of infinite cylinders of homogeneous water-moderated ^{233}U as functions of ^{233}U density.

Figure 4

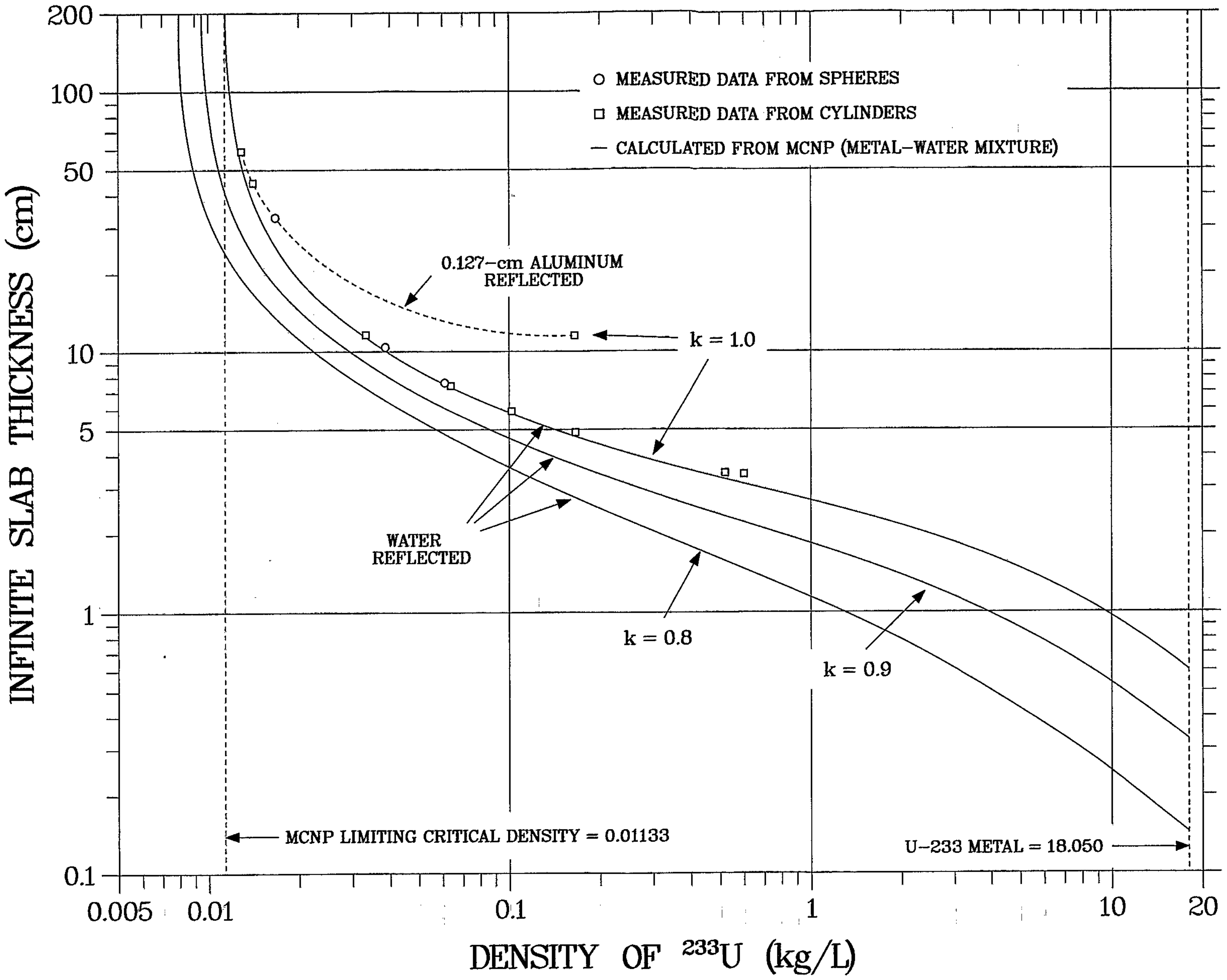

Fig. 5. Thicknesses of infinite slabs of homogeneous water-moderated ^{233}U as functions of ^{233}U density.

