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THE EFFECT OF URANIUM DENSITY ON THE SAFE U-235 ENRICHMENT CRITERION

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THE EFFECT OF URANIUM DENSITY ON THE SAFE U-235 ENRICHMENT CRITERION

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ABSTRACT

A review of the density effect on the criticality of low enrichment uranium has indicated that the nuclear homogeneity of fissionable materials, considered under the safe enrichment criterion of 0.95%U-235, should be established for nuclear safety. It appears that a condition of nuclear homogeneity will be maintained if (a) the uranium density is less than or equal to 3.2 g. U/cm.³, or (b) for densities ranging from 3.2 g. U/cm.³ up to a maximum of 18.9 g. U/cm.³, one dimension of the uranium fuel is less than or equal to 0.02 inch.

THE EFFECT OF URANIUM DENSITY ON THE SAFE U-235 ENRICHMENT CRITERION

INTRODUCTION

The principal factor in handling low enrichment uranium materials without regard to the usual criticality controls of mass or geometry is the establishment of the minimum critical U-235 enrichment for the uranium system of interest. In the case of homogeneous, hydrogen-moderated uranium systems, data obtained with the Hanford physical constants testing reactor, the PCTR, has indicated a minimum critical enrichment of about 1.03% U-235.¹,² This work, together with supporting criticality experimentation at the Oak Ridge National Laboratory,³ forms the bases of the maximum safe enrichment criterion of 0.95% U-235 adopted by the Oak Ridge Gaseous Diffusion Plant.

A concomitant use requirement of the safe enrichment criterion is the specification of a maximum uranium density of 3.2 g. U/cm.³, normally anticipated for diffusion plant operations, ⁴ and which is considered necessary because of possible slight inhomogeneities of plant materials and the generally greater reactivity of high density non-homogeneous uranium systems at low enrichments. However, with increasing interest in higher density systems, those problems concerning the effect of the uranium density on the maximum safe enrichment have become relatively more important and will be discussed briefly herein.

BASIC CRITICALITY CONSIDERATIONS OF HOMOGENEOUS AND NON-HOMOGENEOUS SYSTEMS

Implicit in the determination of the minimum critical enrichment is the concept of an infinitely large mass of fissionable material whose infinite medium multiplication factor, k_{00} , is equal to unity, this being a necessary condition for criticality. Although the uranium density plays an important role in determining the criticality of finite systems, the criticality of infinite homogeneous systems does not depend upon the density, since no neutrons may escape therefrom, but it is principally a function of the ratios of the neutron cross sections of the fissionable and other materials which comprise a specific system.

It may be noted that the effect of density on homogeneous systems has been re-evaluated recently,⁵ with the conclusions drawn being in agreement with those predicated by the PCTR method and the basic theory of neutron chain reactions,⁶ which holds that the k_{OO} of such systems is indeed independent of the uranium density and the approximations of neutron diffusion theory itself.

However, since the uranium density is a particularly important factor in determining the criticality of non-homogeneous systems of low enrichment, and since the minimum critical enrichment of such systems is somewhat less than that of similar homogeneous systems, it is necessary from a nuclear safety standpoint to distinguish between nuclearly homogeneous and non-homogeneous materials. For example, recent Hanford experiments,7 which essentially confirm earlier work at the ORNL,⁸ have indicated that the minimum critical enrichment of high density uranium metal rods and light water, in lattice arrangement, is about 0.71% U-235. It may be noted that the minimum critical enrichment of this high density, non-homogeneous system, is thus significantly lower than the value of 1.03% U-235 as determined for homogeneous systems by the PCTR.

