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**CORRELATIONS OF EXPERIMENTAL AND
THEORETICAL CRITICAL DATA
COMPARATIVE RELIABILITY, SAFETY FACTORS
FOR CRITICALITY CONTROL**



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CORRELATIONS OF EXPERIMENTAL AND
THEORETICAL CRITICAL DATA*
COMPARATIVE RELIABILITY, SAFETY FACTORS
FOR CRITICALITY CONTROL

by

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drawing freely from theoretical work of

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PREFACE

This report is a somewhat updated version of a paper that was prepared for the ENEA Symposium on Criticality Control (Karlsruhe, May 2-5, 1961). It may be fortunate that the topic was assigned -- otherwise, it is probable that all with appropriate interest, like the hapless author, would have continued waiting for a more venturesome individual to take the required liberties with work of others. As it is, the resulting intercomparisons of computed and experimental critical data form a logical extension of the purely experimental compilation of LAMS-2415, CRITICAL DATA FOR NUCLEAR SAFETY GUIDANCE.

CORRELATIONS OF EXPERIMENTAL AND THEORETICAL CRITICAL DATA

For the experimental part, this discussion will concentrate on data that are susceptible to generalization. Practically, the scope will be limited to fissile systems that may be approximated by simple descriptions - - elementary geometry and generally homogeneous fuel regions. This type of information has been termed "basic" nuclear safety data. Thus, we ignore the specialized experiment such as that intended to demonstrate safe operating conditions for a specific processing unit.

Similarly, we will not consider specialized calculations such as those designed to give effects of minor changes in a given type of heterogeneous fissile assembly. Rather, most attention will be given to computation which hopefully may substitute for basic experimental data. Before considering this class of calculation, however, let us discuss a somewhat less demanding application of theory.

The Adjustment of Empirical Data

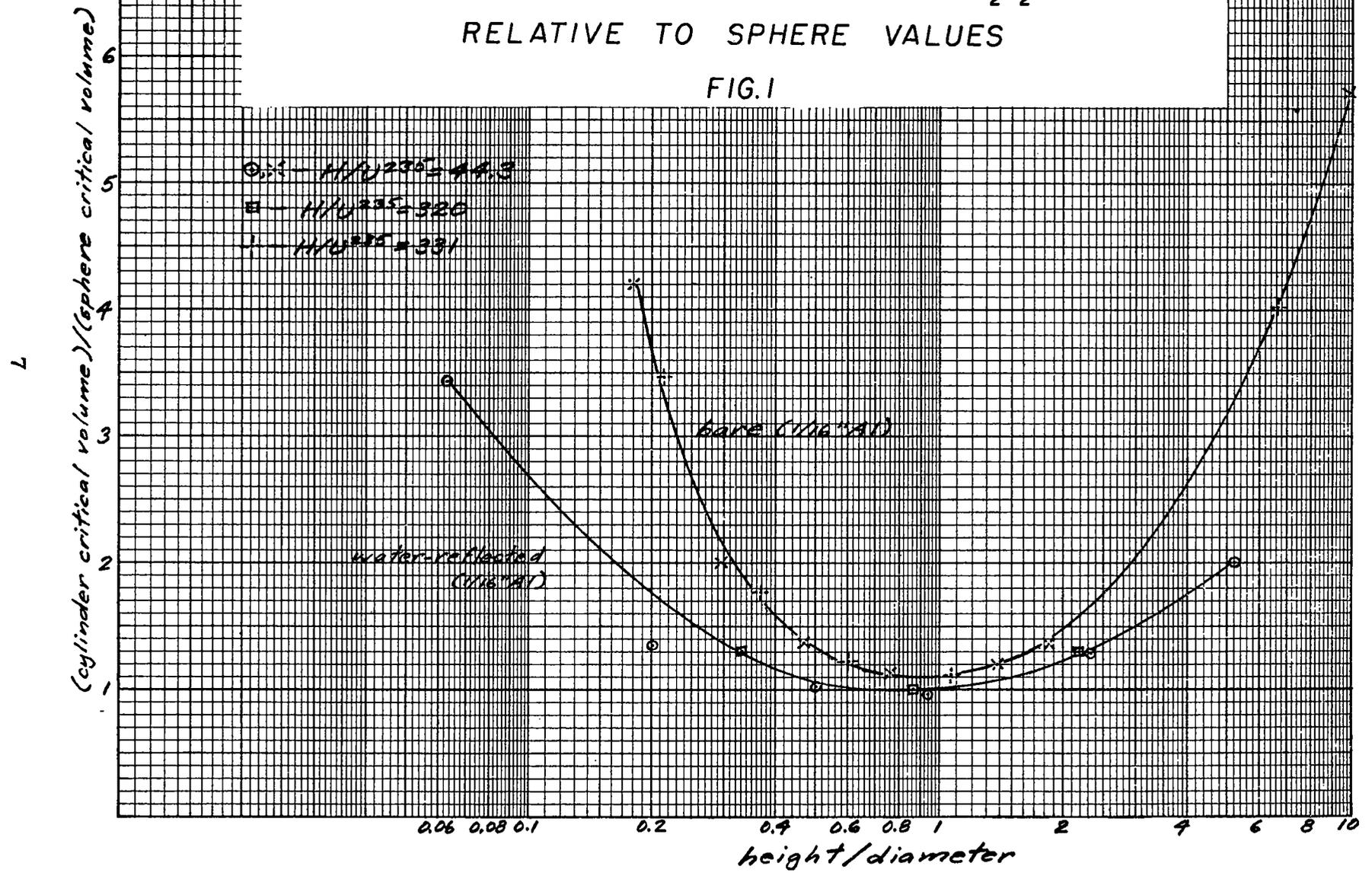
As experimental data seldom apply to ideal systems which may be described in similar terms, empirical

correlation of the data generally requires conversion to uniform conditions. This process, in turn, makes use of data correlations which may be strictly empirical or guided by theoretical models. (It is apparent that successive approximations may be involved if there is a relationship between input and output correlations.) As for example, corrections on critical mass are frequently within 10%, the ultimate in precision is often not required of the model used for correction. Figures 1 and 2 are examples of two empirical representations of the same data, the first without regard to a theoretical model, and the other in a form suggested by such a model (1). In each case the data are critical volumes of families of cylinders of $\text{UO}_2\text{F}_2 - \text{H}_2\text{O}$ solutions in which the uranium is enriched to about 93 w/o U^{235} [we shall symbolize this uranium $\text{U}(93)$].

Figure 1 shows, for cylinders of various height/diameter values, ratios of critical volumes to the spherical value for the same solution. It may be noted that there is no obvious distinction between curves for solutions at two widely different uranium concentrations. In Fig. 2 are represented effective extrapolation distances of critical cylinders, δ_c , which satisfy the buckling relationship

CRITICAL VOLUMES OF CYLINDERS OF $U(93)O_2F_2$ SOLUTION
RELATIVE TO SPHERE VALUES

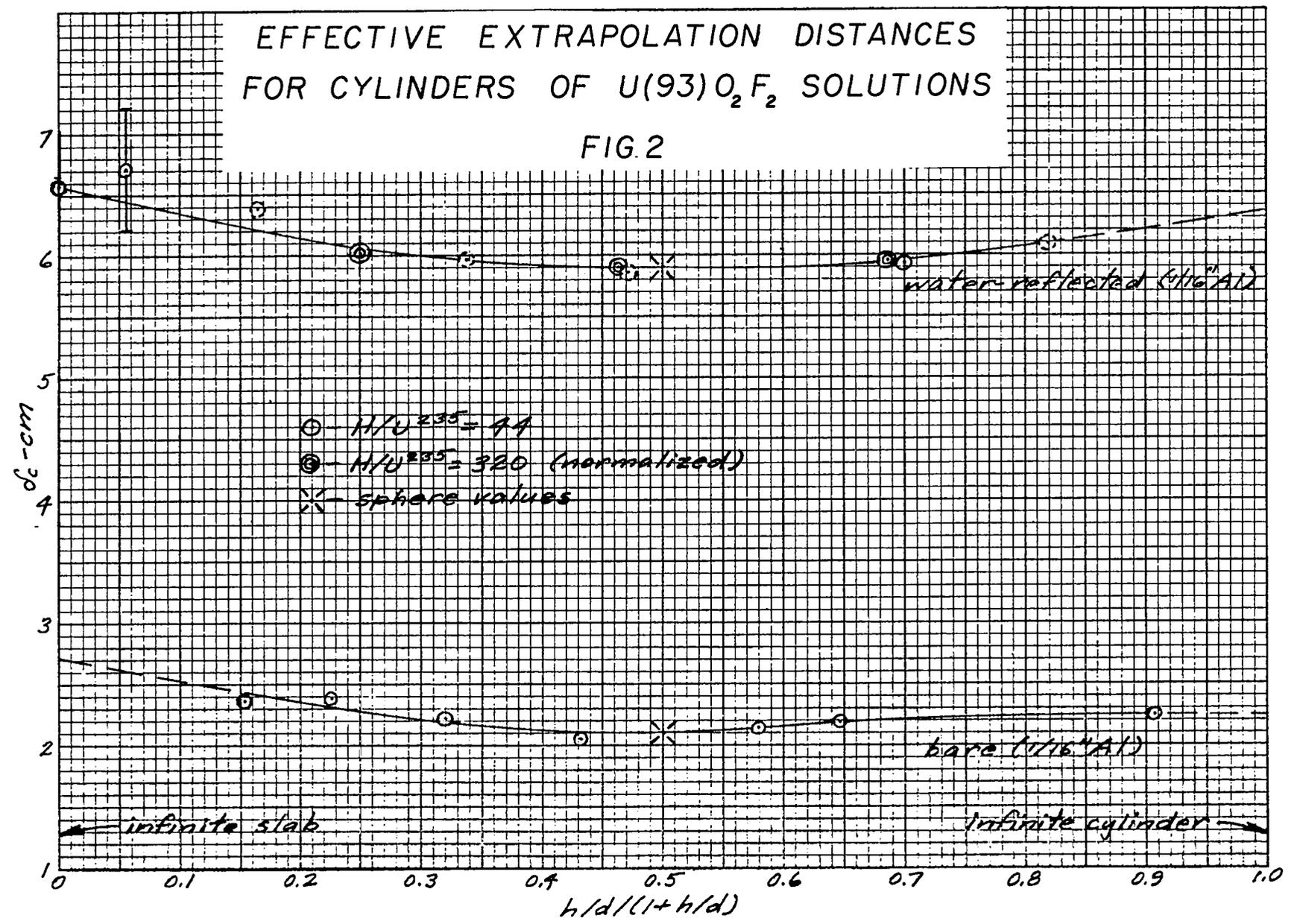
FIG. 1



EFFECTIVE EXTRAPOLATION DISTANCES
FOR CYLINDERS OF U(93)O₂F₂ SOLUTIONS

FIG. 2

8



$$\left(\frac{2.405}{\frac{d}{2} + \delta_c}\right)^2 + \left(\frac{\pi}{h + 2\delta_c}\right)^2 = \left(\frac{\pi}{r_s + \delta_s}\right)^2 ,$$

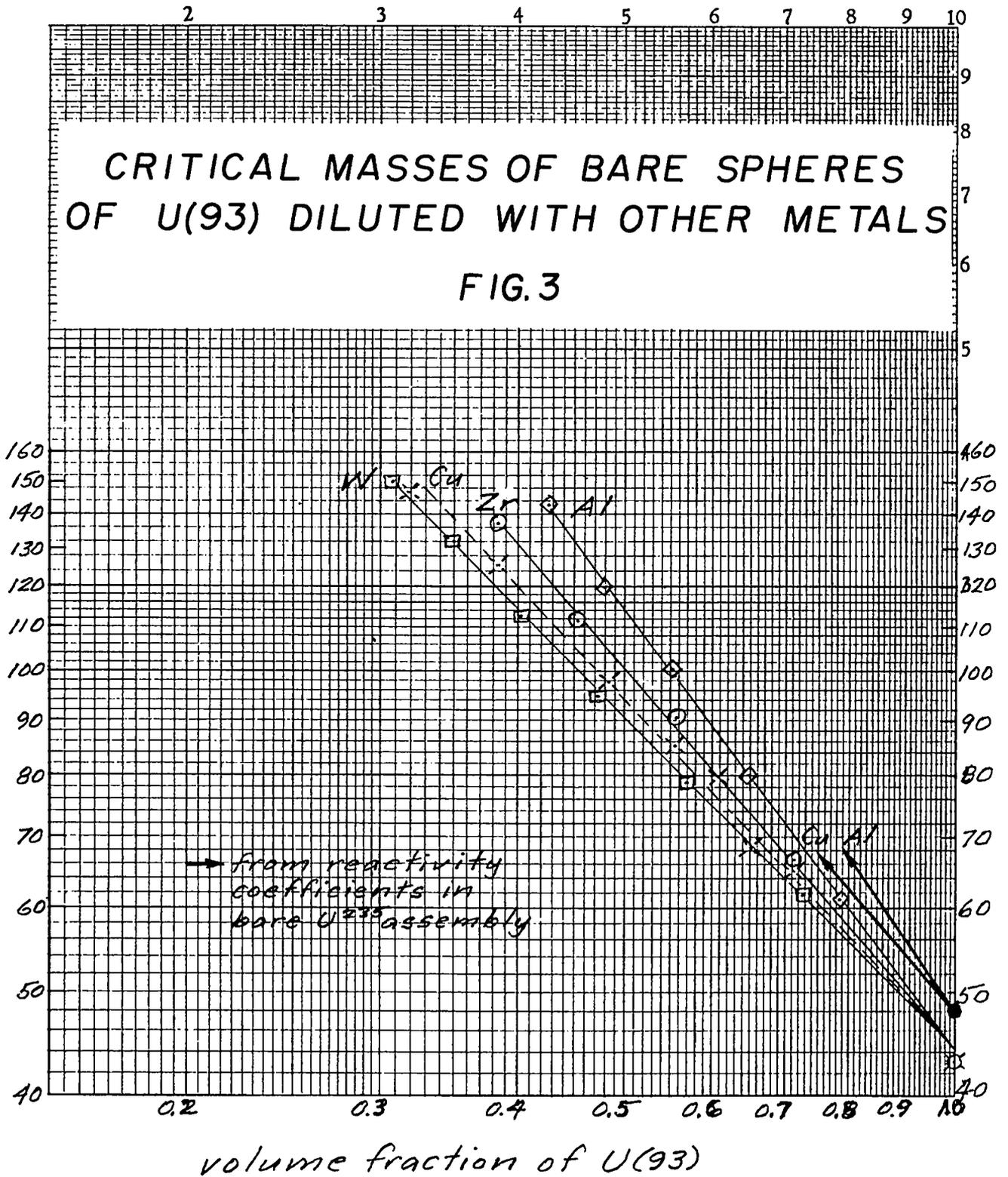
where d and h are diameter and height of the critical cylinder and r_s is the radius of the corresponding sphere. The sphere extrapolation distance δ_s is the sum of an experimental reflector savings and an assumed or computed bare-sphere extrapolation distance. (The latter value, of course, remains an inherent part of the correlation.) If simple theory applied precisely, δ_c would be independent of h/d . As it is, deviations from a constant value are sufficiently small that there may be some confidence in extrapolations to extreme values of h/d . Such extrapolations are impossible with the more direct representation of Fig. 1.