Figure 5

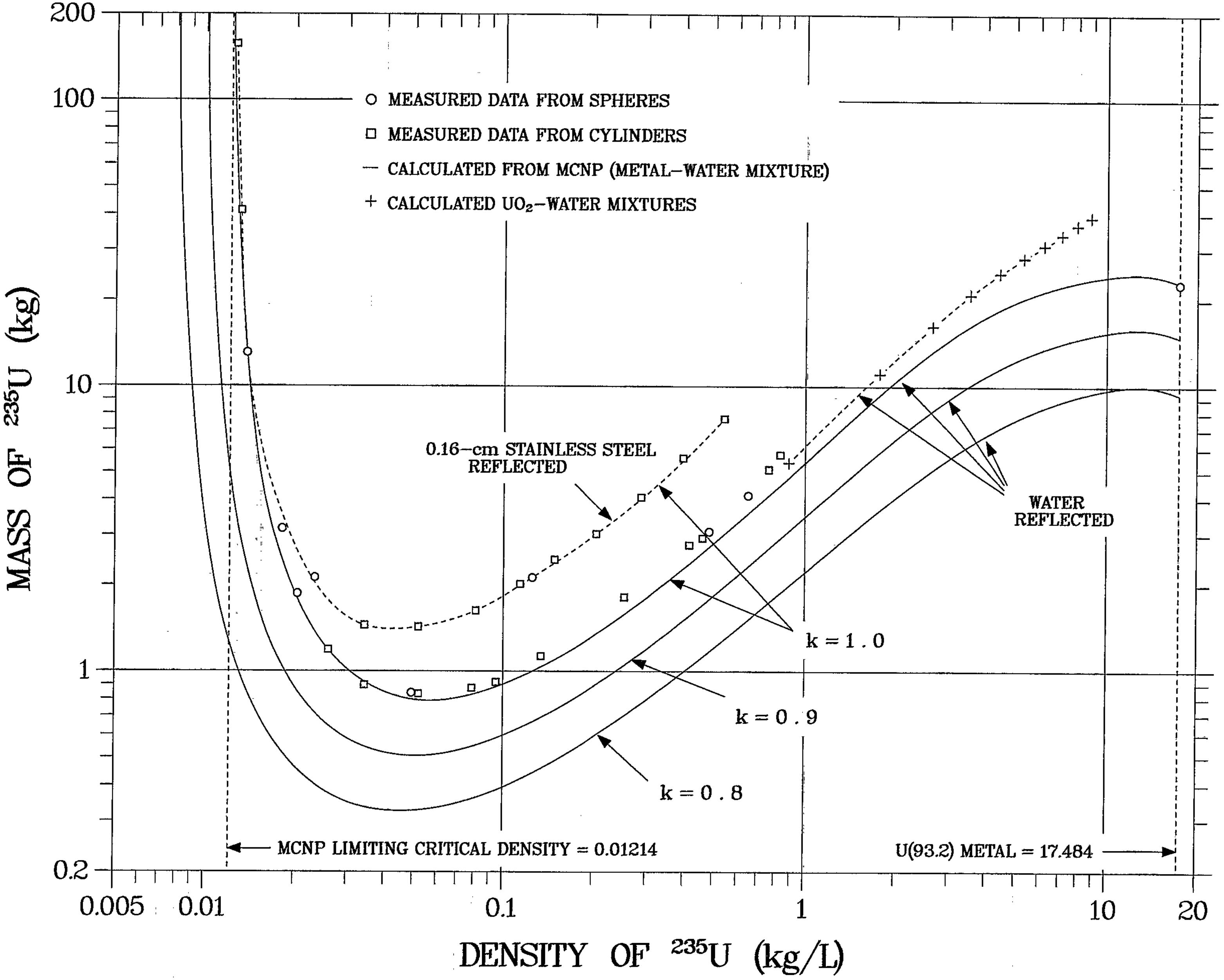

Figure 6

Fig. 6. Masses of spheres of homogeneous water-moderated U(93.2) as functions of ^{235}U density.