BASES OF NUCLEAR HOMOGENEITY

The distinguishing characteristic of a spatially infinite system of nuclearly homogeneous material is the uniformity of the neutron flux throughout the system, whereas the neutron flux in a non-homogeneous system, of identical material composition, will exhibit minima and maxima values according to the regions of fuel and moderator which have been established. A discussion of fuel "lumping" is beyond the scope of this study, however, it is sufficient to note that the principal nuclear safety problem is the degree of non-homogeneity which may be tolerated in a nominally homogeneous system without significantly affecting its over-all criticality. Thus, it may be possible that a system comprised of discrete regions of fuel and moderator could be considered as being nuclearly homogeneous provided (a) the uranium density is sufficiently low that even a latticed configuration of fuel and moderator would not appreciably increase the reactivity or, (b) the dimensional limits of the uranium fuel are sufficiently small and maintained throughout as an intimate mixture of fuel and moderator. In the case of the PCTR determination of the minimum critical enrichment of homogeneous systems, the experimental materials consisted of UO_3 powder at various states of water hydration or absorbed water, with the average particle size of the dry UO_3 powder specified as 60 microns, this corresponding to 2.36 x 10-3 inch.1

CRITICAL ENRICHMENT MINIMA

a. Varying Density

The effect of varying the uranium density on the criticality of non-homogeneous systems may be observed in Figure 1, where the density of the uranium fuel in lattice arrangement with light water moderator is plotted as a function of the estimated minimum critical U-235 enrichment, this corresponding to a $k_{\rm CO}$ of unity. The calculations, which are based on a method outlined by Häfele, see Appendix I, utilize the basic four-factor formulation, consider systems whose fuel dimensions and moderation ratios have been optimized, and are normalized to the experimental determination with natural uranium metal fuel rods in light water.⁷ However, the calculated curve is somewhat lower and is



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thus conservative^{*} with respect to the minimum critical U-235 enrichment of 0.99% derived from less extensive experimental data⁹ for uranium oxide fuel rods in light water where the uranium fuel density was 6.63 g. U/cm.³.

It may be noted from Figure 1 that as the uranium fuel density decreases from a maximum value of 18.9 g. U/cm.^3 necessary for the critical natural uranium system, a corresponding increase in the U-235 enrichment of the fuel is required to maintain the lower density, non-homogeneous systems at criticality. It may also be observed that non-homogeneity does not appear to be a principal cause for nuclear concern at the safe U-235 enrichment of 0.95% providing the uranium density does not exceed the specified 3.2 g. U/cm.³. An infinitely large lattice of such materials would probably be far from critical. On the other hand, if the density of 0.95% enriched uranium is permitted to vary and to approach its maximum value it would be necessary that the dimensional limits of the uranium fuel materials be sufficiently small that the system could be considered nuclearly homogeneous.

b. Maximum Density

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From an examination of the available data, it appears that the dimensional limits which satisfy the condition of nuclear homogeneity for 0.95% U-235 enriched uranium at maximum density of 18.9 g. U/cm.³ will be quite small, as in thin metal chips or turnings, or will be comparatively large as in uranium chunks measuring several inches in diameter. This is indicated in Figure 2, where the minimum critical U-235 enrichment, k $_{\infty}$ of unity, is plotted as a function of the rod diameter of uranium metal fuel rods in light water lattices. As shown in Curve A, which was obtained by guided extrapolations of lattice experiments with uranium fuel-light water, the minimum critical U-235 enrichment of 0.71% is found for a rod diameter of 0.925 inch. Fuel rods which are larger or smaller than this optimum diameter cause the minimum critical enrichment to increase and approach limiting values of 5% and 1.03%, respectively, the 5% value having been established as the minimum critical U-235 enrichment for uranium metal, 10 while the latter value is for homogeneous systems.¹

* The term conservative as used herein is applied to calculations or other estimates where the factors are so chosen that criticality would be predicted for an experimentally subcritical assembly; correspondingly, a critical assembly would be predicted to be supercritical.



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DETERMINATION OF ENRICHMENT MINIMA, MAXIMUM DENSITY

The bases of the U-235 enrichment minima given in Curve A, Figure 2 are discussed below. As mentioned previously, there is strong experimental evidence that the minimum critical enrichment of uranium metal-light water lattices is that of the natural uranium; these data are shown in Figure 3 where the maximum material buckling of the lattice is plotted as a function of the fuel rod diameter. The material buckling, which is another convenient criticality parameter, is negative if the uranium material cannot be made critical, assumes positive values if criticality is possible, and becomes zero at the minimum critical U-235 enrichment. Since a maximum experimental buckling value of zero was obtained for a lattice of 0.925 inch diameter fuel rods, this rod diameter is that plotted in Figure 2 for the minimum critical enrichment value of 0.71% U-235.