Another example of a simple calculation that is useful for adjusting or extending experimental data gives critical masses of modestly diluted metal systems in terms of effective absorption and transport cross sections of the diluent. These cross sections, in turn, are obtainable from reactivity coefficients of diluent and undiluted fissile material. Figure 3 compares directly-observed critical masses of diluted enriched-uranium metal with results from integrated reactivity coefficients that were

CRITICAL MASSES OF BARE SPHERES OF U(93) DILUTED WITH OTHER METALS

FIG. 3

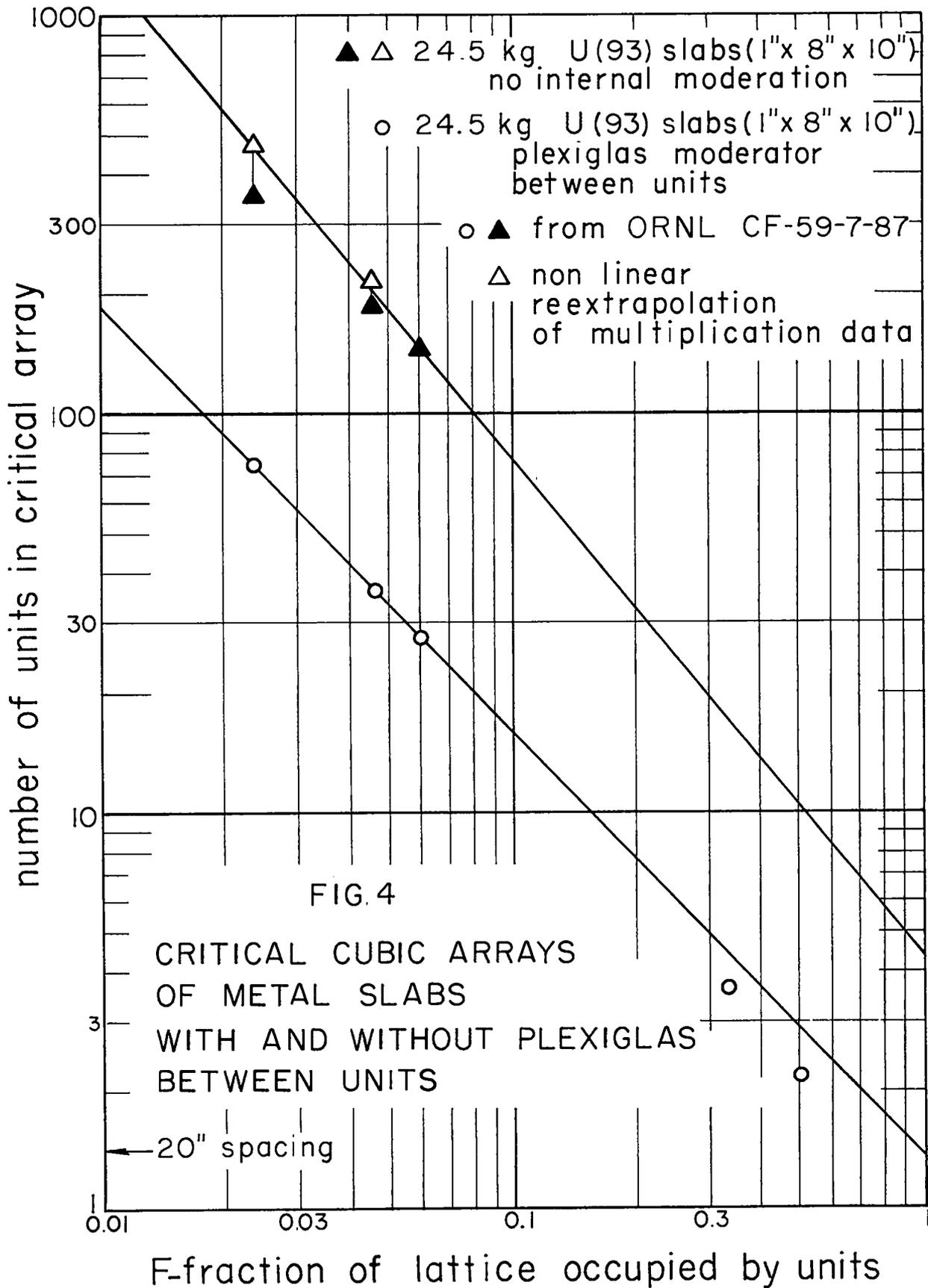
critical mass - kg U²³⁵

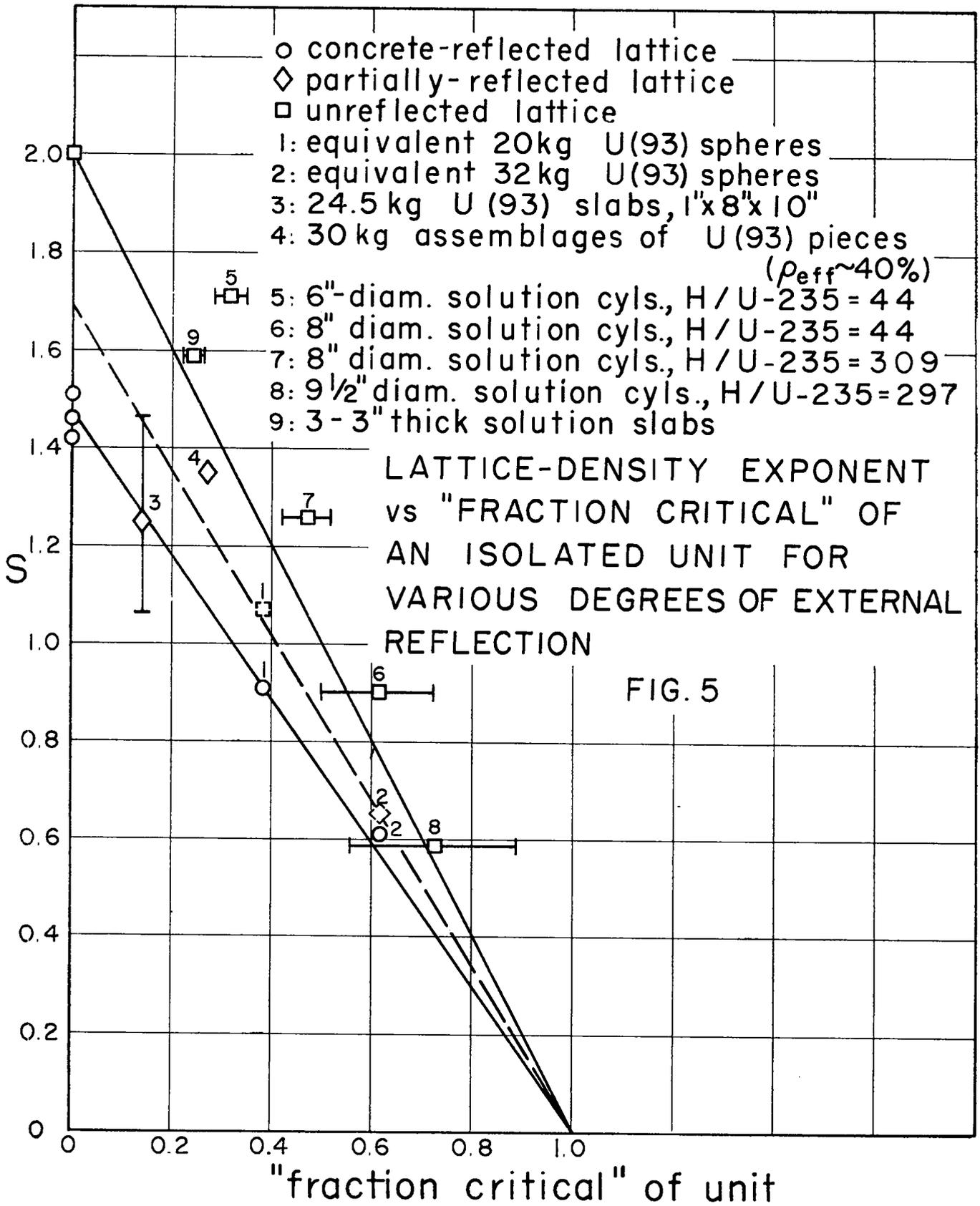


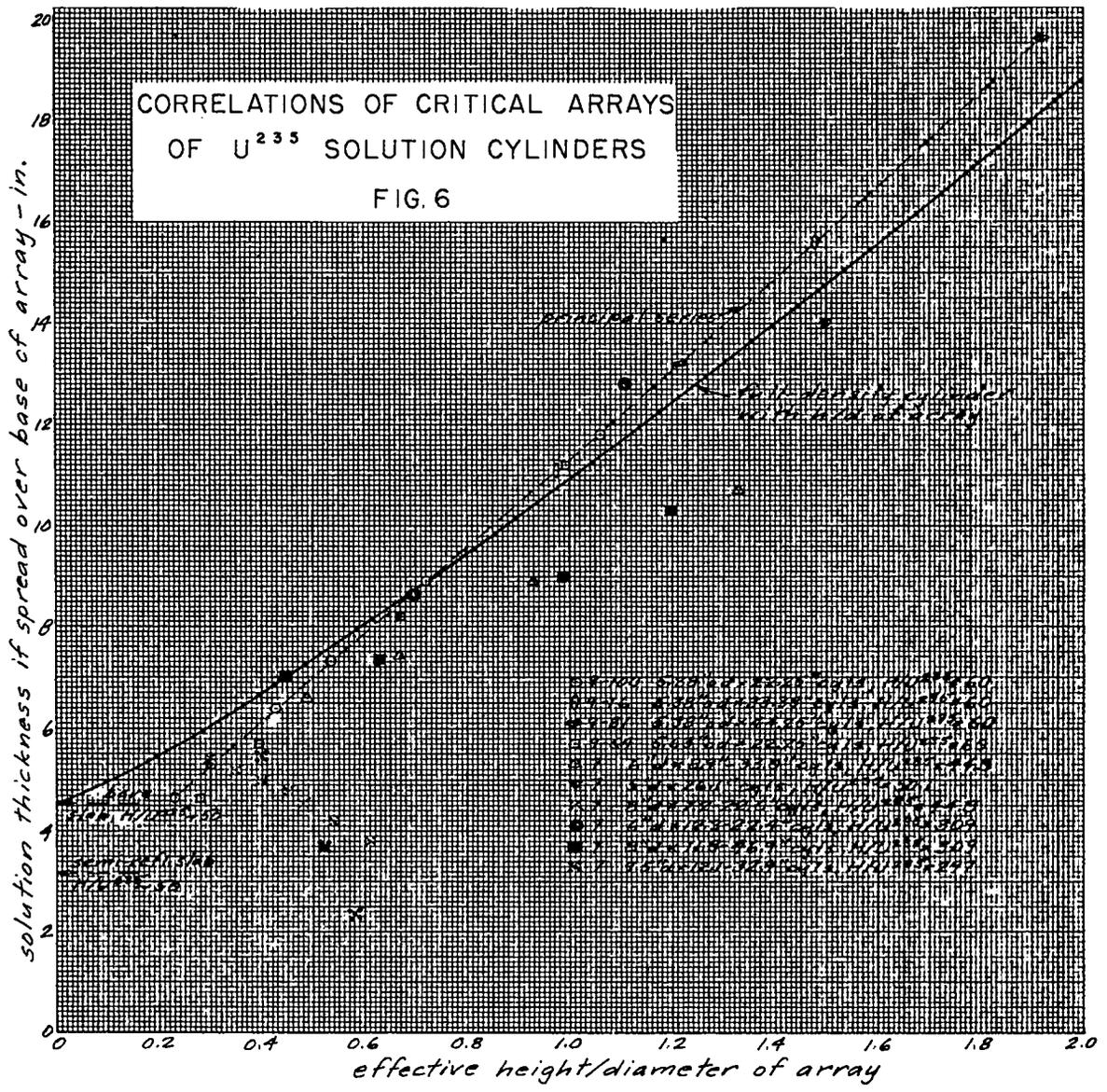
measured in an undiluted sphere. The primary critical data are from measurements by Jarvis (unpublished). The computed relationships are expected to hold through a range in which fixed one-group cross sections apply - - up to about 50 v/o dilution for graphite and perhaps to 70 v/o for most medium-Z and high-Z elements. This sort of correlation provides the bases for corrections of critical data to standard densities and concentrations.