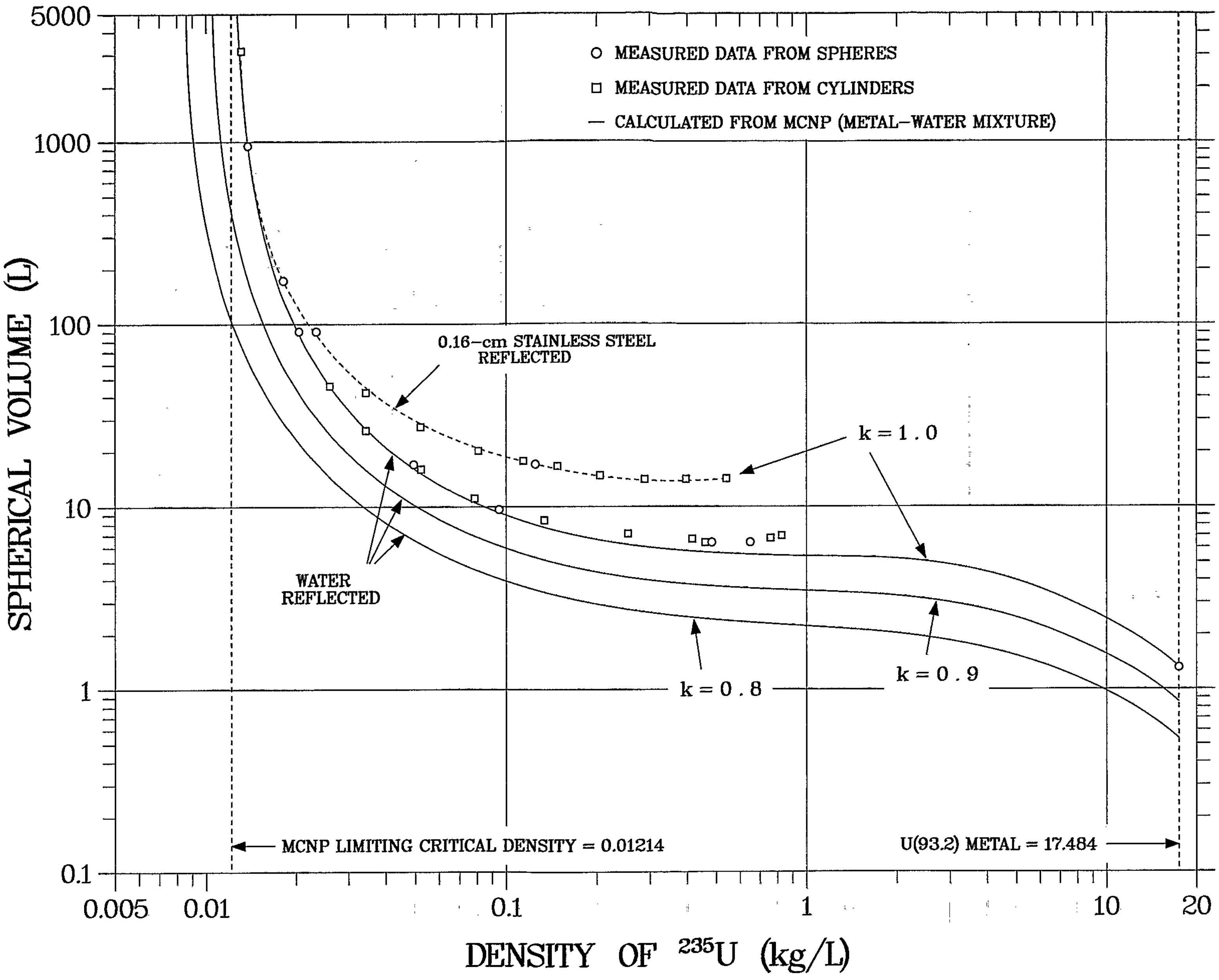

Fig. 7. Volumes of spheres of homogeneous water-moderated U(93.2) as functions of ^{235}U density.

Figure 7

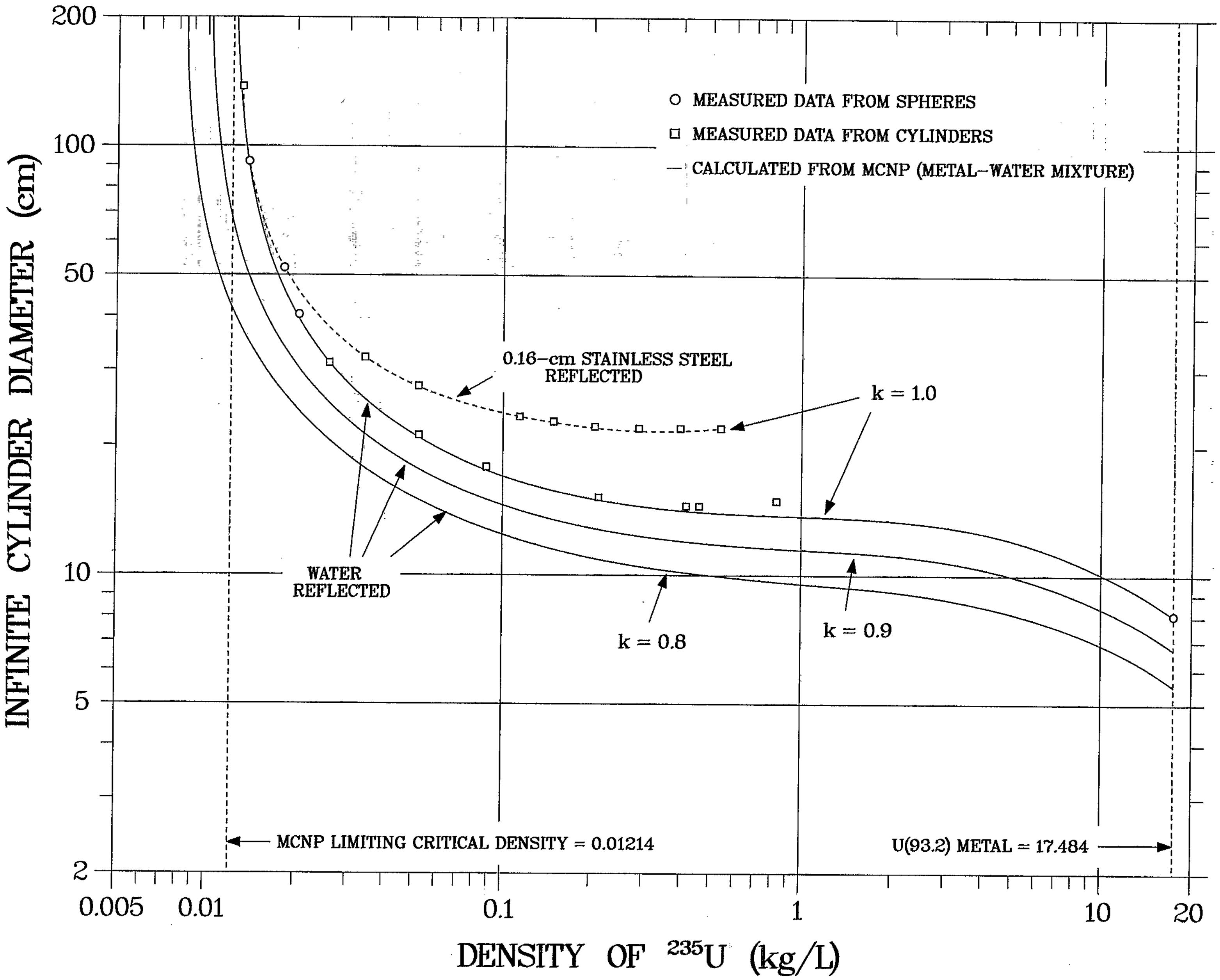

Figure 8

Fig. 8. Diameters of infinite cylinders of homogeneous water-moderated U(93.2) as functions of ^{235}U density.