Although the enrichment minima for lattices of 0.387 and 0.175 inch rods were not precisely determinable as in the case of the natural uranium lattices, the available experimental data, 11, 12 permit short guided extrapolations to the zero buckling as shown in Figure 4. The maximum material buckling curve for experimental lattices of 0.387 inch rods and U-235 enrichments of 1.03%, 1.15%, 1.3%, and 3.06% may be represented by an equation of the form:

$$y = a + bx + cx^2 \tag{1}$$

where the constants a, b, and c have been evaluated by standard curvefitting techniques and have values of - 0.01904, + 0.01605, and - 0.00237, respectively. The minimum critical enrichment for a uranium metal-light water lattice of 0.387 inch diameter fuel rods was thus determined to be 0.77% U-235.*

With respect to the 0.175 inch diameter fuel rods, only one experimental buckling value is given, and this determination is for a U-235 enrichment of 3.06%.12 Again, however, the available data for lower enrichment lattices were used to obtain an additional interpolated maximum buckling value, which is considered to be conservative and which permits an estimate of the minimum critical enrichment for such lattices. In Figure 5, the maximum material buckling of the experimental lattices at 1.027% U-235 enrichment is plotted for 0.387, 0.60, and 0.75 inch rods. Since the U-235 enrichment of these lattices actually corresponds to that of the minimum critical enrichment of the homogeneous systems, a short extension of the experimental buckling curve to the zero intercept may be made in this instance since both the maximum material buckling and the uranium fuel rod diameter approach zero as a limiting value. However, since the fuel rod diameter cannot actually be zero in a critical system, the buckling values in this interpolated region of the curve will be slightly larger and thus conservative with respect to those buckling values which may be found by direct criticality experiment. An estimated maximum buckling value of 1.85 x 10-3 cm.-2 thus obtained for 0.175 inch rods is plotted in Figure 4, which was used in determining a minimum critical U-235 enrichment of 0.86% for such lattices, using the method outlined previously.

The two remaining minima values plotted for Figure 2 are given for rod diameters of 0.065 inch and 3.5 inch for an enrichment of 0.95% U-235, and are theoretical determinations since essentially no experimental data exist in these regions of the curve. The determination for the 0.065 inch diameter rod was made using a four-factor method; the optimum H_2O/U volume ratio as found by these calculations is about 5, see Appendix II. The calculation for the 3.5 inch diameter rods, 13 although not of immediate interest in this review, is useful in determining the shape of the minimum critical enrichment curve.

* An alternate equation, obtained by the method of least squares on the IBM-7090, gives a slightly higher and thus less conservative U-235 enrichment value:

 $y = + 0.002709 + 0.013330 \ln x - 0.001918 \ln x^2$.



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NUCLEAR HOMOGENEITY OF 0.95% U-235 ENRICHED URANIUM

As noted previously, it appears that the nuclear homogeneity of uranium fuels whose densities exceed a value of 3.2 g. U/cm.³ should be established in order to apply the safe enrichment criterion, and that any dimensional limitation chosen should be such that the corresponding homogeneous and non-homogeneous systems are comparable in nuclear safety. Since the maximum safe k_{00} of 0.95% U-235 enriched homogeneous materials has been determined as 0.97, see Reference 1 and Appendix II,

 k_{00} (non-homogeneous) $\approx k_{00}$ (homogeneous) = 0.97.

For the maximum density system, uranium metal fuel rods in light water lattice, it has also been determined that 0.02 inch diameter fuel rods would satisfy this safe k requirement, and, as may be observed in Figure 2, Curve B, a safe rod diameter of 0.02 inch is well below the smallest critical rod diameter of 0.05 inch for 0.95% U-235 enriched uranium. Curve B reflects the lower limits of the critical enrichment minima, see Appendix II, and is therefore useful as a guide for such nuclear safety assessments.