A step further from detailed theory is a scheme for correlating cubic lattices of air-spaced fissile units, which is suggested by the power relationships of Fig. 3. In Fig. 4, data from Oak Ridge National Laboratory (2), the best presently available for cubic arrays, follows the form, critical number of given units = (lattice density)^{-s}, where s is constant over the range of measurement. Figure 5, which shows the way in which s depends upon type of unit and effectiveness of reflector about the array (3), is then a basis for generalization to unmeasured systems.

The picture of relationship between the critical mass of a plane array of discrete units and that of a uniform slab of the same material leads to the correlations of Fig. 6. Here, ORNL data on arrays of solution cylinders (4) are represented in terms of thicknesses of critical solution if spread over the base area of the array. It is







obvious that this representation breaks down at a finite height if the cylinder diameter is such that an individual unit can be made critical. Limited data on plane arrays of massive metal spheres appear to fall into a similar pattern, which suggests the possibility of useful generalization.

One feels less comfortable about the generalization of purely empirical forms for which the reason is not clear. An example is the relatively close clustering of ratios of water-reflected to bare critical volumes for a considerable variety of systems with similar moderation, as represented in Fig. 7 (5). Though this correlation is highly suggestive and establishes reasonable limits, we do not expect it to be reliable in detail. In this case, there happens to be hope for a simple guiding model, because a reasonable trend is from the observed minimum volume ratio of ~ 0.4 (for small, efficient systems) toward unity as neutron leakage becomes successively less important in fission economy (or as k_{∞} decreases). Thus, the greatest problem might not be how to improve the correlation, but, rather, how to find a better representation in practical engineering terms. Though volume ratios at standardized densities may form a nearly unique function of k_{∞} , the latter is not a convenient process parameter.

These illustrations, intended primarily to show the utility of crude theoretical models for data adjustment, will serve a second purpose. They give some basis for judging the uncertainties added to primary data when prepared for correlations.

Computation as a Substitute for Experiment

Attention now will be turned to a realm in which we contend that highly-refined calculation is required. This is where computed values are intended to substitute for experimental data in nuclear safety guidance. Just as experimental values require some index of reliability, computations which replace them should leave the user with a feel for probable error. Practically, there may be even more stringent requirements on computation than on experiment because of doubters who remember the days when a factor-of-two error in calculated critical mass was customary unless nearby experimental data were available for normalization. In spite of vastly improved input data, computational facilities, and techniques, some of the stigma remains; hence our opinion that calculation for the purpose under discussion should be so good as to sell itself to the more skeptical. (After reasonably-complete high-quality computational surveys exist, there may be a

place for short cuts "which seem to work".)

The requirements implied above are: realistic input, convincing method, and a wide enough range of checks against experiment to establish error patterns. The most useful form in which data might emerge is a computational survey. The classic of this type is the series of surveys of bare, spherical critical systems undertaken by Safonov a number of years ago (6). For U^{233} , U^{235} or Pu^{239} , diluted by H_2O , D_2O , Be, BeO or C, he presented critical sizes and certain spectral indices as functions of moderator-fuel ratio. Other series cover the ternary systems of U^{235} - U^{238} -C, U^{235} -Be-C, and, at elevated temperature, U^{235} - U^{238} - H_2O . Parameters for his multigroup diffusion method were chosen carefully, and reasonably represent processes including inelastic scattering and resonance region effects. As the diffusion approximation cannot apply precisely to a small system for which radius and mean-free-path for fission are of the same order, it was necessary to adjust high-energy transport cross sections for agreement with critical data from fast-neutron assemblies.

Recently, Mills (7) has applied more modern techniques to the binary systems that Safonov surveyed, and, for H_2O -moderated U^{233} , U^{235} or Pu^{239} , he included, with spheres,

critical cylinders of infinite length and slabs of infinite extent, and added H_2O reflection. Also covered, is a portion of the $U^{235}-U^{238}-H_2O$ series for bare spheres. The DSN method that Mills employed (8) has the advantage of applying realistically to fast-neutron systems, and input data presumably have been improved since the time of Safonov's work - - particularly a specific coded scheme of Bell's for averaging cross sections throughout resonance regions (9). The high energy cross section set is that of Hansen and Roach (10), in which parameters have been adjusted within their ranges of uncertainty to fit a wide variety of integral fast-neutron data.

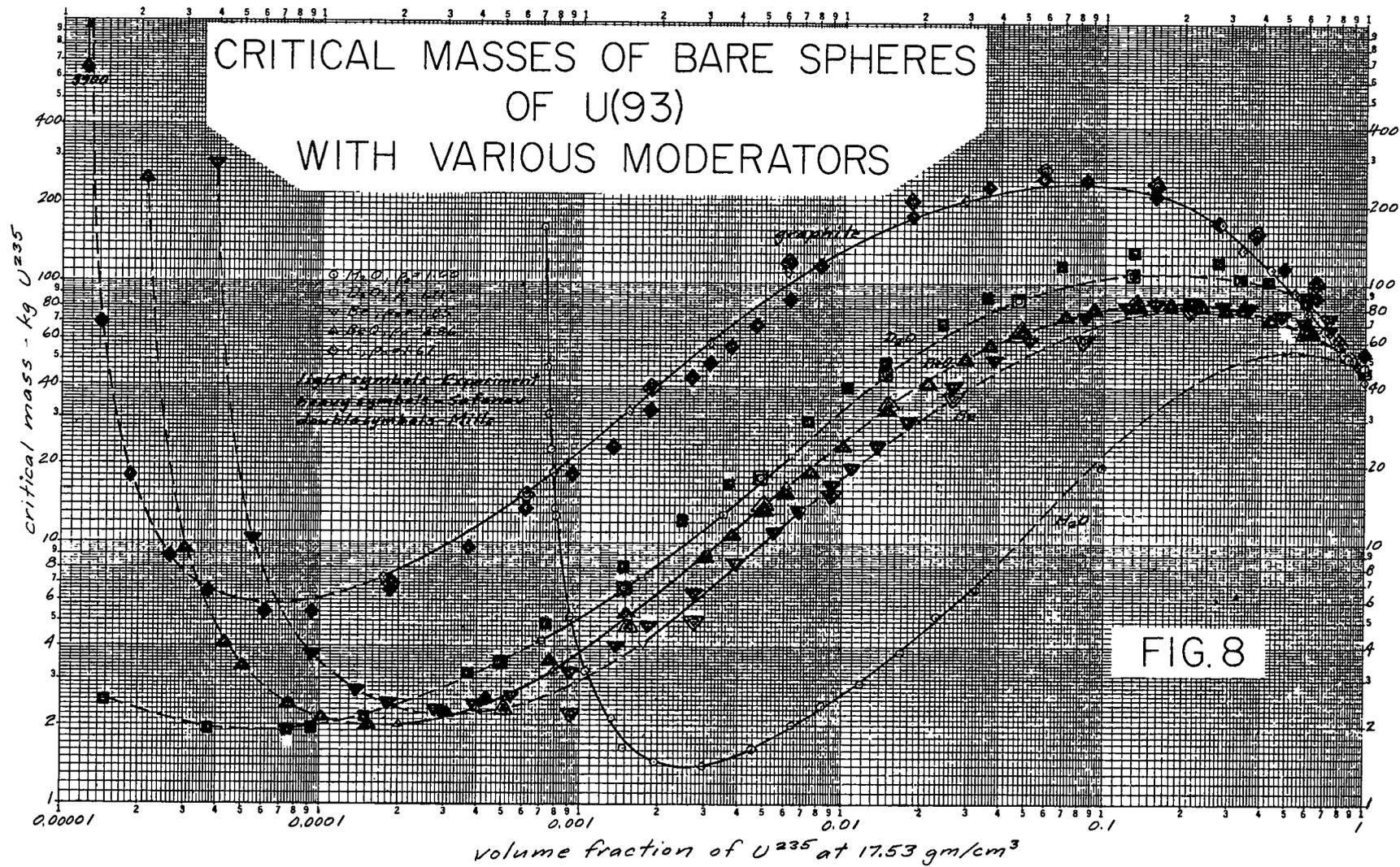
Other DSN surveys of interest to us were conducted by Roach for water-reflected $U(93)-H_2O$ and $Pu^{239}-Pu^{240}-H_2O$ spheres at normal and reduced densities, and by Stratton for bare and water-reflected $U(93)-H_2O-C$ spheres (both sets unpublished, but cross sections appear in Ref. 10). A difference with Mills' procedure, expected to be of minor influence, is an alternative hand method of averaging resonance cross sections. Anisotropic scattering cross sections for hydrogen are used in each case (except Mills' points of Fig. 21).

It is appropriate to mention that detailed computation, like good experiment, is expensive. The DSN method requires a modern, high-capacity computer, and the necessary time

increases with degree of moderation - - where advantages over the less-demanding multigroup diffusion method largely disappear. For this reason, Goodwin uses multigroup diffusion calculations for extensive nuclear-safety surveys, supplemented by fewer DSN results to provide check points and to cover neutronic complexities that are beyond the simpler method (for example, 11). We shall have occasion to use some of Goodwin's results, while concentrating on those of Safonov, Mills and Roach.

Comparisons of Experiment and Computation

Experimental data for bare spheres of highly-enriched uranium metal diluted in various degrees by D_2O , Be, BeO and C (graphite) are represented in Fig. 8. Water-moderated systems, which will be considered in more detail, are included only for comparison of form. Errors in data interpretation (shape conversion unspecified impurities, inhomogeneities, and incidental reflection) should be generally within 10%, with limits for H_2O and D_2O solutions appreciably smaller. Exceptions are the lower point that applies to undiluted metal (the shape was extreme and density uncertain), and possibly the point for ~ 90 v/o H_2O , where there was a compositional correction. Density corrections, according to critical mass = const (density)⁻²,



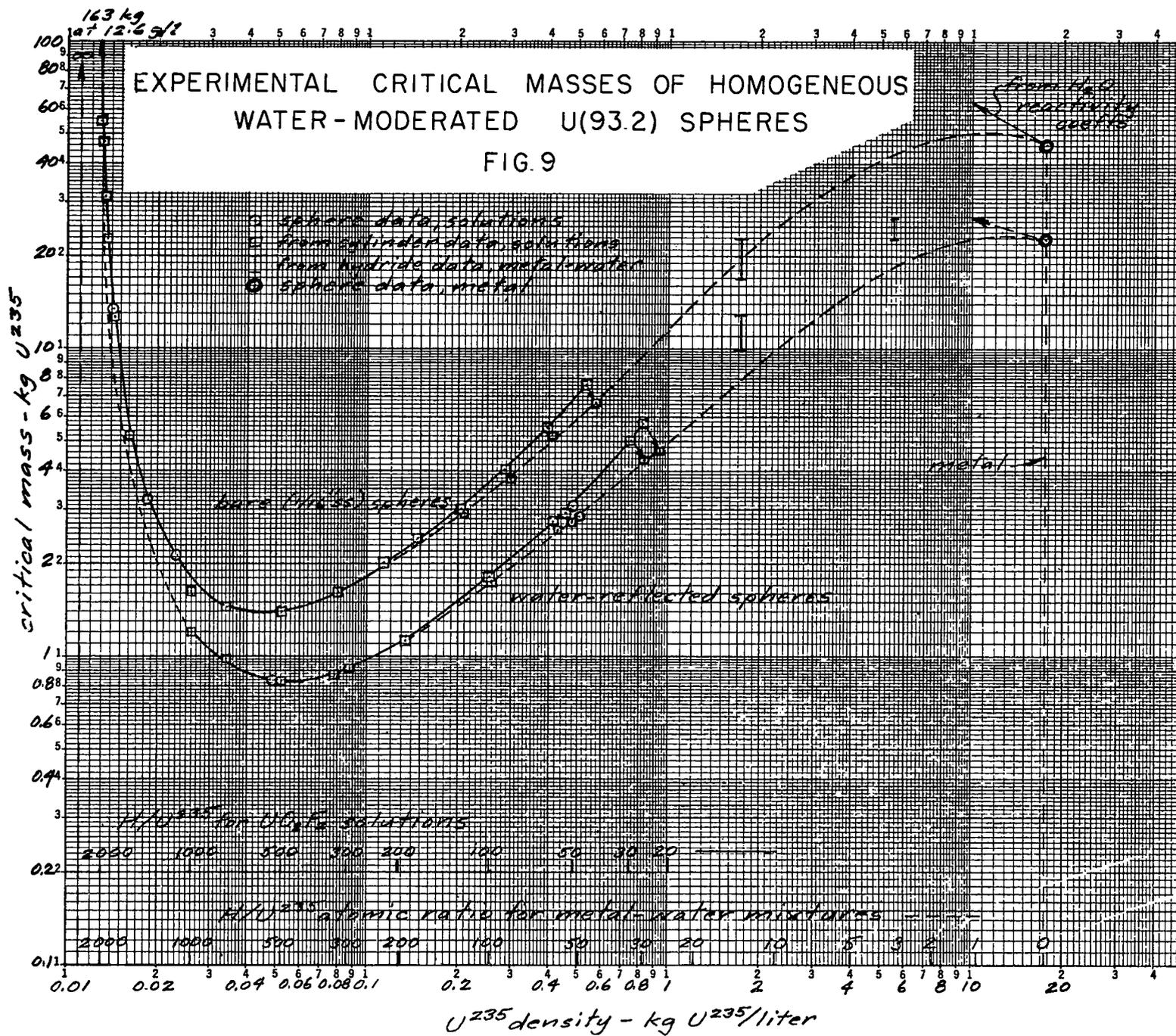
are quantitative. Calculations by both Safonov and Mills agree reasonably with experiment, maximum deviations being about 20%. In Safonov's case, this is particularly significant because most of the critical experiments followed his work. As expected, Mills' results show improvement.