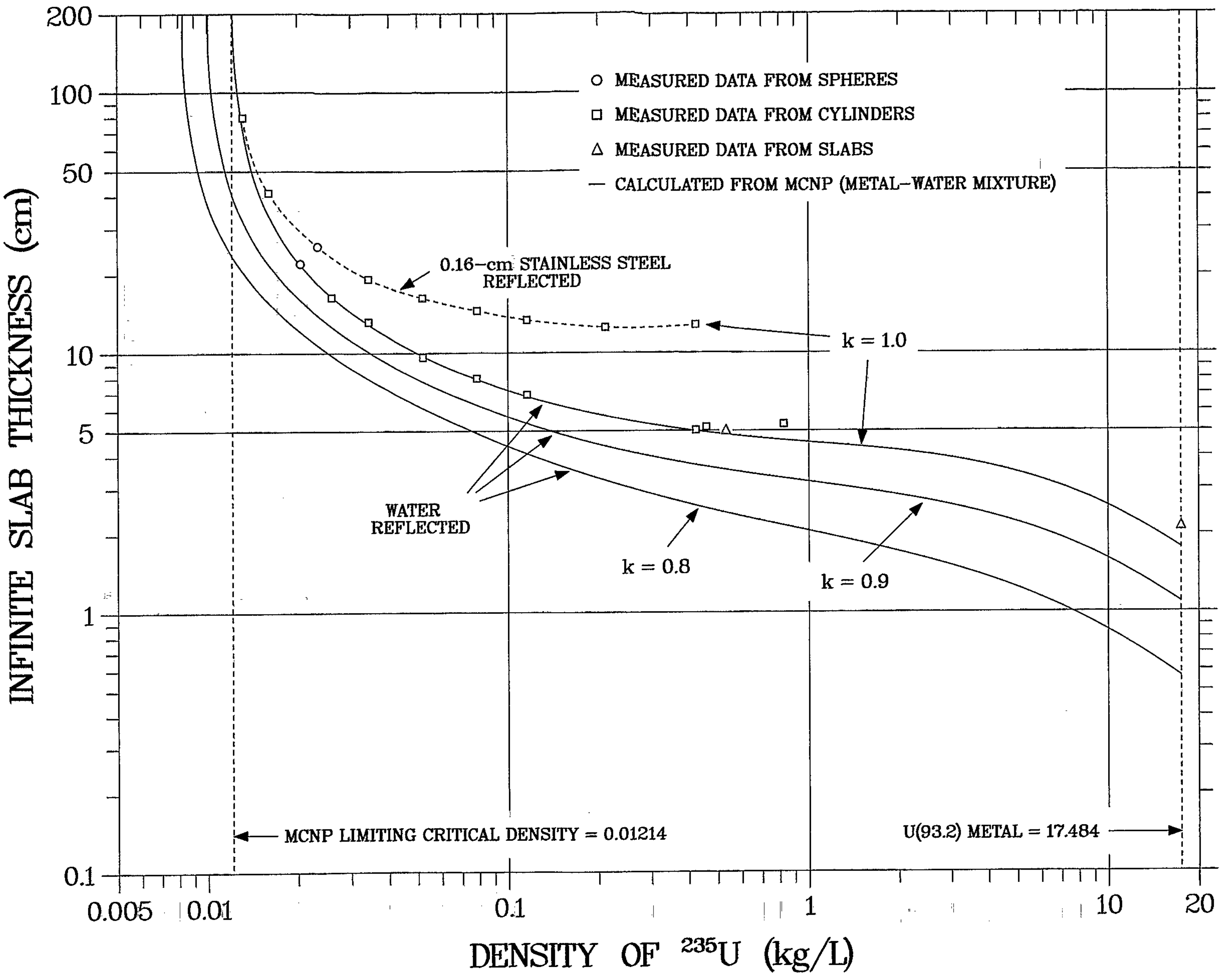

Figure 9

Fig. 9. Thicknesses of infinite slabs of homogeneous water-moderated U(93.2) as functions of ^{235}U density.