Although a safe dimensional limit of 0.02 inch was derived for uranium fuel rods, it is also considered applicable to other geometrical shapes, since the effect of geometry on the U-238 resonance escape probability, which is the predominant criticality factor of low enrichment systems, appears to be essentially negligible for uranium lumps of this small magnitude.* Further, since it is somewhat unlikely that the experimental uniformity of the fuel and moderator will be encountered in actual plant conditions and since random non-uniformities should effectively decrease the reactivity of the fuel, this may be considered as an additional factor of conservatism. Thus, for any discrete particle of 0.95% U-235 enriched uranium fuel, a necessary but sufficient condition of nuclear homogeneity is that one dimension be less than or equal to 0.02 inch.

 $D_{\text{(one dimension of uranium fuel)}} \leq 0.02 \text{ inch.}$

CONCLUSIONS

A review of the density effect has indicated that a change in the uranium density does not alter the infinite medium criticality used in the determination of a safe U-235 enrichment criterion for homogeneous uranium materials. However, since the available experimental and theoretical data show the density effect to be so pronounced for low enrichment non-homogeneous uranium materials, a specification for the nuclear homogeneity of 0.95% U-235 enriched uranium, which appears appropriate for nuclear safety considerations, has been derived upon the bases of

* For a detailed discussion of the geometrical effects, see Dresner, Resonance Absorption in Nuclear Reactors, Pergamon Press, Inc., 1960. a dimensional limitation of the individual uranium fuel particle. It is to be emphasized, however, that this specification was derived for hydrogen-moderated uranium systems, and that moderators such as carbon, beryllium, and deuterium are specifically excluded.

ACKNOWLEDGEMENTS

Several helpful discussions concerning the effect of the uranium density on low enrichment systems were held with W. Häfele, of the Kernreacktor, Karlsruhe, Germany, see Appendix I, and with L. Dresner of the Oak Ridge National Laboratory. Acknowledgement is gratefully made herewith.

APPENDIX I

CRITICALITY CALCULATIONS OF LOW-ENRICHMENT URANIUM LATTICES WITH LIGHT WATER MODERATOR

The following calculations were first outlined by W. Häfele, during an ORNL seminar in February 1960, and are intended to show the effect of the uranium fuel density on the minimum critical U-235 enrichment of latticed systems. The notation is principally that of Glasstone and Edlund.¹⁴

The maximum material buckling, B_m^2 , inferred from the experiments with natural uranium light water lattices,⁶ may be expressed as follows:

$$B_m^2 = \frac{k_\infty - 1}{m^2} = + 30^{\circ} \times 10^{-6} \text{ cm.}^{-2}$$
 (I-1)

For hydrogen-moderated systems, a representative value of the migration area, M², is taken as 33 cm.². The infinite medium multiplication factor, k_{00} , is found by the standard four-factor formula, η f p ε ; other nuclear quantities used in the calculations are:

$$N_{o} \approx 0.45 \cdot 10^{23} \cdot \left(\frac{\rho_{o}}{18.9}\right)$$

$$\sigma_{\overline{o}} \approx [650 \ \alpha + (1 - \alpha) \ 2.8] \cdot 10^{-24} = (650 \ \alpha + 2.8) \cdot 10^{-24}$$

$$\Sigma_{o} \approx (29 \ \alpha + 0.126) \left(\frac{\rho_{o}}{18.9}\right) \ \text{cm.}^{-1}$$

$$\Sigma_{1} \approx 0.02; \ V_{1}/V_{o} \approx 1.5; \ \overline{\phi}_{1}/\overline{\phi}_{o} \approx 1.5;$$

where ρ_{0} is the uranium density, and α is the U-235 enrichment. Subscripts 0 and 1 refer to the uranium fuel and water moderator.

The thermal utilization factor, f, is given by the expression:

and

$$\mathbf{f} = \frac{\Sigma_{o}}{\Sigma_{o} + \Sigma_{1} \left[\frac{V_{1}}{V_{o}} \cdot \frac{\overline{\varphi}_{1}}{\overline{\varphi}_{o}} \right]}$$
(I-2)

* Reactor Physicist, on leave from Kernreaktor, Karlsruhe, Germany.

where
$$\begin{bmatrix} V_1 & \overline{\phi}_1 \\ V_0 & \overline{\phi}_0 \end{bmatrix}$$
 is the thermal disadvantage factor.