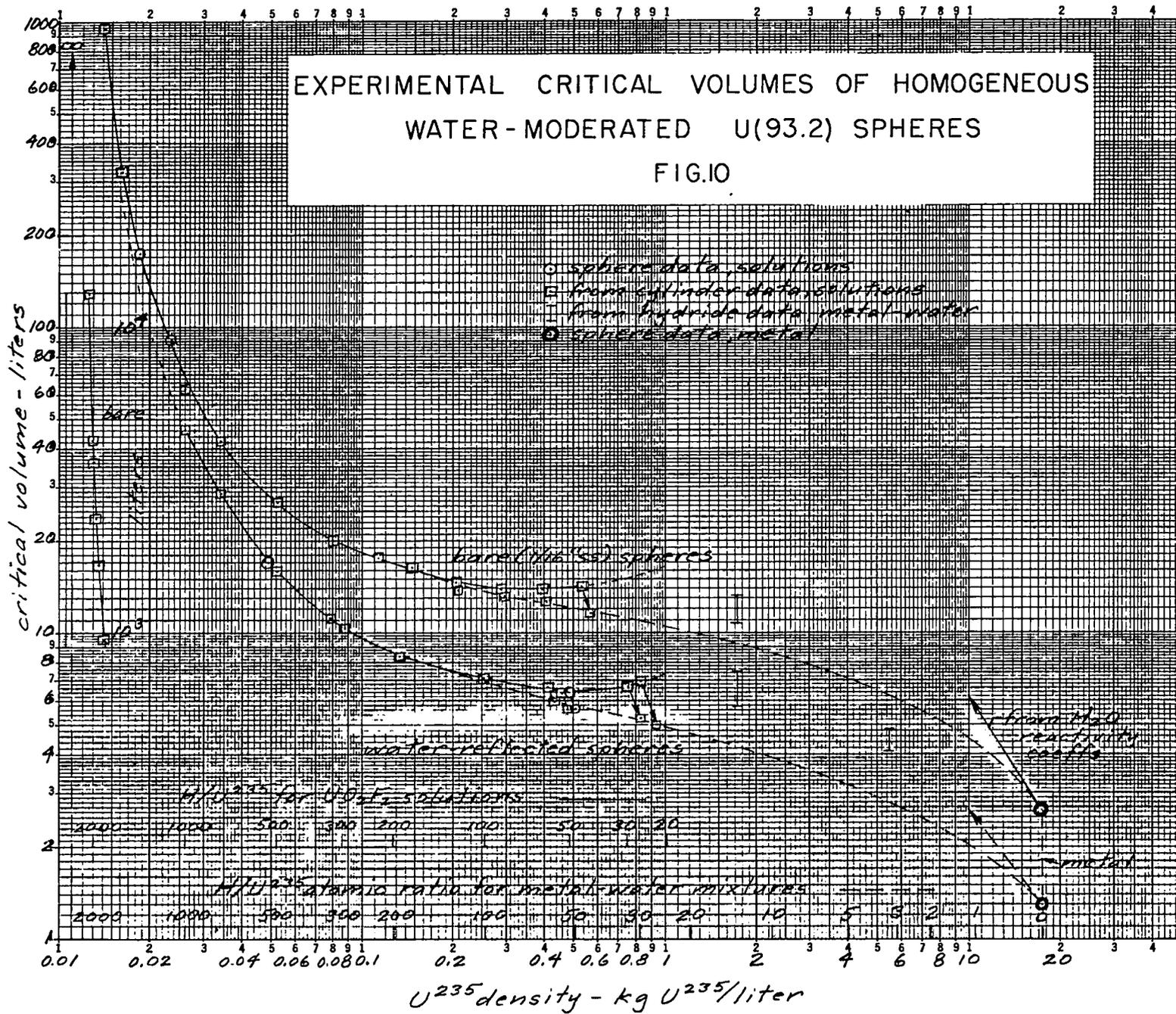
Experimental critical masses for water-moderated highly-enriched uranium spheres, both bare and water-reflected, are presented in Fig. 9. The importance of changes in U^{235} density at constant H/U^{235} atomic ratio is illustrated by the dual representation of data for concentrated solutions, first at normal densities then at the larger densities corresponding to idealized homogeneous metal-water mixtures. Even though such mixtures cannot be attained in practice, they represent an upper limit to density at a given value of H/U^{235} , hence, a lower limit to critical size. From the corresponding critical volume curves, Fig. 10, we see that the minima for solutions disappear for metal-water mixtures.

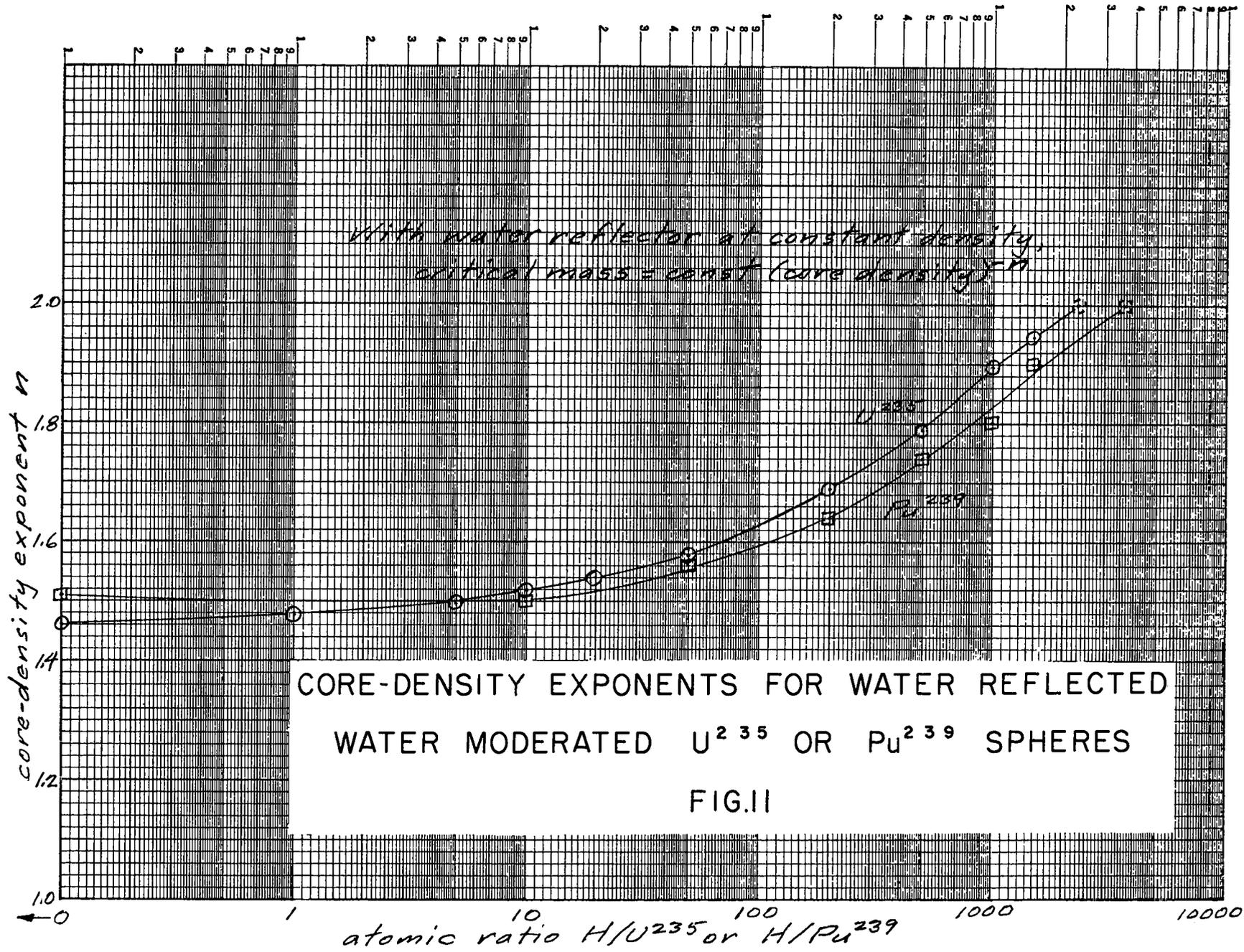
As has been noted, corrections, for example, to densities of mixtures of uranium compounds and water are straightforward for unreflected systems. When water reflection is added, however, the influence of a core-density change is reduced, as indicated by Fig. 11 for water-moderated water-reflected spheres. The core-density