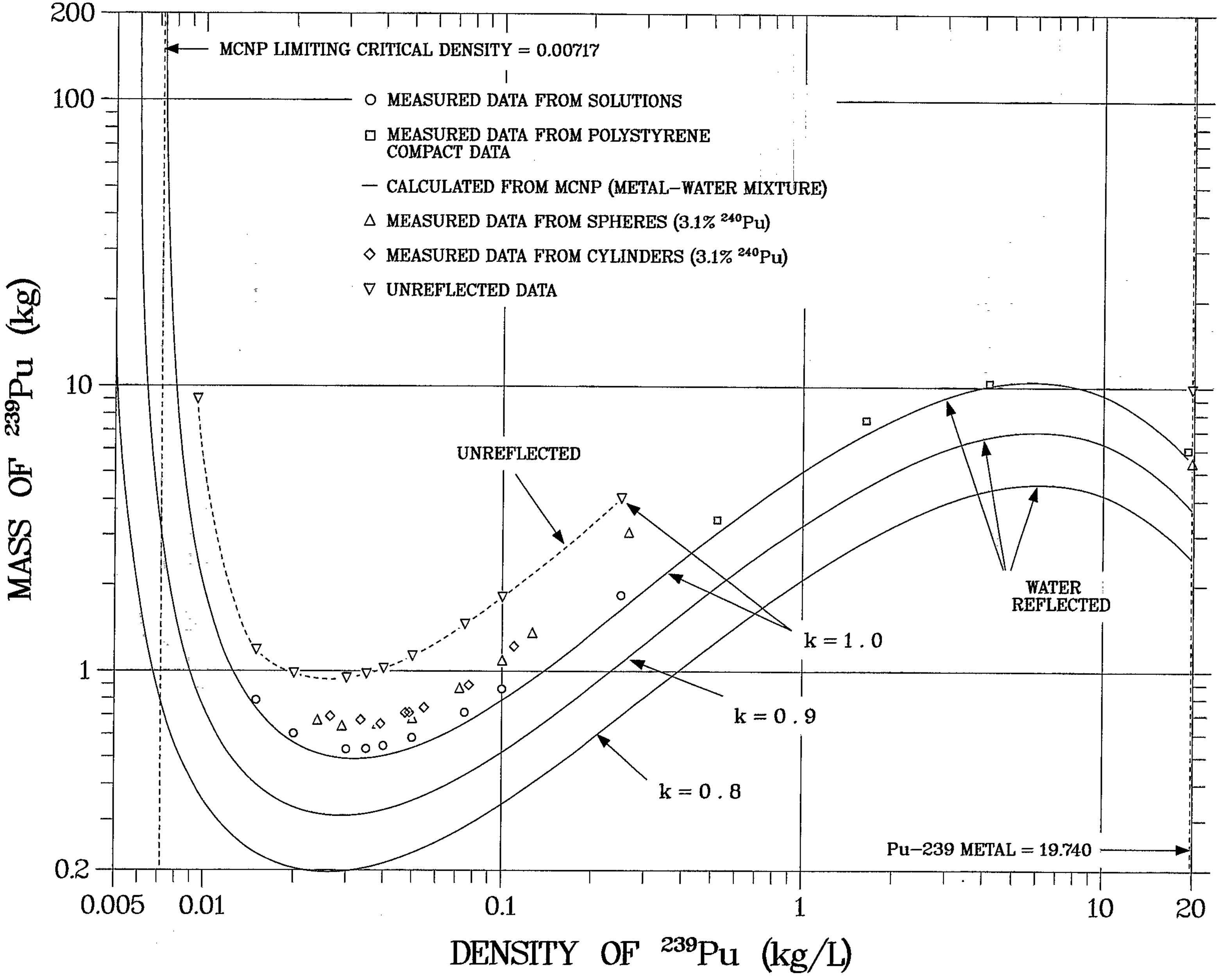

Fig. 10. Masses of spheres of homogeneous water-moderated ^{239}Pu as functions of ^{239}Pu density.