The fast fission factor, ϵ , may be represented by:

$$\epsilon = 1.00 + 0.07 \left(\frac{\rho_0}{18.9}\right)$$
 (I-3)

and η the average number of fission neutrons emitted per thermal neutron capture in the uranium fuel is expressed by

$$\eta = \frac{2.5}{1 + \frac{\Sigma_0}{\Sigma_f}}$$
(I-4)

where

$$\frac{\Sigma_0}{\Sigma_{\rm fr}} = \frac{100 \ \alpha + 2.8}{350 \ \alpha}$$

It is now possible to test the following empirical expression for the U-238 resonance integral RI, based upon an interpolation between the non-homogeneous latticed systems and the homogeneous systems of identical composition.

$$RI_{(lattice)} = RI_{(homogeneous)} \left[1 - C \left(\frac{\rho_0}{18.9} \right) \right]$$
(I-5)

The U-238 resonance escape probability, p, which is obtained from the four-factor formula and Equations I-1 through I-4, is determined to have a value of 0.770 for the experimental natural uranium light water lattice. Thus, solving the following expression:

$$p \cong \exp\left[-\frac{N_{o} V_{o} \overline{\phi}_{o}}{V_{l} \xi \Sigma_{s} \overline{\phi}_{l}} RI\right] \cong \exp^{-0.26l}$$
(I-6)

it is determined that RI(lattice) has a value of 14 barns.

The U-238 resonance integral for homogeneous systems is simply:

$$RI_{\text{(homogeneous)}} = 3.9 \left[\frac{\Sigma_s}{N_o} \right]^{0.42} = 22 \text{ barns.}$$
(I-7)

Using the resonance integral values from Equations I-6 and I-7 and inserting in Equation I-5, it is determined that the constant C assumes a value of 0.36.

For a lower uranium fuel density of 4 g. $U/cm.^3$, RI (homogeneous) is 37 barns, and from Equation I-5:

$$RI_{(lattice)} = 37 \left[1 - 0.36 \left(\frac{4}{18.9} \right) \right] = 32 \text{ barns.}$$

As a further check on this resonance integral value for the lower density fuel, the theoretical resonance integral expression for latticed systems,

$$RI_{(lattice)} = 9 + 24 \left[\frac{S}{M} \left(\frac{\rho_0}{18.9} \right) \right]$$
(I-8)

where the surface area to mass ratio $\frac{S}{M}$ may be taken as 0.20, is used to determine the U-238 resonance integral of the lower density uranium fuel; close agreement is noted between a theoretical value of 31 barns and that obtained from Equation I-5. Calculations were thus performed to determine the minimum critical enrichment α , which satisfied the condition of the infinite medium multiplication factor of unity, where the uranium fuel densities were 4.0 g. U/cm.³, and 3.2 g. U/cm.³, values of 1.0% and 1.1% U-235 were obtained, respectively. A factor of conservatism was introduced in the calculations by considering the thermal disadvantage factor,

$$\left[\frac{\mathtt{v}_1}{\mathtt{v}_{\circ}}\cdot\frac{\overline{\mathtt{p}}_1}{\overline{\mathtt{p}}_{\circ}}\right]$$

as unity, thus increasing the thermal utilization factor f to a value larger than it actually should have. The predicated U-235 enrichment minima should, therefore, be somewhat lower than those which could probably be obtained by experiment with lattices of low density uranium fuels.