EXPERIMENTAL CRITICAL MASSES OF HOMOGENEOUS WATER-MODERATED U(93.2) SPHERES

FIG. 9



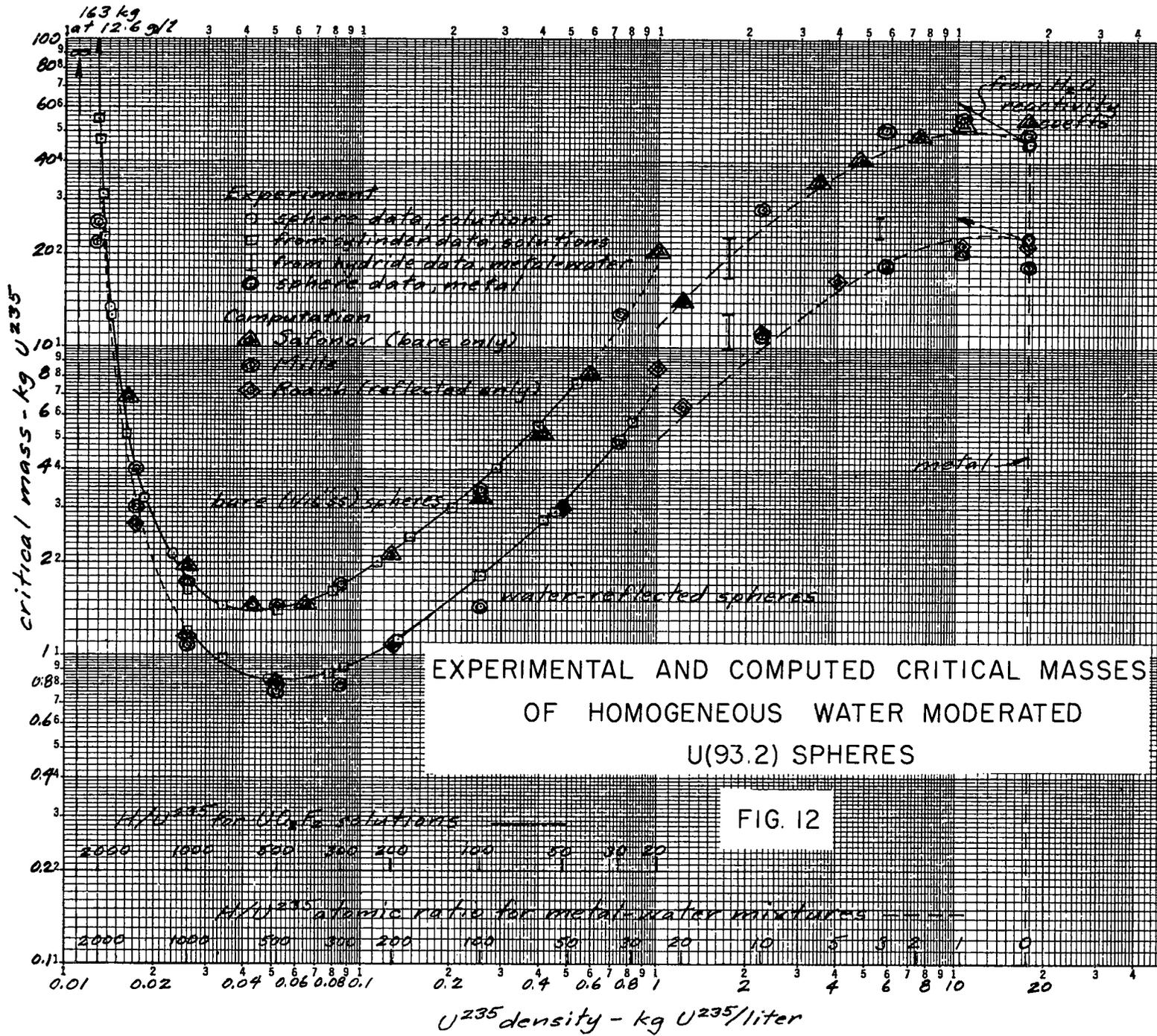


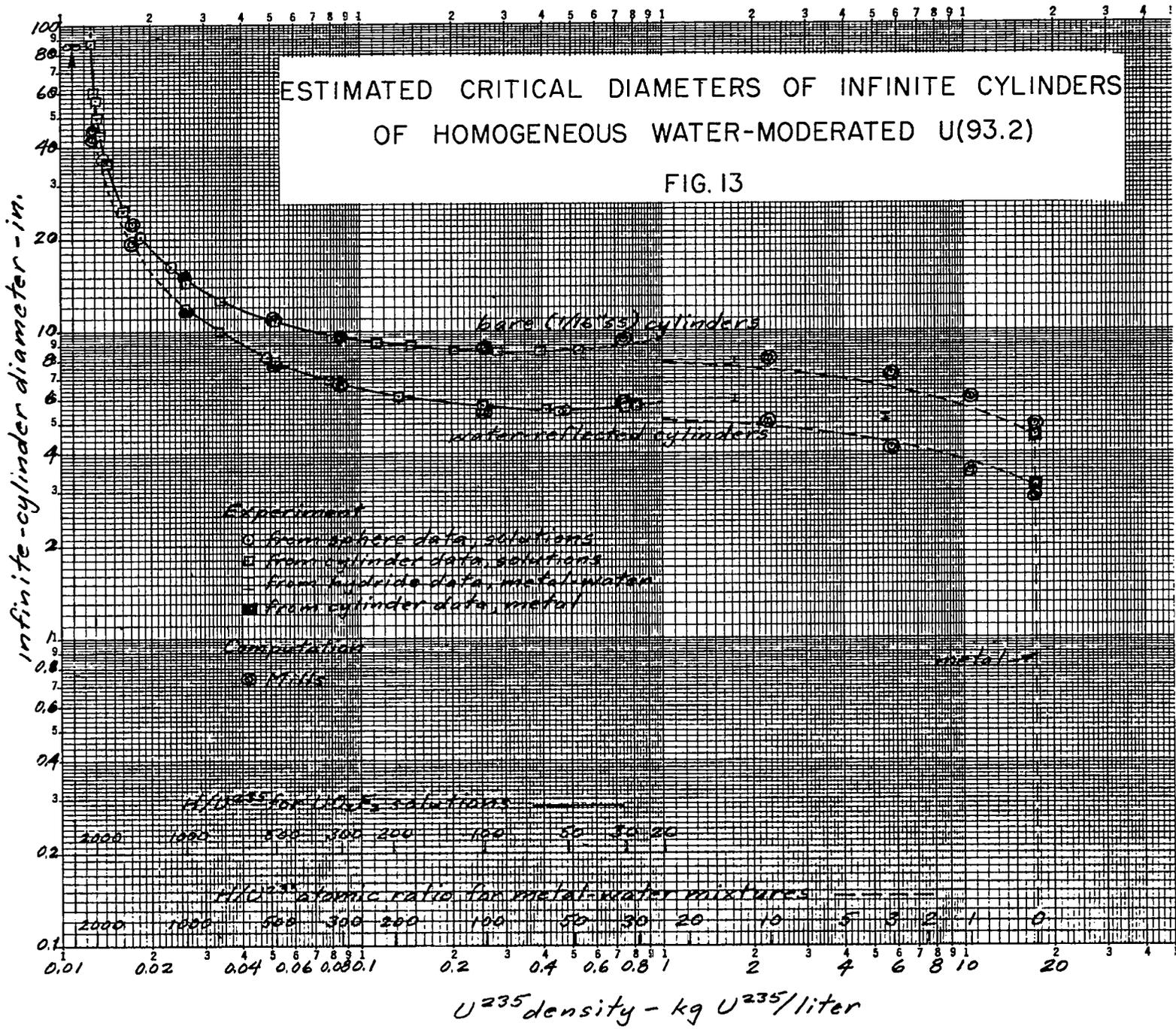


exponents shown, which are from critical calculations by Roach at two sets of densities differing by 20%, are used here at face value.

With Fig. 12, we return to the U^{235} critical-mass curves with density-corrected computations superposed. Only Mills has values for both bare and reflected spheres. Though experimental points for solutions appear to be accurate to within 5%, the two water-reflected points that are derived from "hydride" assemblies should be discounted because of serious uncertainties in composition and reflector-savings corrections (recently-"improved" corrections led to alarming shifts). Direct checks of computation against the undistorted hydride data would serve a better purpose than inclusion on these curves. It should be noted that experimental "unreflected" points are biased downward, perhaps 2% for concentrated solutions, by the nominal 1/16" stainless-steel container which was not included in calculations. Safonov's values, which may be a few percent high due to a thermal base of 500^oF, have been chosen as the latest and best of several sets. Generally, the agreement between solutions data and calculation is appreciably better than the maximum deviation of ~ 20%.

Empirical data for infinitely-long cylinders and slabs of infinite extent (Figs. 13 and 14), of course are simply

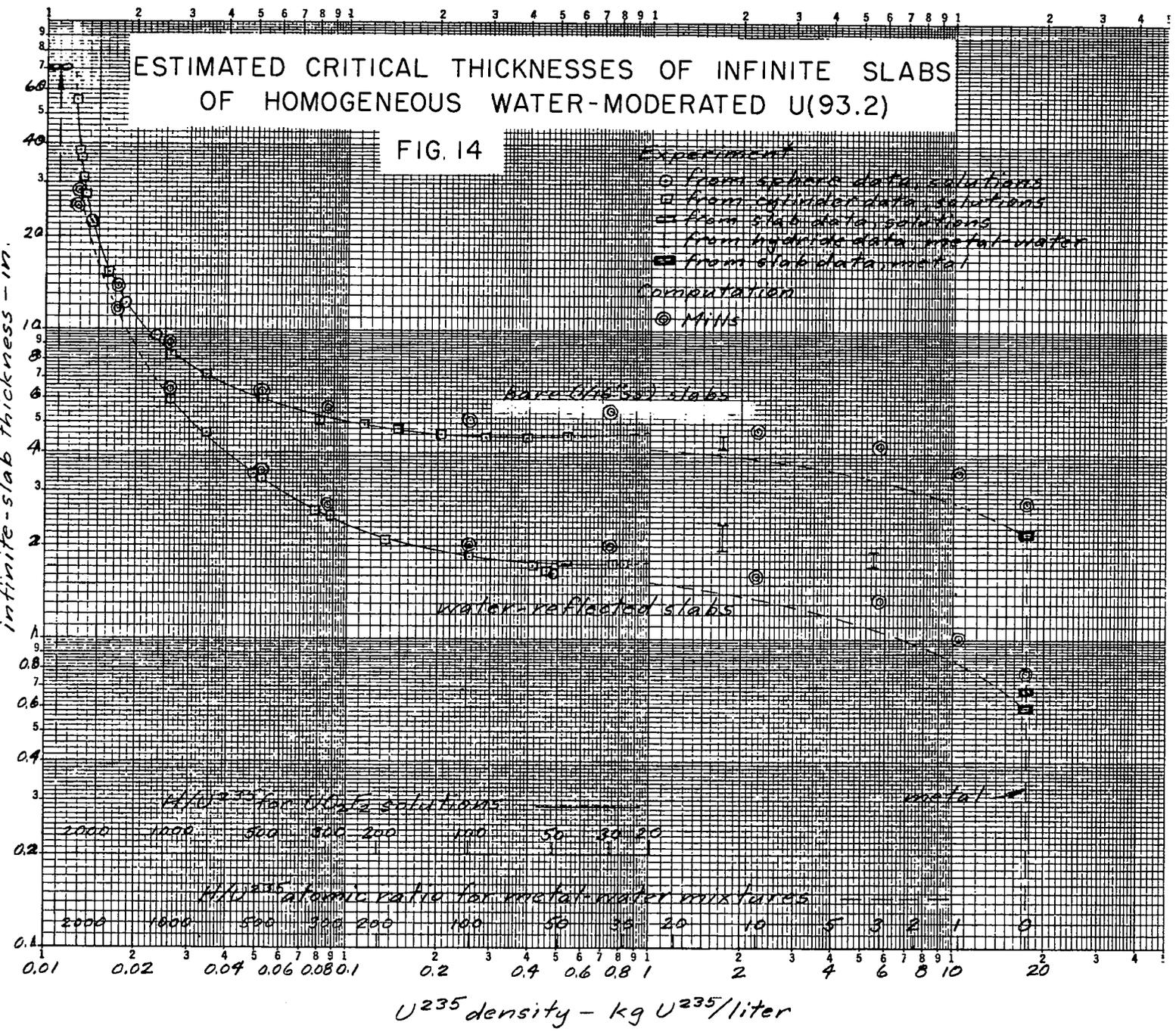




ESTIMATED CRITICAL THICKNESSES OF INFINITE SLABS OF HOMOGENEOUS WATER-MODERATED U(93.2)

FIG. 14

infinite-slab thickness - in.



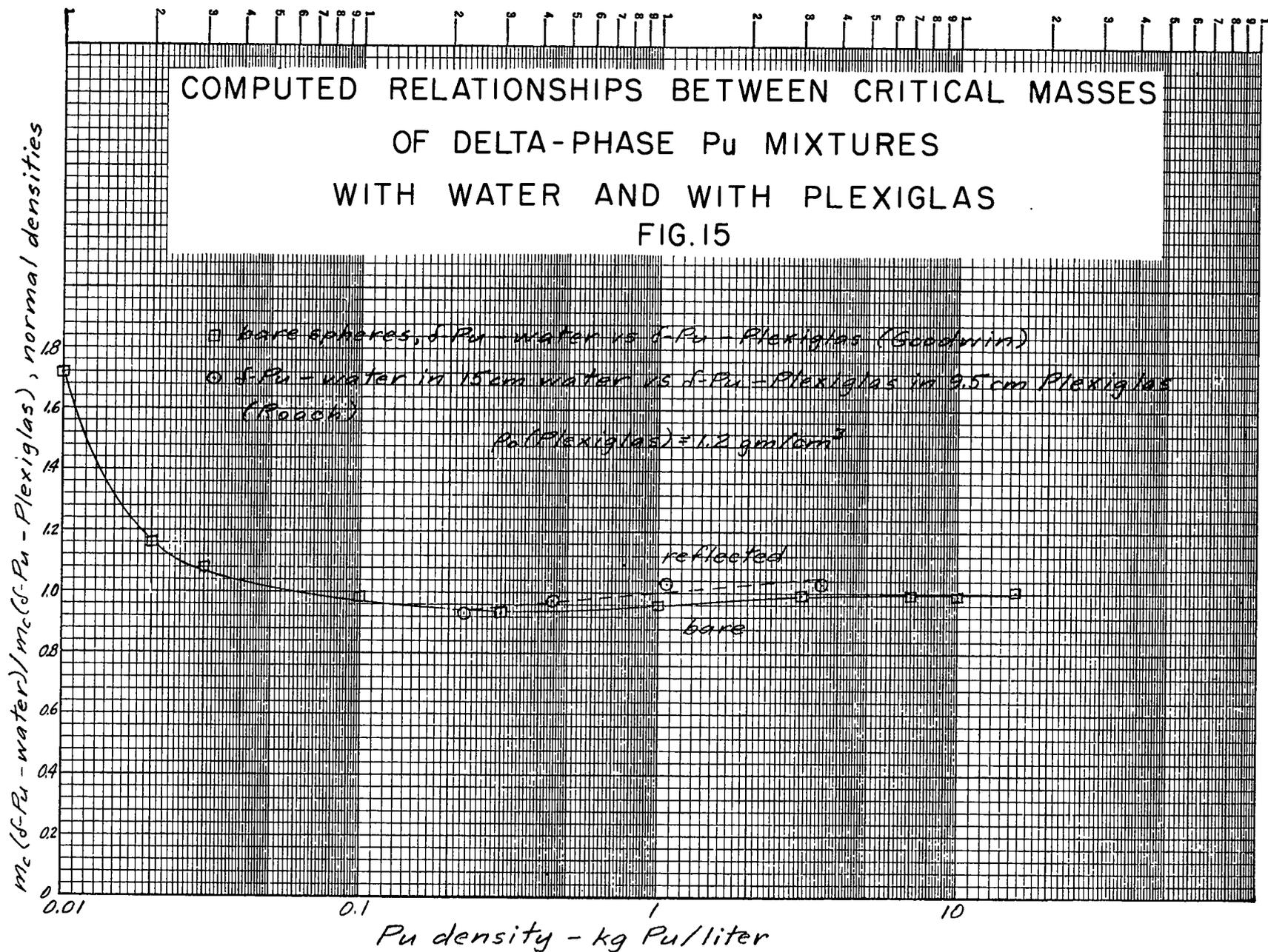
U^{235} density - kg U^{235} /liter

based on observation. For undiluted metals and solutions near the critical minima, measurements on long cylinders and broad slabs lead to reasonably convincing values for the infinite systems. In other regions, estimates of slab thicknesses are considerably more sensitive to conversion errors than are the cylinder diameters. In spite of this, it appears that Mills' slab values are systematically high, as contrasted with his calculations for cylinders.