Figure 10

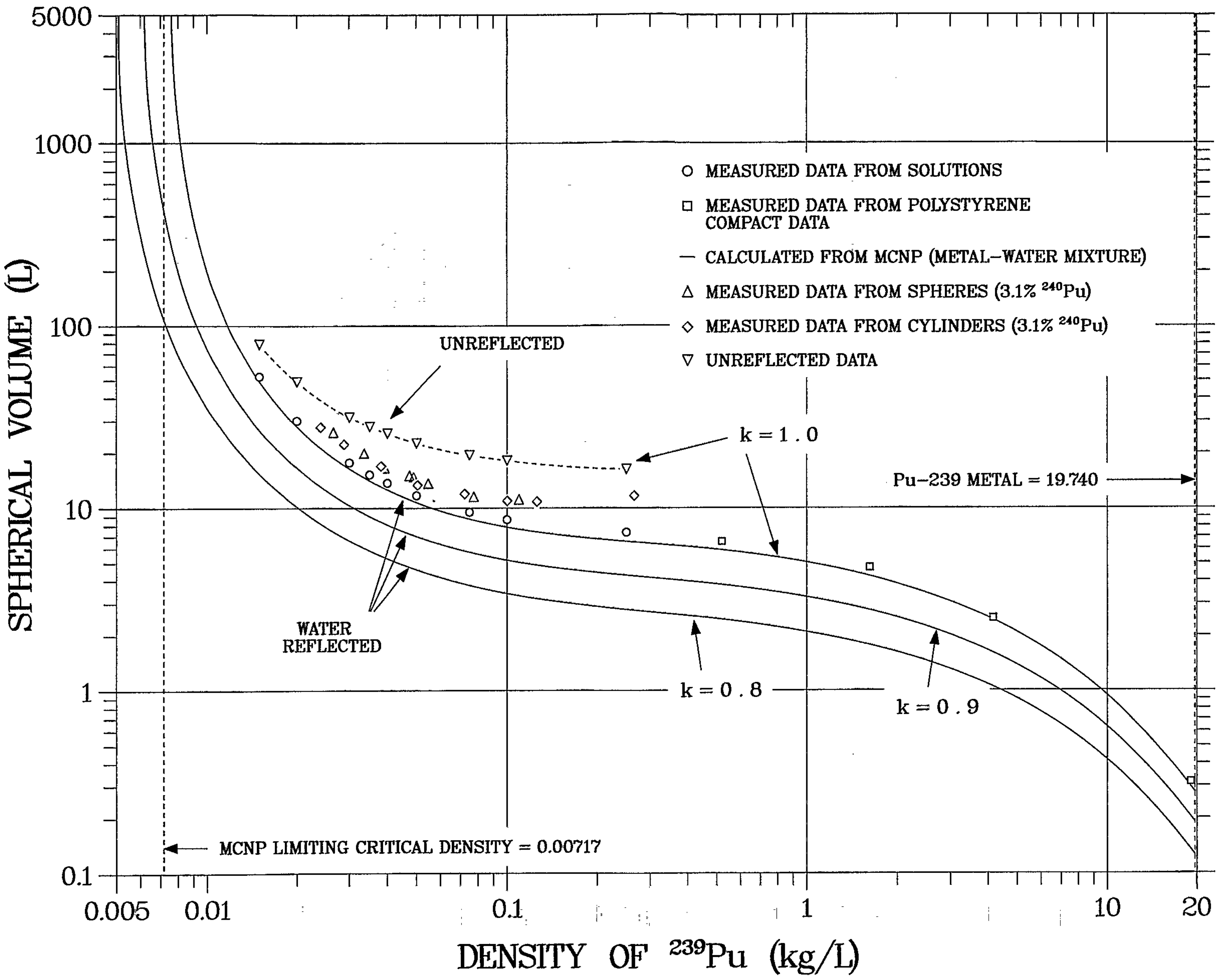

Fig. 11. Volumes of spheres of homogeneous water-moderated ^{239}Pu as functions of ^{239}Pu density.

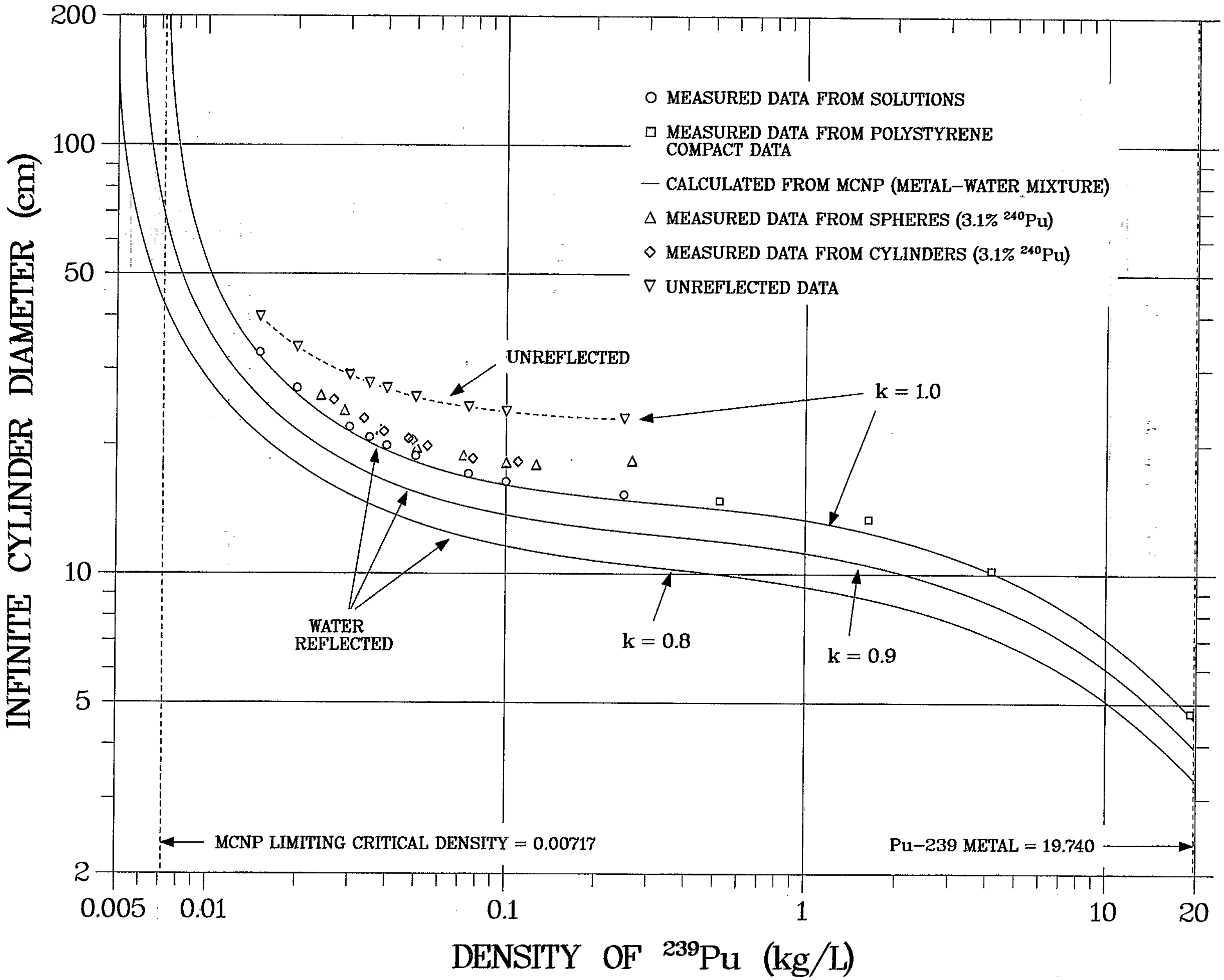

Fig. 12. Diameters of infinite cylinders of homogeneous water-moderated ^{239}Pu as functions of ^{239}Pu density.