An indication of the degree of conservatism of the predicated results for low density uranium is obtained from the data of Figure I-l which plots the maximum experimental buckling, see References 7, 8, 11 and 12, as a function of the U-235 enrichment both for uranium metal rods and UO_2 rods (7.5 g./cm.³) in light water lattices. The critical enrichment minima inferred from these data are 0.702% U-235 and 0.99% U-235 for uranium densities of 18.9 g. U/cm.³ and 6.6 g. U/cm.³, respectively; these minima may be compared with those of the calculated critical curve shown in Figure 1.



k_{∞} Determinations of 0.95% enriched non-homogeneous systems

The predominant factor in the higher reactivity of low enrichment nonhomogeneous systems is the increased probability that neutrons will escape U-238 resonance capture as they lose energy by the various collision processes. This is due largely to the surface absorption of resonance energy neutrons at the outer layers of the uranium fuel "lumps," which thereby increases the overall probability that neutrons will pass through the fuel without U-238 capture and thus may be available for U-235 fission capture at the lower thermal energies. This effect may be noted in Figure II-1 where the resonance escape probability p is plotted as a function of the rod diameter for lattices of uranium metal rods and light water. The values of p given for 0.387 in. and 0.60 in. rods have been reported previously, 11 however, values of p for the homogeneous mixtures considered as the limiting case for zero rod diameter, were determined from the resonance escape formula,

$$p = \exp\left[-\frac{N_{o}}{\xi \Sigma_{g}} RI\right]$$
(II-1)

It may be noted that Equation I-6 for non-homogeneous systems reduces, as it should, to Equation II-1 since the flux ratio $\overline{\phi}_{0}/\overline{\phi}_{1}$ becomes unity at all energies and the fuel and moderator volumes V_{0} and V_{1} become identical for homogeneous mixtures of fuel and moderator.*

However, offsetting the gain in resonance escape probability is a corresponding decrease in the utilization of thermal neutrons due to the lowering of the thermal flux in the uranium fuel lumps of non-homogeneous systems. This effect may be noted in Figure II-2 where the product ηf , η being the neutron regeneration constant and f the thermal utilization factor, is plotted as a function of the fuel rod diameter for lattices of 0.95% U-235 enriched uranium in light water lattices. As the rod diameter becomes small ηf approaches values obtained for homogeneous mixtures since the thermal disadvantage factor becomes unity and Equation I-2 also reduces to,

$$f = \frac{\Sigma_0}{\Sigma_0 + \Sigma_1}$$
(II-2)

* Values of p were actually determined for 1.027% U-235 enriched uranium; however, they may be used for uranium of 0.95% U-235 enrichment since the slight change in U-235 content will not appreciably affect the total scattering cross section per U-238 atom.



Values of ηf shown in Figure II-2 for 0.387 in. and 0.60 in. diameter rods were obtained by a short extension of data reported for 1.027% U-235 enrichment;¹¹ however, the homogeneous values of ηf were determined from Equation II-2 above, using a value of 1.440 for η .*

A k_{00} determination using the standard four-factor formula is shown in Table II-1 below for 0.05 in. and 0.02 in. diameter uranium metal rods in light water lattices, and for homogeneous system, of light water. Values of the fast fission factor, ϵ , are those reported for 0.25 in. diameter rods, ¹⁵ which introduces a slight factor of conservatism, in these results, since ϵ becomes smaller with decreasing fuel rod dimensions and approaches the lower values which are obtained for homogeneous mixtures of fuel and moderator.

TABLE II-1

INFINITE MEDIUM MULTIPLICATION FACTOR, k_{∞} 0.95% U-235

System	<u>(H/U)</u>	<u></u>	p	e	k
Homogeneous, Light Water	2.8 4.2 5.6 6.0	1.340 1.281 1.228 1.214	0.663 0.723 0.766 0.775	1.0555 1.0436 1.0342 1.0310	0.937 0.966 0.972 0.970
Lattice, 0.02 in. Di- ameter Uranium Rods, Light Water	2.8 4.2 5.6 6.0	1.339 1.280 1.226 1.212	0.673 0.732 0.775 0.785	1.0555 1.0436 1.0342 1.0310	0.951 0.977 0.982 0.980
Lattice, 0.05 in. Di- ameter Uranium Rods, Light Water	2.8 4.2 5.6 6.0	1.337 1.277 1.223 1.208	0.686 0.745 0.788 0.797	1.0555 1.0436 1.0342 1.0310	0.968 0.992 0.996 0.992

Note: 1.4 (H_2O/U) volume ratio = (H/U) atomic ratio.

* Although some variation of η may be possible because of the Wigner-Wilkins hardening of the thermal spectrum with decreasing (H/U) ratios, this effect is small.

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