Information about water-moderated plutonium assemblies is not as clean as that for U^{235} . Existing critical data, for metal and for dilute solutions only, has recently been supplemented by subcritical measurements of Schuske and his coworkers at Rocky Flats on plutonium metal sheet with Plexiglas as moderator and, in some cases, as reflector (12). But data on the influence of ever-present Pu^{240} , and on effects of variation in moderator composition are scanty. For this reason, it is desirable to use detailed computation for adjusting the Rocky Flats observations. Relative critical masses of Plexiglas-moderated plutonium (bare or Plexiglas reflected) and of water-moderated plutonium (bare or water reflected) from such calculations appear in Fig. 15. Plutonium densities remain at normal values for the mixtures. The curve for bare spheres was obtained by Goodwin, and that for reflected spheres by Roach. Figure

COMPUTED RELATIONSHIPS BETWEEN CRITICAL MASSES
 OF DELTA-PHASE Pu MIXTURES
 WITH WATER AND WITH PLEXIGLAS
 FIG. 15

31



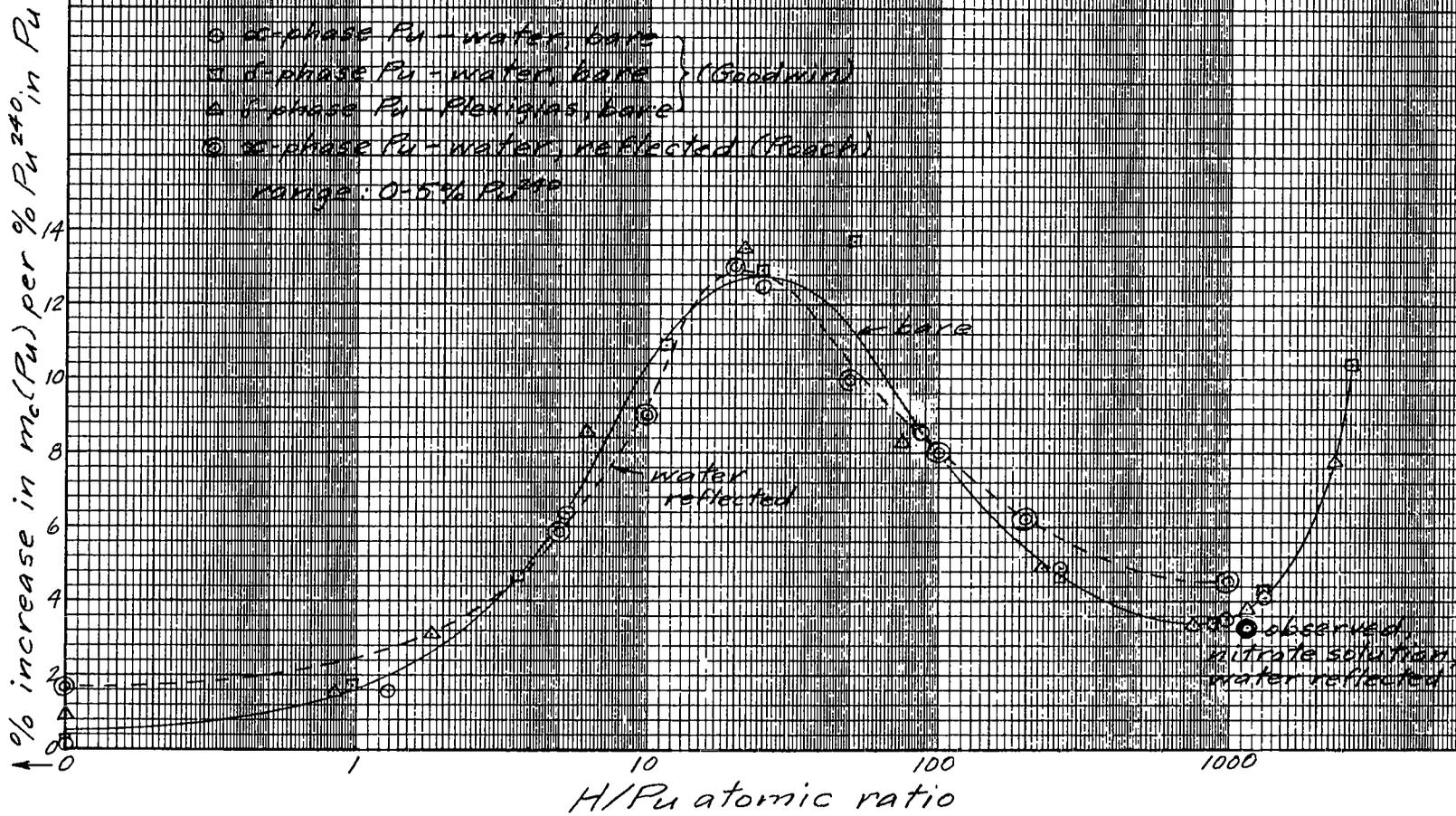
16, also due to Goodwin and Roach, gives the percent increase in sphere critical mass of total Pu per percent Pu²⁴⁰ content of the plutonium.

Making use of the last two sets of curves and density exponents from Fig. 11, critical masses from the Plexiglas-moderated assemblies were joined with the older data for solutions and unmoderated metal as in Fig. 17. The "idealized solutions" represented in addition to metal-water mixtures have a relationship between fuel density and hydrogen-fuel ratio which is analogous to that of UO₂F₂ solutions. Practical densities generally would be less because of the excess acid required for stable Pu solutions. In Fig. 18, where computed results are added to the experimental curves, the reasonable clustering is particularly significant, as only Safonov used the points applying to moderated assemblies for cross section adjustment (he used just dilute-solution data for this purpose). Here it is difficult to judge whether values from experiment or from calculation are the more reliable. If consistence is a valid criterion, it appears that critical masses are tied down to within 20% over most of the range of moderation.

Except for water-reflected metal, observations on critical cylinders and slabs containing Pu are so limited that conversions to the infinite forms must be based on