Figure 12

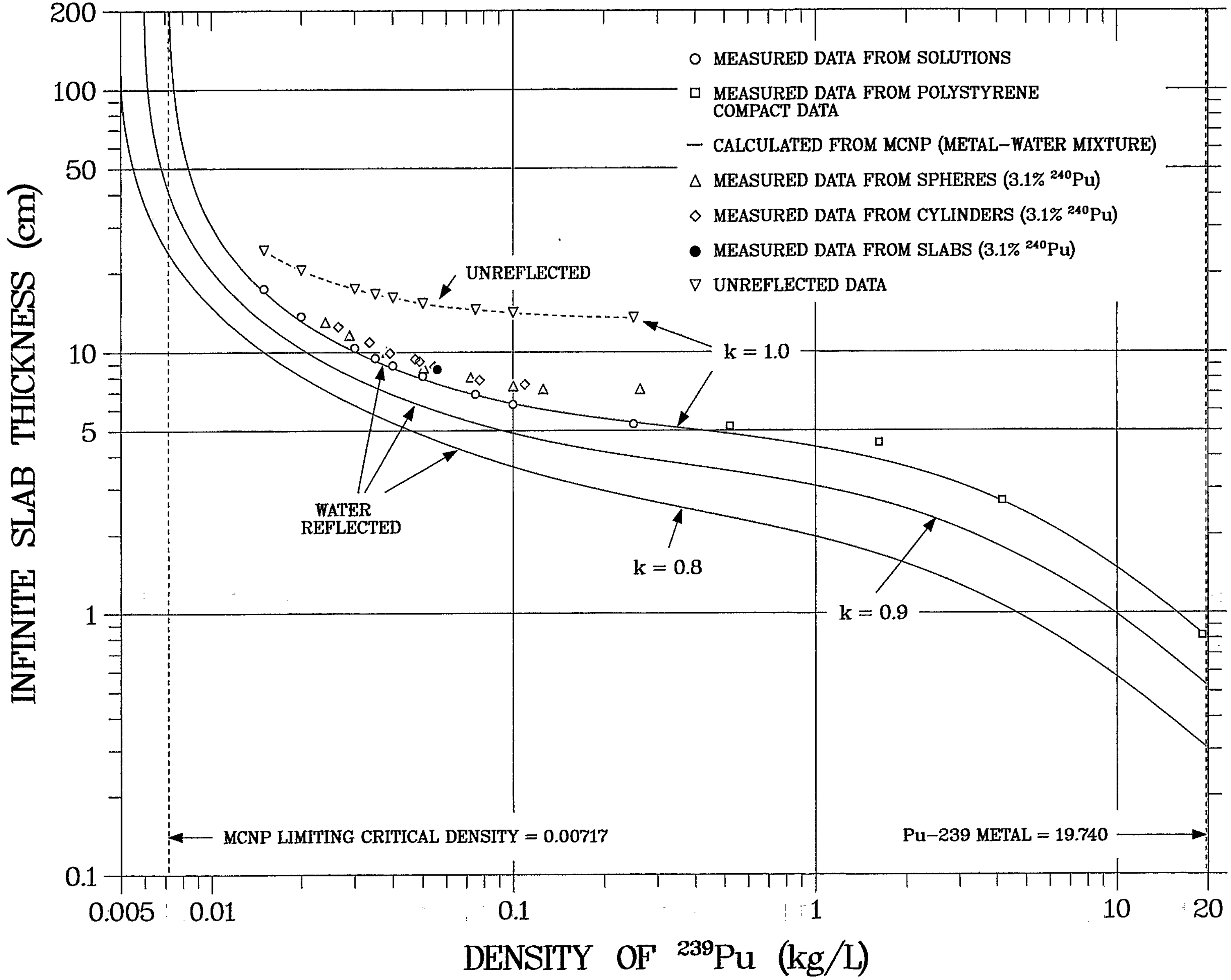

Fig. 13. Thicknesses of infinite slabs of homogeneous water-moderated ^{239}Pu as functions of ^{239}Pu density.

Figure 13

2. Low-Enriched Uranium

Application of the ^{235}U limits of Table 1 and Figures 6 through 9 to uranium at low enrichments would result in safe but very uneconomic criticality safety criteria. Strict administrative controls to establish the enrichment and to maintain material identification are mandatory in order to take advantage of realistic limits for uranium of low enrichment. It should be noted that criticality is not possible for uranium metal containing less than approximately 5 wt% ^{235}U.[82–83]

Table 8 gives more realistic subcritical limits for uranium enriched to 10 wt% or less ^{235}U.[4] The limits in Table 8 with footnote "a" must be decreased if the uranium density for a saturated solution can be exceeded, as by precipitation.