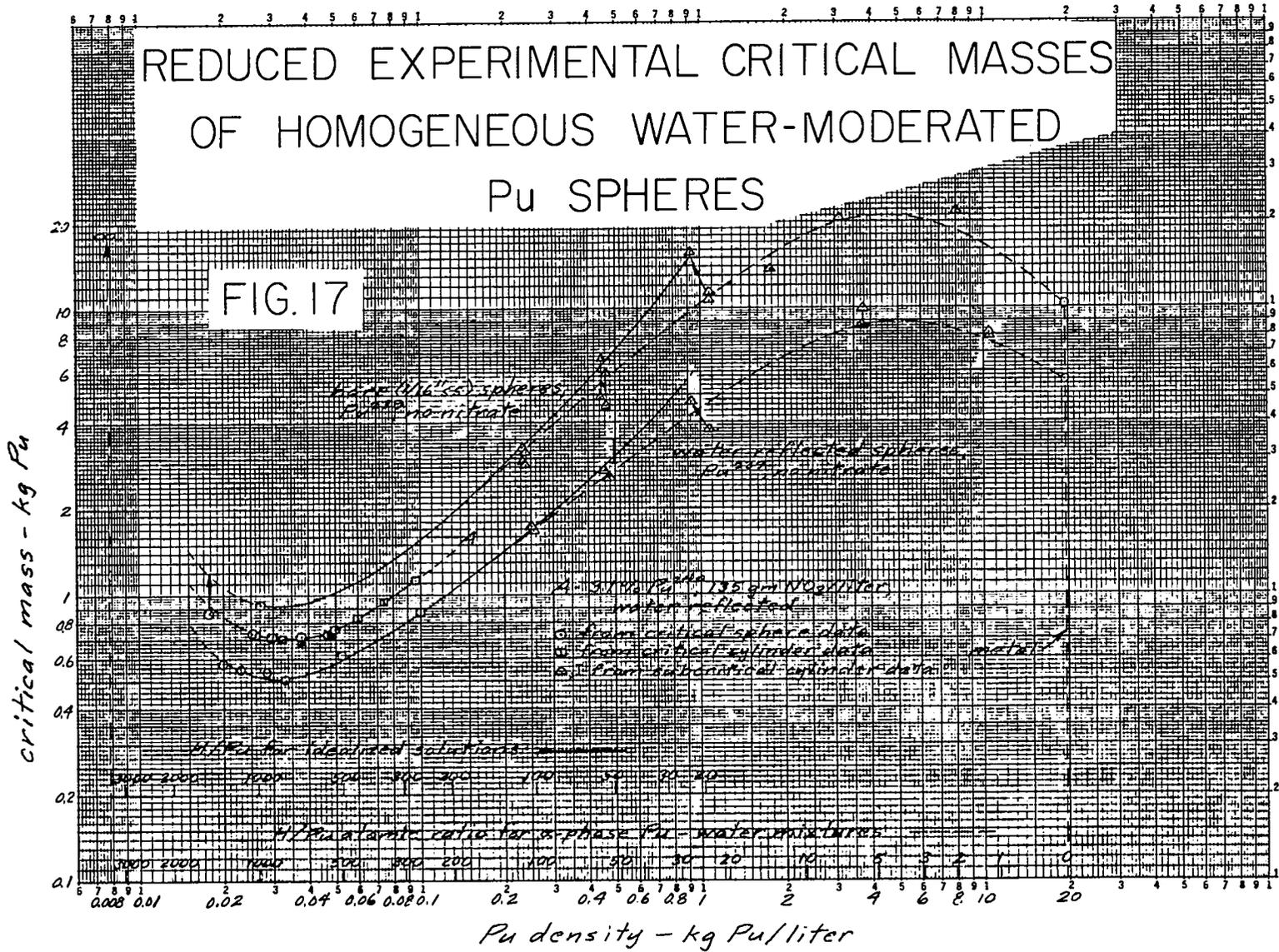
COMPUTED INFLUENCE OF Pu^{240} ON CRITICAL MASS OF WATER-MODERATED Pu SPHERES

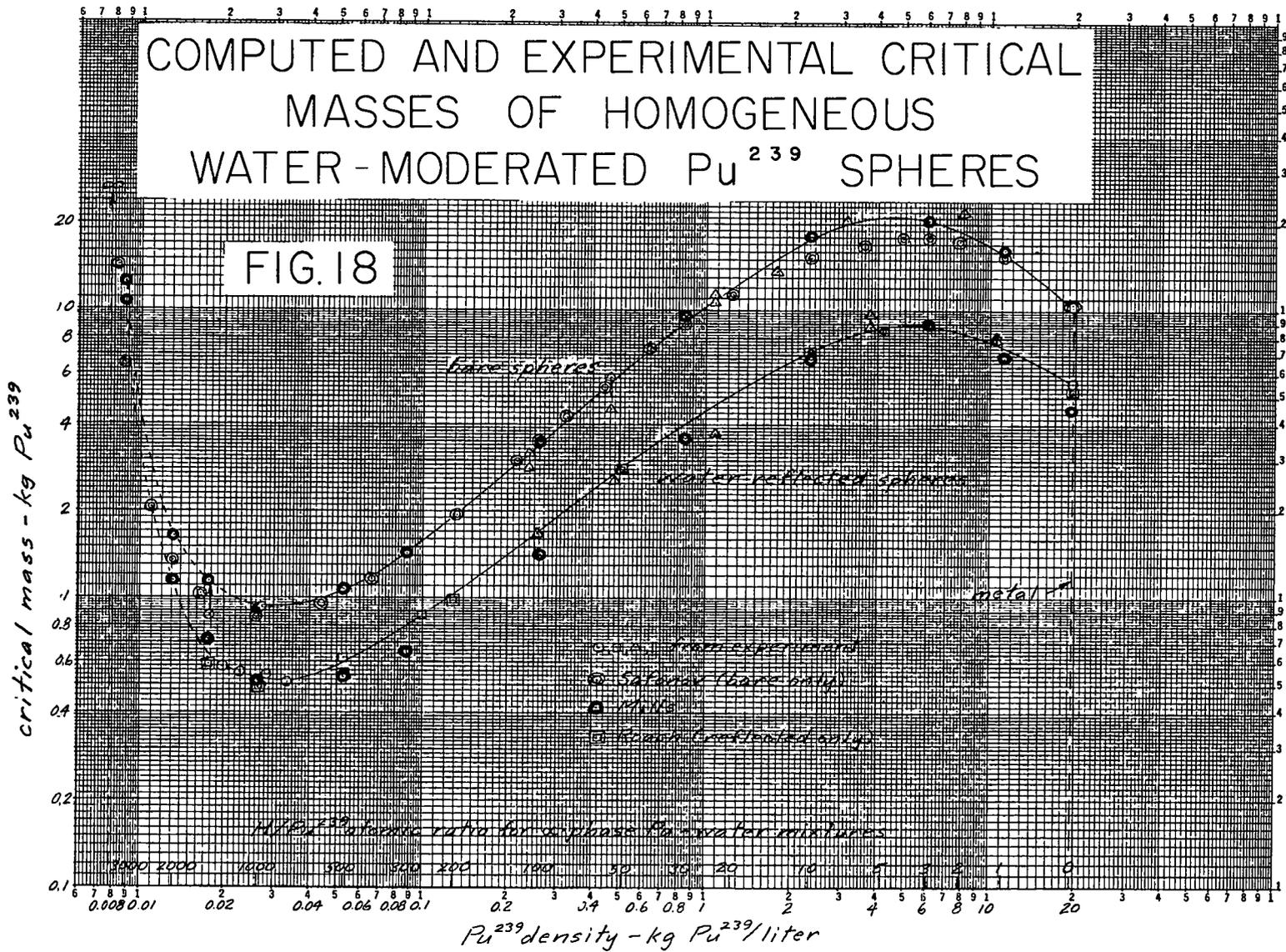
FIG. 16



REDUCED EXPERIMENTAL CRITICAL MASSES OF HOMOGENEOUS WATER-MODERATED Pu SPHERES

FIG. 17



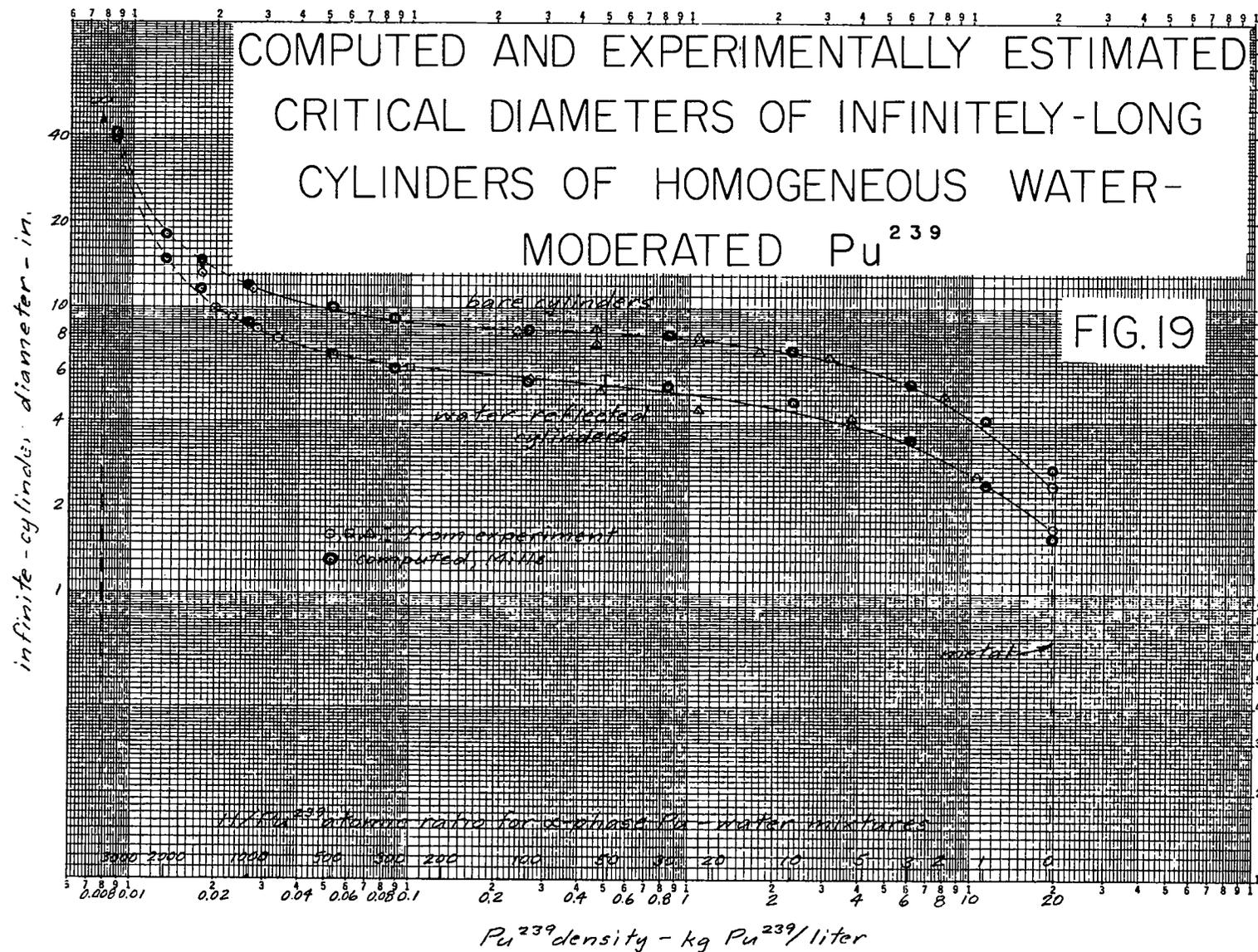


models which apply to U^{235} (Figs. 19 and 20). In spite of this added uncertainty, differences between points based on experiment and those computed by Mills are similar to the uranium cases.

Computation with Limited Experimental Confirmation

Fissile systems that are sensitive to more than two constituents demand computational guidance to a greater extent than the essentially two-component mixtures upon which we have concentrated. The simple reason is that the new dimension (or more) makes complete experimental coverage prohibitive. Examples of practical interest are U^{235} - U^{238} - H_2O combinations for partially-enriched fuel-element materials, U^{235} -C- H_2O for impregnated graphite elements, and U^{235} -Be- H_2O for impregnated beryllium.

As the transportation and general handling of partially-enriched uranium compounds (which may become moist) is a current problem which requires attention, let us see what is known about critical masses of water-moderated U^{235} - U^{238} combinations. The principal family of curves in Fig. 21 are from Safonov for a temperature of $260^{\circ}C$ ($500^{\circ}F$), but corrected to normal metal-water densities. To indicate the order of neutronic changes when the temperature is dropped to $20^{\circ}C$, Mills calculates a 25% decrease in

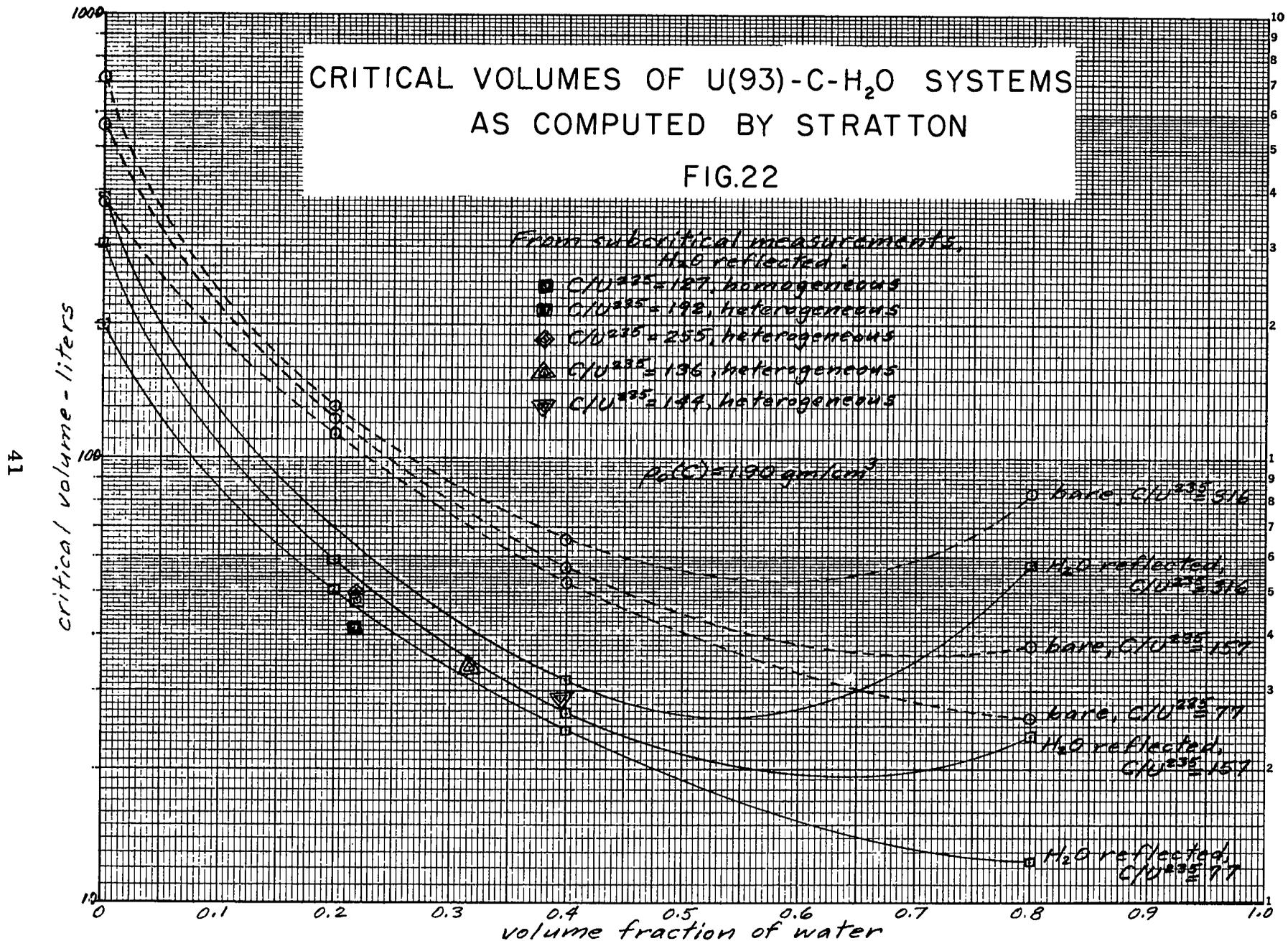


the minimum critical mass for U(2% U²³⁵) versus a decrease of just 5% for U(93). The gas model which he used may overestimate the influence of temperature change, and we do not know what differences Safonov might have found. So it is with apology that we have added to Fig. 21 room-temperature points from experiment and from a preliminary survey by Mills (for this survey, convergence requirements were ~ 1% on radius instead of the customary factor of 10 less). Though temperature corrections would bring data of the three general types more or less together, limitations on experiment, present uncertainties in Mills' calculations, and the inappropriate temperature of Safonov seem to combine to call for a thorough computational survey. Mills has undertaken this, including the establishment of asymptotes on the low-moderation side of minimum.

The next illustration is a set of critical masses for U(93)-C-H₂O which was computed by Stratton specifically for nuclear safety (Fig. 22). As method and cross section sets are identical to those used by Roach, comparisons of Roach's results and experiment for straight water moderation (Fig. 9) are pertinent. Computed critical masses for graphite-U²³⁵ systems fall closely with those of Mills on Fig. 8. The few experimental values are from subcritical measurements and all but the left-hand point refer to

CRITICAL VOLUMES OF U(93)-C-H₂O SYSTEMS AS COMPUTED BY STRATTON

FIG.22



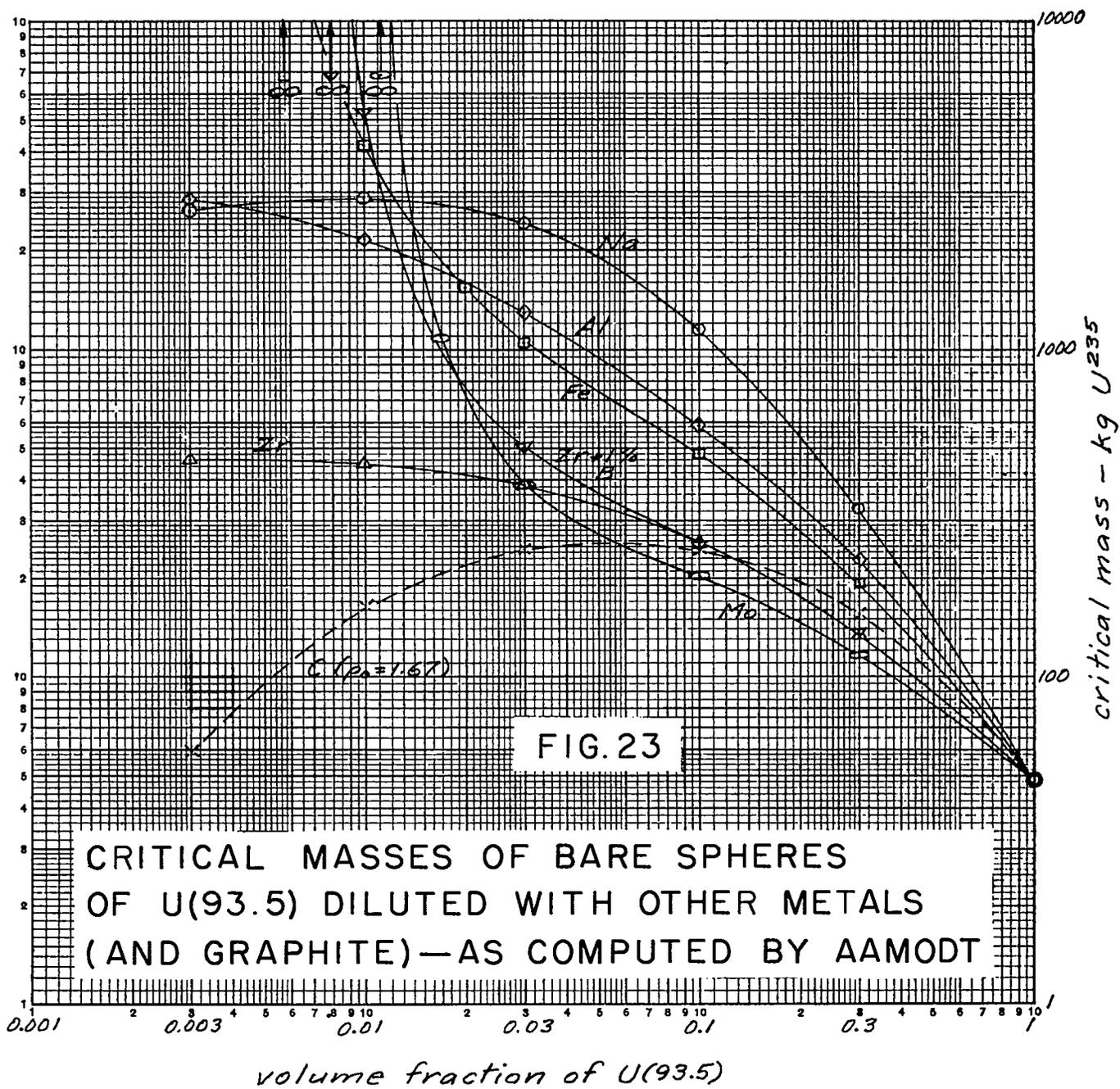
systems with inhomogeneous hydrogen distribution. Errors of interpretation could be as great as departures from computed values. For our purposes, limits were established by scaling down computed curves to pass through the lower experimental points, then applying a factor of safety of at least 2. From the apparent reliability of calculations with each moderating component separately and by comparison with the crude points for ternary assemblies, it is expected that most of this safety factor will be preserved.

Comments on Reliability and Safety Factors

The correlations which we have seen should leave us with a feeling as to the potential reliability of both experimental and computed critical parameters. In general, experimental data fall into two categories: precision measurements (critical or near subcritical) which require only well-defined corrections to standard conditions, and information which, because of imprecise critical determination, unclean assembly, or a degree of inappropriateness, serves only as a crude guide. Fortunately, there are reasonably abundant (though never sufficient) data of the former class.

The ideas that we have developed about the precision of detailed computation may be supplemented by what can be

judged from certain controlling input parameters and their reliability. To illustrate this point, Fig. 23 shows results of DSN computation (R. Aamodt-unpublished) which extend the very limited experimental values of Fig. 3. The answer to the question as to whether the calculations for dilute ranges may be used for nuclear safety guidance is not a clear "yes" or "no". Within regions to the right of upturns toward asymptotes, departures from the important anchor-point for undiluted uranium are controlled by relatively straightforward scattering properties of the diluent. Critical-mass values will not be a factor of two high if capture by the diluent has less than 10% influence on $\bar{\eta}$, the effective excess of neutrons produced per absorption. This criterion will be satisfied over most of the range that we are considering. Where curves break away toward asymptotes, however, critical masses (and the limiting critical compositions) become extremely sensitive to capture cross sections, for which 50% errors have been common. In view of this, I would not use, for nuclear safety purposes, either critical masses above the scattering base or asymptote locations unless there were experimental confirmation. (Actually the need for such guidance would imply the existence of enough material to make possible some sort of experimental check.)



Before expressing an opinion about the general reliability of detailed computation, I wish to make a side-line observation about non-input errors which are not uncommon. Any detailed calculation such as DSN is similar to a complex experiment that requires special procedures to verify the proper functioning of equipment and guard against extraneous effects. With the ever-present possibility of transcription error or machine malfunction, there should be a careful consistency check of any isolated problem before the result is accepted. The advantage of a parametric survey is that difficulties of this type are likely to result in inconsistencies that are obvious. Provided calculations have been properly checked, I believe that a probable error in critical mass of $\pm 10\%$ to 15% is about right for existing DSN techniques with proven cross section sets. This does not apply to regions in which critical masses change precipitously with composition.

A complete discussion of factors that should be applied to critical data for establishing nuclear safety limits would require a full description of the operations for which the limits are intended and of the nature of controls. I do not know how to generalize past the recommendations (and added precautions) which appear in the NUCLEAR SAFETY GUIDE (13). When the necessarily arbitrary

safety factors which appear there are translated to new situations, it is obvious that there should be added allowance for uncertainties in data. The importance of such an allowance diminishes if the limit applies to an idealized situation that is actually unapproachable in practice. Examples would be the application to normal vessels of limits for fully-reflected systems, and storage-shelf limits that are based on a cubic array of maximum-size units at the minimum allowed spacing. Hidden, but generally large, factors of safety in these cases greatly relieve the demands on guiding data. Information specific to a plant layout, in contrast to that which appears in our surveys, of course would eliminate many of the hidden safety factors that are now necessary.

Final Remarks

Any worker in the nuclear safety field, when faced by gaps in critical data, feels that his efforts are tentative. In exactly this sense the present review is recognized as describing just the beginning of an effort that should be carried on by many. A continuing, possibly coordinated, computational program is needed - - and let us hope that it will be of the best.

In representing detailed computational methods by the

DSN and multigroup diffusion techniques, it has not been the intention to discriminate against alternatives such as Monte Carlo. This method could make its appearance when correlations are extended beyond the simple systems that we have considered.

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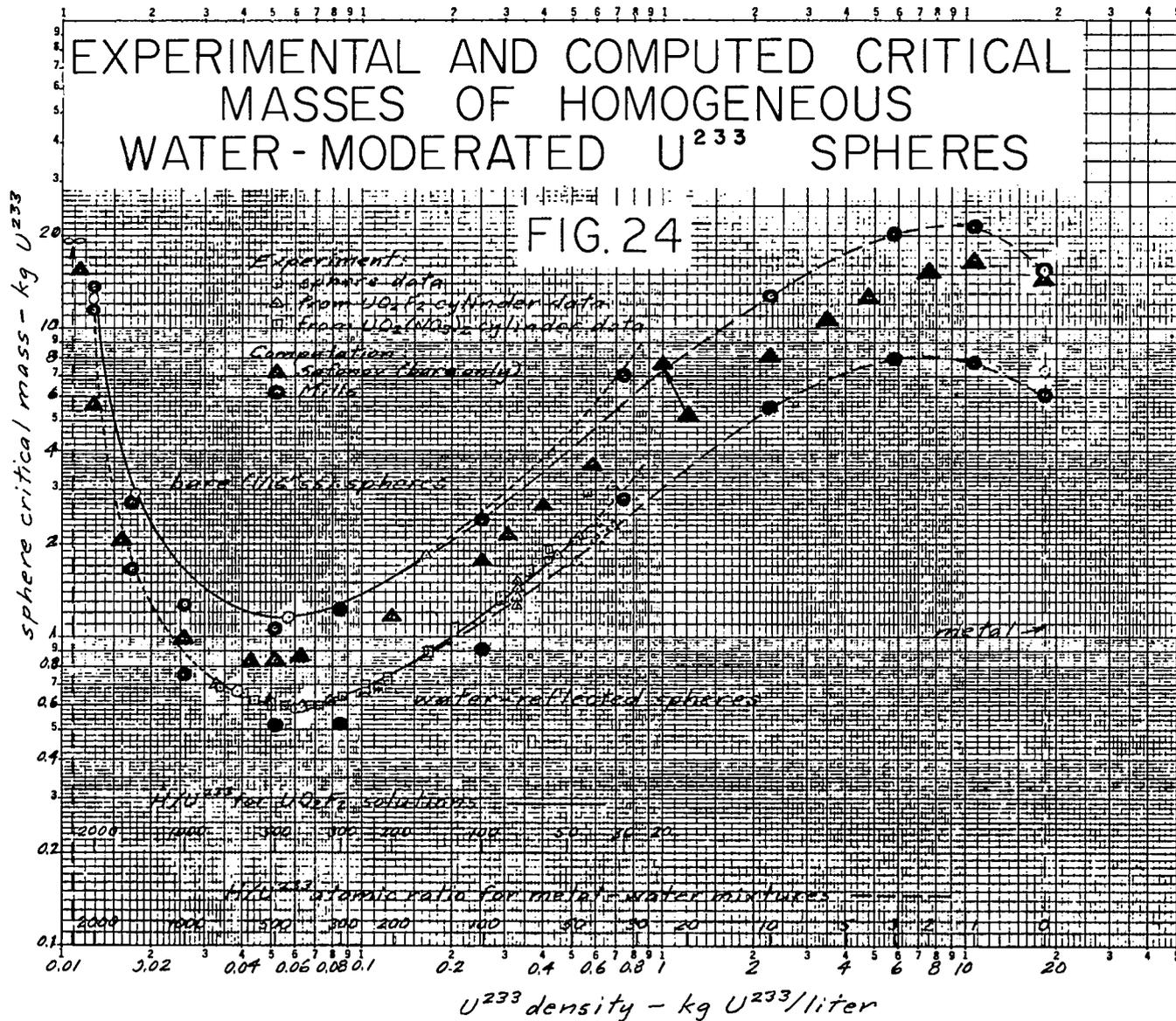
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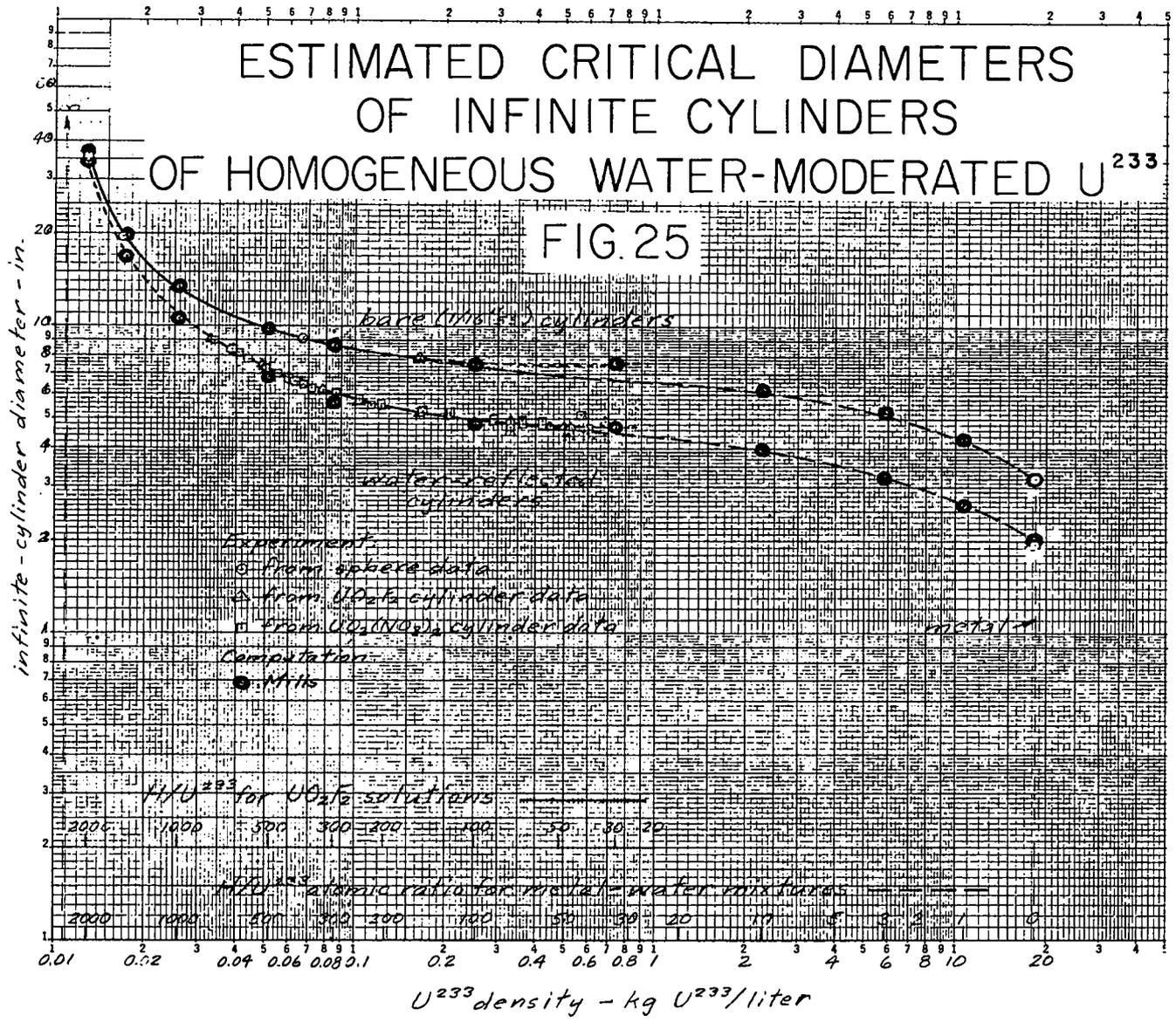
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APPENDIX

Data for U²³³

Though most nuclear safety problems center about U²³⁵ and Pu²³⁹, experimental and computed data for U²³³ are added here for completeness. In Fig. 24, critical masses of U²³³ solutions (and unmoderated metal), reduced to apply to spheres, are compared with results of calculations by Mills and by Safonov. Figures 25 and 26 give corresponding critical diameters of infinite cylinders and thicknesses of infinite slabs, both as estimated from experimental data and as computed by Mills.





ESTIMATED CRITICAL THICKNESSES OF INFINITE SLABS OF HOMOGENEOUS WATER-MODERATED U^{233}

FIG. 26