The subcritical masses presented in Table 8 for solutions of low-enriched uranium compounds are not applicable when applied to lattices such as exist in power reactors. This is a consequence of the absorbing characteristics of ^{238}U for neutrons having energies of a few electron volts, a property called resonance absorption.[23] When the uranium is latticed, as in a reactor, there is a greater probability of immediate neutron energy degradation from the high energy at which neutrons are produced by fission to less than that at which ^{238}U is strongly absorbing. These neutrons "escape" the ^{238}U resonance absorption and the probability of the escape is a measurable and calculable property of such lattices. The maximum ^{235}U enrichment of the uranium at which latticing can reduce the critical mass is estimated to be about 6 wt% ^{235}U. As noted above, the critical mass of uranium below this enrichment can be lower for a heterogeneous system than for a homogeneous system. Therefore, subcritical limits of a lattice are smaller than for homogeneous uranium of the same enrichment. If the particles constituting a mixture are uniformly distributed and are larger than 127 microns (i.e., not capable of being passed through a 120-mesh screen), the mixture should be considered as heterogeneous unless demonstrated otherwise.[84–85]

Subcritical limits on masses and dimensions of lattices in water of $U(\leq 5)$* metal or oxide rods of any diameter or lattice spacing surrounded by a thick water reflector have been calculated.[86–87] These limits can be applied to other heterogeneous arrangements of uranium in water. Limits derived for rods of optimum diameter latticed at the most reactive spacing can be applied conservatively to other sizes, shapes, or distributions. Experiments[88–89] indicate that a random arrangement is less reactive than is a uniform array of rods at optimum spacing; the actual spacings in the random array may be distributed about the most reactive spacing.

Subcritical limits for uranium and uranium oxide in heterogeneous mixtures[86–87] are given in Table 9. The limits are applicable regardless of the size or shape of the metal or oxide pieces; they apply only if the environment does not return neutrons more effectively than water (see Table 6). For comparison with lattices, limits for homogeneous uranium oxide water mixtures from the reference are included in Table 9.

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*Read as uranium enriched to less than or equal to 5 wt% in ^{235}U.

Table 8

Subcritical Limits for Solutions of Low-Enriched Uranium, Thick Water Reflector

		Subcritical Limit				
Solution	Enrichment (wt% ^{235}U)	Mass (kg ^{235}U)	Volume (L)	Cylinder Diameter (cm)	Slab Thickness (cm)	Density of U (g/L)
UO_2F_2	1.45	–	–	–	–	1190.0[a]
	2.0	8.00[b]	340.0[b]	63.0[b]	36.5[b]	770.0[c]
	3.0	2.75	77.0[b]	37.4[b]	20.0[b]	470.0[c]
	4.0	1.98	42.7[b]	30.2[b]	15.1[b]	335.0[c]
	5.0	1.64	30.6	26.6	12.6	261.0[c]
	10.0	1.07	14.8	20.1	8.3	123.0[c]
$UO_2(NO_3)_2$	2.88	–	–	–	–	594.9[a]
	4.0	6.50[b]	273.0[b]	58.6[b]	33.7[b]	375.0[c]
	5.0	3.30	111.0[b]	42.7[b]	23.4[b]	283.0[c]
	10.0	1.47	26.7	25.2	11.9	128.0[c]

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[a] Density below which criticality is unattainable.

[b] This value is for a saturated solution; the minimum occurs at a greater density of the salt.

[c] Saturated solution, assuming that a molarity of 5 for UO_2F_2 and 2.5 for $UO_2(NO_3)_2$ is not exceeded; at larger molarities, the above limits are not valid.

Table 9

Subcritical Limits for Low-Enriched Uranium as Oxide-Water and Metal-Water Lattices, Thick Water Reflector

		Subcritical Limit			
Material	Enrichment (wt% ^{235}U	Mass (kg ^{235}U)	Volume of Lattice (L)	Cylinder Diameter (cm)	Slab Thickness (cm)
Latticed Oxide	1.0	19.50	480.0	72.0	41.0
	2.0	3.37	60.3	34.0	17.4
	3.0	2.12	33.4	27.4	13.0
	4.0	1.59	24.0	24.5	11.2
	5.0	1.36	19.5	22.3	9.8
Latticed Metal	1.0	13.10	234.0	55.5	31.6
	2.0	2.94	43.0	30.1	15.0
	3.0	2.03	25.8	24.9	11.5
	4.0	1.54	18.3	21.7	9.8
	5.0	1.29	14.5	20.0	8.2
Homogeneous Oxide	2.0	5.55	109.0	41.9	22.4
	3.0	2.84	49.4	31.4	15.5
	4.0	2.00	33.5	26.9	12.6
	5.0	1.58	25.2	24.6	11.1

It is unclear from available data whether natural uranium metal rods can become critical in water if they are of the appropriate diameter and spacing. The minimum ^{235}U enrichment of critical homogeneous aqueous mixtures is about 1 wt%.* Table 10 gives the ^{235}U enrichments at or below which several compounds will be subcritical as homogeneous aqueous mixtures or solution.

Table 10

Subcritical ^{235}U Enrichment Limits for Uranium Mixed Homogeneously with Water

Compound	Subcritical Limit (wt% ^{235}U)
Uranium metal	0.93
UO_2, UO_3, U_3O_8, or UO_2F_2	0.96
$UO_2(NO_3)_2$	1.96

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*See Fig. 22 of Ref. 11.